Understanding Stickiness of Sticky-Cohesive Foods

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To my parents, in loving memory.

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List of accepted conference abstracts

Kazemeini, S.M., Rosenthal, A, J., 2018, Effect of stickiness of solid food products on trigger of food swallowing, Post graduate symposium, University of Nottingham, Sutton Bonington campus, UK. Oral presentation.

Kazemeini, S.M., Rosenthal, A, J., 2019, Effect of stickiness of model food products on trigger of food swallowing, Post graduate symposium, University of Nottingham, Sutton Bonington campus, UK. Poster presentation.

Kazemeini, S.M., Rosenthal, A, J., 2019, Effect of stickiness of solid food products on trigger of food swallowing, 33rd EFFoST International Conference, Postillion Convention Centre, WTC Rotterdam, The Netherlands. Poster presentation.

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List of Publications

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Kazemeini, S. M. & Rosenthal, A. J. 2022. Artifacts and errors in the measurement of the stickiness of liquid foods with tack tests. J Texture Stud. (Published- see Section 7.8), this publication was a study on stickiness with different food samples compared to the current PhD study.

Glossary of abbreviations

Abbreviation	Meaning
FA	Fast Adapting
SA	Slow Adapting
DE	Dextrose Equivalent
GMO	Genetically Modified
G″	Loss modulus
G′	Storage modulus
LVE	Linear Viscoelastic Range
ω	Angular frequency
γ	Shear strain
η*	Complex viscosity
H	Lambda
tanδ	Tan delta or loss factor
rad	Radian
s	Second
Pa	Pascal
kPa	kilo Pascal
mPa	mega Pascal
Hz	Hertz
AHS	Adhesive Hard Sphere
fMRI	functional Magnetic Resonance Imaging
TI	Time Intensity
DTI	Discrete Time Intensity
°C	Degrees centigrade
T1R	Type 1 taste Receptors
EMG	Electromyography
sEMG	surface Electromyography
ESS	Electronic Sensing System
mL	millilitre
l	litter
g	gram
kg	kilogram
min	minute

mm	millimetre
cm	centimetre
m	metre
m^3	Cubic metre
mN	millinewton
N	Newton
TA	Texture Analyser
TPA	Texture Profile Analysis
BMI	Body Mass Index
SSC	Sensory Science Centre
hr(s)	Hour(s)
W	Watt
SNR	Signal to Ratio
DTOS	Double Threshold Onset Segmentation
th	Threshold
BL	Baseline noise extracted from the signal vector
μ_{BL}	Mean of the baseline
σ_{BL}	Standard deviation of the baseline
κ	Predefined factor
W_{crit}	Critical value
W	Window length
t_{on}	Onset time
t_{off}	Offset time
BD	Burst Duration
CD	Cycle Duration
$IChT$	Interchew Time
T	Total time of the mastication
M	The number of total chews
$IEMG$	Chew Work
ChW	Total Chew Work
WR	Chew Work Rate
pW	Proportional Work
NCh	Number of Chews
ACh	Average Duration of Chews
ChR	Chew Rate
pk	Vector of peaks

MA	Maximum Voltage Peak Amplitude
AV	Average Voltage Peak Amplitude
MFS	Median Frequency Shift
MF	Median Frequency
ChT	Chew Time
K	Bulk modulus
P	Pressure
ANOVA	Analysis Of Variance

Thesis aims and objectives

The current research focused on the stickiness of model foods containing sugar and starch. The overall aim of this project was to deepen the understanding of stickiness as a multidimensional textural feature.

The main objectives of this PhD research were as below:

1. Investigate the development of stickiness during chewing and determine its magnitude immediately prior to swallowing.
2. Investigate the most commonly used TA parameters to measure stickiness and validate whether they are influenced by other textural attributes.
3. Measure the degree of stickiness as reflected in the activity of the jaw muscles.
4. undertake correlations between instrumental measurements and physiological parameters to establish instrumental parameters as predictors of perceived stickiness.

The hypotheses associated with the objectives that were tested in this study were:

1. To determine whether the stickiness of model foods decreases to a certain level before swallowing.
2. The necking of sticky foods can affect the validity of parameters determined by the texture analyser during a compression-separation test.

3. Parameters obtained from instrumental measurements will significantly correlate with sensory and/or electromyographic measures.

Thesis outline

Stickiness is a textural characteristic of food that is a difficult area of research from both a human perception and instrumental measurement perspective. For this reason, different approaches were used in the present work to explore the challenges. Since the final stages of oral food processing lead to a sticky-cohesive texture, it was hypothesised that the sticky-cohesive texture would reach a certain level in all model foods of the present work.

The first chapter of this thesis systematically addresses a comprehensive and detailed literature review on topics related to stickiness, such as failure mechanisms related to the different methods used in the present study (instrumental and physiological methods).

Thereafter, various topics were covered in the Materials and Methods section. Efforts to develop six model foods with different degrees of stickiness are presented (section 2.1.1). Sensory measurement (section 2.2) and physiological experiments (section 2.3) are then presented to provide a deeper comprehension of human responses to different levels of stickiness. In the physiological section the influence of the stickiness of model foods on electromyography parameters are measured by recording the activity of mastication muscles (temporalis and masseter). In addition, the texture analyser is used as an instrumental method to measure the surface stickiness of model foods by performing a compression-separation test (section 2.4.1). In order to investigate the internal properties of model foods, a stress relaxation test is also carried out using a rheometer (section 2.4.2). In the present study, the compression-separation test represents the surface stickiness of model foods without catastrophic changes in the sample, while the stress relaxation test provides insight into the rheological behaviour of model foods and their responses to applied deformation. The other instrumental approach is to measure the bulk modulus of the model foods. This measurement was initiated by developing an instrument to measure bulk modulus and establishing a specific protocol to accurately measure bulk modulus values.

Carrying out the above measurements resulted in the collection of a huge amount of data. The different nature of the data (e.g., electromyography data that is not monotonic) was a reason to approach the data analysis with different statistical methods (e.g., Pearson correlation and ANOVA). The results for each specific method are presented and discussed in chapter 3. In addition, possible correlations between sensory, electromyographic and instrumental measurements are also explored. In addition, the results of each method were also correlated with each other to find out whether they correlate with each other or not (sections 3.2.2, 3.2.4, 3.2.5, 3.3.2, 3.3.3).

The final step of the present work is to bring together all the data from the instrumental and sensory analysis by performing a principal component analysis to draw the overall picture of how human perception of stickiness in terms of muscle activity correlates with surface stickiness and rheological properties (section 3.4) to establish if instrumental parameters can predict perceived stickiness.

Abstract

Stickiness is a property of foods that can be both desirable and undesirable. It is described as a necessary characteristic of sticky table rice, while it can be deleterious in food processing, such as the excessive stickiness of bread dough. It is therefore important to understand the underlying principles of stickiness, as a complex textural attribute.

In order to investigate stickiness in this thesis, six model foods were first developed based on different amounts of sugar and heating times. The stickiness of the model foods was measured by two instrumental techniques (texture analyser, rheological test) and by sensory evaluation, while the muscle activity of assessors was measured by electromyography.

Most of the instrumental measurements (except distance to adhesive peak from texture analyser) were able to provide significant predictions of stickiness ($p < 0.05$). Two parameters, force of the adhesive peak and pre-area showed the strongest correlations with other measured values for surface stickiness using texture analyser. In the current study, two new parameters, the pre-area and the initial gradient, were introduced for measuring surface stickiness with the texture analyser.

The results of the rheological parameters using a stress relaxation test showed that the model foods with low stickiness exhibited a solid behaviour in which the modulus of elasticity dominates over the loss modulus, while it was the opposite for liquid-like samples. It was also found that energy dissipation is an important parameter in defining the degree of stickiness, with slower energy dissipation being associated with a higher degree of stickiness.

In addition, sensory evaluation was performed by 10 assessors and using a discrete time intensity method. All assessors rated the overall stickiness of the model foods as significantly different ($p < 0.05$). The sensory evaluation results showed that the model foods with higher stickiness did not undergo maximum stickiness reduction before swallowing. At the same time as the

sensory evaluation, the muscle activity of assessors was also recorded by surface electromyography. Among the features extracted from the electromyography, Chew Work and Chew Time yielded the highest significant correlations ($p < 0.05$) among the model foods. The continuous reduction of the Chew Work values towards the end of oral processing is striking, indicating a structural breakdown of the model foods.

The combination of instrumental and physiological methods was performed by Principal Component Analysis (PCA). By applying PCA to the data, several parameters were grouped together (such as the pre-area and the total-area parameters with the rheological parameters). PCA also provided an overview of positive or negative correlations between the parameters as the overall stickiness positively correlated with most of electromyography features while the correlations were mainly negative with instrumental measurements.

1 Literature Review

1.1 Food texture

Food texture is a concept with many parameters and is highly dependent on the particular food, as each person perceives texture differently (Szczesniak, 1963, Bourne, 2002, Nishinari and Fang, 2018). However, it plays a crucial role in food acceptance (Dar and Light, 2014). Texture is defined by the Oxford Dictionary as “the way food or drink tastes or feels in your mouth, for example, whether it is rough, smooth, light, heavy, etc.”(Oxford, 2019). However, this definition only refers to the oral cavity and does not consider all aspects of texture perception. Szczesniak (1963) discusses several researchers' definitions of texture and concludes that texture is a combination of the physical state of the material (e.g., rheological, and mechanical aspects) and the way it is processed and perceived in the mouth. The importance of human perception in perceiving food texture led to the suggestion of Kilcast (2004) that texture is a property with different aspects (such as appearance, touch and hearing) that together are responsible for food enjoyment. In addition to oral texture perception, it has also been argued that texture is a sensory attribute with different criteria that is first perceived through the sense of touch as a boundary sense and encoded in the brain through the activation of cutaneous pressure receptors (Jowitt, 1974, Bourne, 2002, Szczesniak, 2002, Nishinari, 2004, Stokes et al., 2013). Furthermore, the sense of sight and hearing should also be included (Jowitt, 1974, Bourne, 2002). For example, the visual impact of the Japanese dish *kaiseki ryori* plays a major role in consumers' perception of its texture. If the product is pureed (e.g., to facilitate its use by the elderly), it would not be appealing compared to its original and intact appearance (Nishinari, 2004) which highlights the importance of visual contact in texture perception (Dar and Light, 2014). Overall, addressing the importance of multisensory integration in the perception of food texture and considering multiple parameters when discussing the food texture perception is of great

importance. For this purpose, oral processing, the role of saliva and its lubricating properties, and chewing parameters, such as the number of chews, should be considered (Hutchings and Lillford, 1988).

Defining the individual properties of food texture (either through instrumental or sensory measurements) used to be a major challenge for researchers. It was common practise for researchers to define texture properties for their own research activities, which made it difficult to transfer the results to other products. In a first attempt to increase the comparability of different research results and to promote a universal understanding of texture across different disciplines, Szczesniak (1963) proposed a standard classification of texture attributes (Table 1-1). In this classification, the five most important texture characteristics are hardness, cohesiveness, viscosity, elasticity and adhesiveness.

Table 1-1. Classification of textural attributes (Szczesniak, 1963).

Texture attribute	**Definition**
Hardness	Required force in order to deform the food
Cohesiveness	Strength of the internal bonds making the structure of food product
Viscosity	Resistance of a fluid food to flow
Elasticity	The ability of a food to retrieve its original form after removing a deforming force
Adhesiveness	Required force to separate the food from contacting surface

Although there have been some suggested amendments to Szczesniak (1963), this standard classification has been widely accepted by texture scientists and remains so today. For example, a suggested modification to the above texture classification was made by Sherman (1969). It was proposed to group texture attributes into primary (initial perception such as visual appearance), secondary (initial perception on palate such as adhesion to palate) and tertiary (after application of high shear stress and at the mastication point such as lumpy or pasty). Furthermore, Jowitt (1974) suggested that the classification be modified to use only three general

groups of structure, texture and consistency, where food texture could be defined using adjectives such as rubbery, adhesive, elastic and sticky.

Among these five textural attributes, adhesiveness, or in other words stickiness, is the focus of this project and its definition is discussed in the following section.

1.1.1 Stickiness definition

Stickiness is a multidimensional textural characteristic that has been defined from different angles. One of the first published attempts to define stickiness described it as a tactile sensation when the human skin is pulled away from a sticky material (Zigler, 1923). Other definitions of stickiness by Civille and Szczesniak (1973) and Hoseney and Smewing (1999) also emphasised on the forces required to separate contacting surfaces. In relation to the oral cavity, Jowitt (1974) describes stickiness as “the tendency to adhere to contacting surfaces, especially the palate, teeth and tongue during mastication’’ or in other words, “the sensory experience of mechanical adhesiveness” (Adhikari, 2003). These definitions focus on stickiness as a surface property of food that humans perceive during temporary contact. It is interesting to note that most established instrumental measurements (such as the tack or TPA test), as well as the descriptions used in sensory evaluation of stickiness, are based on the above definitions. Although these definitions provide a general measurement of stickiness, a single physical attribute is not sufficient to draw a complete picture of stickiness. Therefore, it should be emphasised that linking mechanical and sensory measurements of stickiness is of great importance to gain a more comprehensive understanding of stickiness (Peyron et al., 2011). Regardless of the type of food, stickiness can be classified as an undesirable property or an important and essential sensory property of some foods (Kilcast and Roberts, 1998, Russell and Kim, 1999, Fiszman and Damásio, 2000a). For example, while a high level of stickiness is necessary and required for caramel sweets, older people with oral processing disorders may perceive excessive levels of stickiness as an unfavourable property of foods.

Stickiness, as a multidimensional textural attribute is closely associated with other terms such as adhesiveness, cohesiveness, and tackiness. These terms are often used interchangeably and some are favoured over others by different researchers (Jowitt, 1974, Russell and Kim, 1999). For research purposes, it is important to provide clear definitions for each term, and when comparing research studies, it is important to understand how each team understood the terms used in their research. However, the terms most commonly associated with stickiness are briefly defined below:

Adhesiveness is the term most often used in place of stickiness or vice versa. Adhesiveness is defined as the tendency of two materials to stick to each other without a barrier in between (Kilcast and Roberts, 1998). In the case of oral processing, these materials may be a food surface and teeth, while in a compression test it may be a food surface and a stainless steel probe. Adhesion has a similar definition to stickiness and usually refers to surface properties of the food and is therefore a part of stickiness definition. In the literature it can be observed that stickiness is used as a surface property similar to adhesiveness which is not scientifically correct.

Cohesiveness is characterised as the strength of the internal bonds that gives food its structure (Szczesniak, 1963). It has been suggested that cohesion is easily distinguished from adhesion, as it refers to the strength of the internal structure or similar parts of the product to cohere or stick together, while adhesiveness is a surface property of the material that makes the particles stick or cohere together to external surfaces (Adhikari et al., 2001, Nishinari et al., 2019). Some researchers have defined the term cohesiveness differently in the context of adhesiveness/stickiness, e.g., “internal adhesiveness” (Fiszman and Damásio, 2000a) and “intrinsic stickiness” (Carson et al., 2002a). It should be noted that when a sticky food product is torn open, the internal structure becomes a surface property, which is a conversion from cohesion to adhesion. Cohesion is an important parameter for the type of failure, which is discussed in section 1.1.7.

Tackiness is defined as the adhesive failure (see section 1.1.7) of a substance, which is different from the definition of stickiness, as the latter

results mainly from the combination of both adhesive and cohesive forces (Koç et al., 2013, Noren et al., 2019). Tackiness has also been defined by Gay and Leibler (1999) who described it as a property that exists when “a substance appears sticky and some work is required to remove one’s finger from it”. Moreover, tacky texture and perception during oral processing have been defined as the ability of a food (e.g., Turkish Delight) to adhere and cling to the teeth. Tackiness has also been shown to correlate strongly with stickiness (Mayhew et al., 2018). Tackiness seems to be more similar to adhesiveness, while stickiness can be defined not only by one of the three definitions above, but by their combinations.

1.1.2 Theories of adhesion

Various theories have been developed to explain and understand the mechanisms of adhesion and they mainly explain why two surfaces stay together (Nussinovitch, 2017). The theories on the mechanisms of adhesion can be mainly divided into two main groups: mechanical interlocking or entanglement and the charge interactions of materials (Pizzi and Mittal, 2017). In this section, the theories relevant to food and the oral adhesion mechanism are summarised as follows:

Mechanical interlocking is the oldest theory of adhesion, proposed about a century ago (Nussinovitch, 2017). Mechanical interlocking occurs when a viscous adhesive penetrates the surface pores (irregularities) of a solid material (substrate) (Figure 1-1). The mechanism can be initiated or accelerated by a rise in temperature, so that the viscous part penetrates or flows more easily into the pores of the solid surface, and when the temperature subsequently drops, it forms the mechanical interlock (Adhikari et al., 2001, Espinosa, 2011). The softness or deformability of the food, and the smoothness of the surfaces are important parameters for the possibility of mechanical interlocking. The processing of foods with rough surfaces leads to increased adhesion due to mechanical interlocking (Noren et al., 2019). Stickiness can also be a problem when oral hygiene is not

maintained, and hot foods are eaten, and the food debris causes mechanical interlocking by penetrating the irregularities of the teeth.

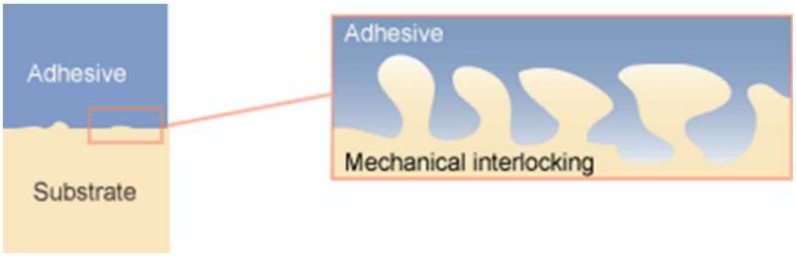

Figure 1-1. Illustration of mechanical interlocking theory (Schaubroeck, 2015).

Wettability theory plays a crucial role in adhesion (Mittal, 1977). When a drop of a liquid material is applied to a solid surface, the tendency with which the drop spreads is called wettability. Normally, higher wettability is a sign of high quality adhesion affinity between the two materials (Michalski et al., 1997). The wettability of a solid and a liquid is assessed by measuring the contact angle of the liquid on the surface of the solid (substrate) (Figure 1-2). Young's equation represents the contact angle between the liquid drops and the substrate as follows (Ismail et al., 2019):

$$\gamma_L \cos\theta_Y = \gamma_S - \gamma_{SL} \qquad (1\text{-}1)$$

θ_Y: Liquid-solid contact angle

γ_L: Liquid-air interfacial tension (surface tensions)

γ_S: Solid interfacial tension (or energy per unit area)

γ_{SL}: Solid-liquid interfacial tension (or energy per unit area)

According to Lee et al. (2015), a contact angle ranging from 0º to 90º signifies high wettability, indicating that no external pressure is necessary for achieving good contact between the liquid and solid surfaces. On the other hand, a contact angle ranging from 90º to 180º indicates low wettability, implying that external pressure is required to establish acceptable contact. In

general, favourable wettability conditions result in enhanced adhesion properties between the liquid and solid surfaces.

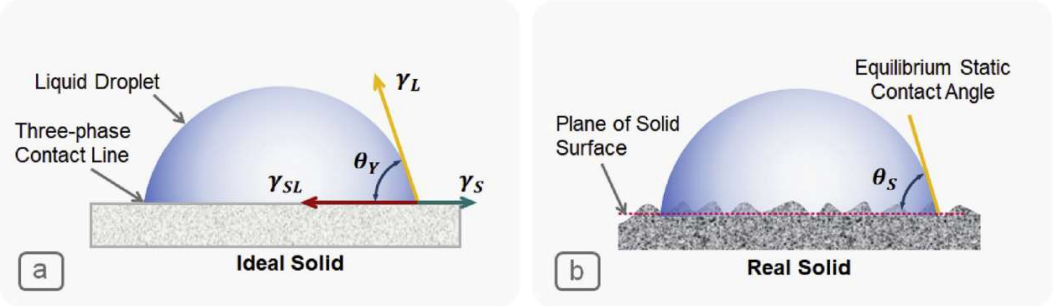

Figure 1-2. Illustration of contact angle based on Young's modulus (Ismail et al., 2019). "a": is an ideal solid where there are no pores or irregularities, while "b" represents a real solid with different levels of pores or irregularities.

If the wettability is acceptable, pores or irregularities on the substrate would generally increase adhesion. If, on the other hand, wettability is low, irregularities would reduce surface adhesion (Mittal, 1977).

Adsorption theory (thermodynamic adsorption); In most of the literature reviewed, adsorption is associated with wettability. However, wettability refers more to the contact angle between the food and the surface, whereas adsorption focuses on the bonds and the nature of the contacts. There are two main types of adsorption: physisorption and chemisorption. Physisorption is mainly controlled by secondary bonding modes such as hydrogen or van der Waals forces. Chemisorption, on the other hand, occurs due to stronger affinities such as covalent bonds (Mittal, 1977, Adhikari et al., 2001, Espinosa, 2011).

Diffusion theory was first presented by Voyutskii and Vakula (1963). This theory is applicable to systems involving two mobile polymeric surfaces and whose mobility force indicates their diffusion or adhesion strength (Mittal, 1977, Espinosa, 2011). When these polymeric materials are in close contact, they begin to diffuse towards each other (Figure 1-3). The adhesion strength is a function of temperature, viscosity, molecular weight, and contact time. Figure 1-3 illustrates the interdiffusion theory (Schaubroeck, 2015).

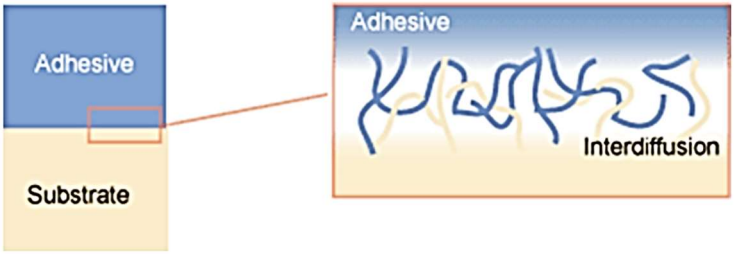

Figure 1-3. Illustration of interdiffusion theory.

In relation to biological surfaces, mucoadhesion plays an important role in diffusion theory. Mucoadhesion is defined as the interaction between a synthetic or natural polymer and a mucin surface (Sau-Hung Spence and Robinson, 1987) and plays an important role in oral drug delivery (Alaei and Omidian, 2021). Mucin forms a biological surface found in many human tissues (e.g., nasal and ocular) (Shaikh et al., 2011). Mucoadhesion is mainly applicable to adhesion theories where there is no solid surface. Currently, there is no single theory that can explain the concept of mucoadhesion. Therefore, it is important to remember that the combination of several theories facilitates the understanding of the mucoadhesive mechanism.

Electrostatic theory occurs when two ionised surfaces come into close contact and an electrical double layer is formed by the transfer of charge between the surfaces. These surfaces can be solid or between food (polymer) and the mucus glycoprotein network on the surface of body tissues (Michalski et al., 1997, Espinosa, 2011).

The importance of adhesion theories is also evident in relation to the adhesion of food to the surface of the mouth, i.e., to the teeth, and the development of caries, leading to cavities and other serious dental problems. For example, beverages such as roasted coffee and green tea have a caries-inhibiting effect on teeth by reducing the adhesion of food to the teeth, which can act as a mediator between Streptococcus mutans and the surfaces of the mouth (Gazzani et al., 2012).

Since foods are complex systems, stickiness can be defined using different adhesion theories. For example, various types of adhesion can occur

simultaneously in the oral cavity, e.g., mechanical interlocking in the presence of moisture. It should also be noted that a comprehensive understanding of stickiness mechanisms through the application of adhesion theories could add value to the food industry, such as the use of mucoadhesive polysaccharides for highly efficient flavour release in the oral cavity (Cook et al., 2017).

1.1.3 Factors affecting stickiness

Food stickiness is a multidimensional textural characteristic that is influenced by several parameters, making it difficult to predict using simple scales. The parameters that influence the perception of stickiness may be related to the intrinsic or extrinsic properties of the food. Some of the most important factors influencing stickiness are explained below.

- **Viscosity** is an important parameter for mechanical interlocking because a change in temperature changes the viscosity of the product which affects mechanical interlocking and finally stickiness. If a food has a high viscosity, more force is required to pull two contacting surfaces apart than if the food product has a low viscosity. This means that the food is classified as stickier (Van Aken et al., 2007). Also, when viscosity increases to the glassy point (transition from rubbery/viscous texture to crystalline/ brittle texture), it can prevent cohesive failure (see section 1.1.3), resulting in a decrease in stickiness (Howes et al., 2003). Viscosity is related to molecular mobility in food. Higher mobility means a decreasing tendency of viscosity which leads to a softening of the material and increases mobility, which in turn leads to a lower stickiness of the system (Adhikari et al., 2001).
- **Water:** the water content of food plays an important role in its stickiness. Depending on the food material, a higher or lower water content can affect stickiness. In the case of hard caramels, for example, a higher water content can lead to higher stickiness (Ergun et al., 2010). Conducting a peel test on caramel samples with different moisture content showed that a moisture content of 11% had a higher stickiness than 10 and 12%

(Wang and Hartel, 2021a). This could indicate that an increase in moisture content does not always lead to higher stickiness, as there seems to be a critical value for moisture content in relation to stickiness. This could also apply to perceived stickiness in oral processing, where excessive lubrication of sticky foods leads to a reduction in stickiness. In addition to bulky foods, for many powdered foods (e.g., icing sugar), "caking" is a phenomenon caused by an undesirable increase in moisture content, which, for example, causes food particles to stick together, reducing their mobility and leading to aggregation, which is closely related to stickiness (Chuy and Labuza, 1994).

- **Temperature:** considering the water content of a food, a rise in temperature can increase the mobility of the water so that it acts as a plasticiser and consequently reduces the surface viscosity. This can lead to a higher possibility of mechanical interlocking, as the sample can more easily penetrate a porous surface. If the temperature is subsequently lowered, this will result in an increased viscosity of the food, which will increase the possibility of mechanical interlocking. An increase in temperature can also alter the failure mechanism (see section 1.1.7). This phenomenon occurs by increasing the viscoelastic compliance of the material, leading to an increase in cohesive failure (internal bonds) compared to adhesive failure (surface interactions). As a result, food materials are perceived as stickier as more residues remain on the surface (Schmidt et al., 2018). As an example, fruit juice powders have a high concentration of low molecular weight sugars, making the product stickier due to the lowering of the glass transition temperature (Jaya and Das, 2009). In general, low glass transition temperature in foods is an important phenomenon affecting stickiness (Adhikari et al., 2001). Foods with a low glass transition temperature can become sticky even at room temperature (Jaya and Das, 2009).
- **Composition of ingredient types:** ingredients such as proteins normally contribute little to stickiness due to their structure as well as their high glass transition temperature (Adhikari et al., 2001). When whey protein isolate was added to honey during spray drying process, an increased

powder yield of 28% to 80% was observed, which was due to the reduction in stickiness. The whey protein isolate does this by forming a glass-like "skin" around the honey droplet. This layer forms as soon as the droplet encounters hot air during spray drying, thus reducing stickiness (Adhikari et al., 2007b). However, the presence of low molecular weight sugars such as sucrose, glucose and fructose increases the stickiness of foods (Jaya and Das, 2009). The Dextrose Equivalent (DE) of corn syrups is associated with food stickiness. Wang and Hartel (2021a) reported that an increase in DE leads to higher values of measured surface stickiness of caramel samples (Adhikari et al., 2001).

The contribution of the various food ingredients to stickiness is shown in Table 1-2.

Table 1-2. Food ingredient's relative contribution to stickiness in food products (Adhikari et al., 2001).

Food ingredient	**Relative contribution to stickiness**
Protein	0
Fats	1
Low molecular sugars	2
Organic acids	2
Water/relative humidity	3

0: lowest contribution, 2: average contribution 3: highest contribution.

By knowing the contribution of each ingredient, it is possible to get some indication of the degree of stickiness of the final product. This knowledge can be used for food design as well as for solving industrial problems with stickiness.

- **Compression:** when a force or compression is applied to a solid food or food powder, the particles are very close together, leading to an increase in stickiness and caking of the particles (Adhikari et al., 2001). In a study by Fiszman and Damásio (2000b), the effects of different degrees of compression on model foods on the instrumental measurement of stickiness were investigated using model gels with different contents of carrageenan, locust bean gum and sugar. They concluded that the higher the degree of compression, the higher the stickiness of a food. The higher

degree of compression also had a plasticising effect on the structure, resulting in increased extensibility of the samples. They suggested that extensibility could also be a parameter for defining stickiness.

The above parameters provided useful insights into the reasons for increased or decreased stickiness. An example can be used to illustrate the relationship between several parameters in the development or control of stickiness. A food product containing water experiences a certain increase in temperature during the process, which might lead to a reduction in viscosity. As a result, the reduced viscosity increases the penetration of the sample onto the contact surfaces (e.g., the conveyor belt). As the sample is transported in bulk, there is a constant pressure on the samples coming into contact with the surface, which in turn increases the rate of penetration into the surface. Therefore, if the process described above is not carefully controlled, serious production problems can occur. From this example, it can be seen that control of the ingredients and the individual process steps is of great importance in order to minimise the negative effects of stickiness.

1.1.4 Instrumental measurement of food texture

Characterising the mechanical properties of food is one of the main reasons for instrumental measurements of food texture (Friedman et al., 1963), taking into account production processes and consumer preferences. Since food production (e.g., pumping through pipes) and chewing by consumers are very dynamic processes and involve a large number of parameters, the result of the instrumental measurement usually has a low correlation with the real situation. One suggestion to obtain more reliable data is to expand the range of instrumental techniques, methods and data analysis used in the experiments to obtain a more comprehensive picture of the nature of the food sample. Another suggestion is to include possible parameters from oral processing and/or technological processes. Indeed, these parameters represent some of the most important limitations of instrumental measurement. These limitations include, but are not limited to, test temperature, test speed, the presence of saliva or a biomimetic mucosal

surface, the manipulation of the food by the tongue in the mouth and the size and shape of the food (Bourne, 2002, Foegeding and Drake, 2007).

Instrumental methods are categorised in different ways based on the concept used in each specific method. The methods used by researchers can assess different parameters, including: empirical (mimetic approaches, e.g., artificial jaw), compression, puncture (penetration), shear (torsion/twisting), cutting, rheology, extrusion methods and tensile techniques (tension/bending) (Szczesniak, 1973, Kilcast, 2004). There are many instrumental techniques for measuring food texture. The most popular instrument is the texture analyser, which is used to examine a variety of food textures (Rosenthal, 2010). The technique was originally introduced by Friedman et al. (1963) and has been shown to provide an acceptable correlation between instrumental and sensory evaluation. The texture analyser can be used to perform many different measurements such as compression (e.g., TPA test), penetration and tensile tests. Among the different methods, the TPA and the simple compression test are the most commonly used by researchers.

1.1.5 Texture Profile Analysis (TPA) and single compression test

One of the most important tests performed with the texture analyser is Texture Profile Analysis (TPA). This is a widely accepted test consisting of two successive compressions to mimic jaw movement (Friedman et al., 1963). An illustration of a TPA test can be found in Figure 1-4. Before performing the TPA experiment, a sample is placed between the probe (moving plate) and the base. The experimental procedure is divided into two sections: first bite and second bite. During the first bite (first compression), the probe compresses the intact sample at a certain compression rate. This is followed by decompression when the probe is lifted. The second bite or compression follows on the deformed sample (Bourne, 2002). The settings used for this test are critical for reproducibility and comparison of research results from other groups. There can be large differences in the measurements when the speed of the moving plates is changed while in contact with the sample (Kazemeini and Rosenthal, 2021b, Kazemeini and Rosenthal, 2022).

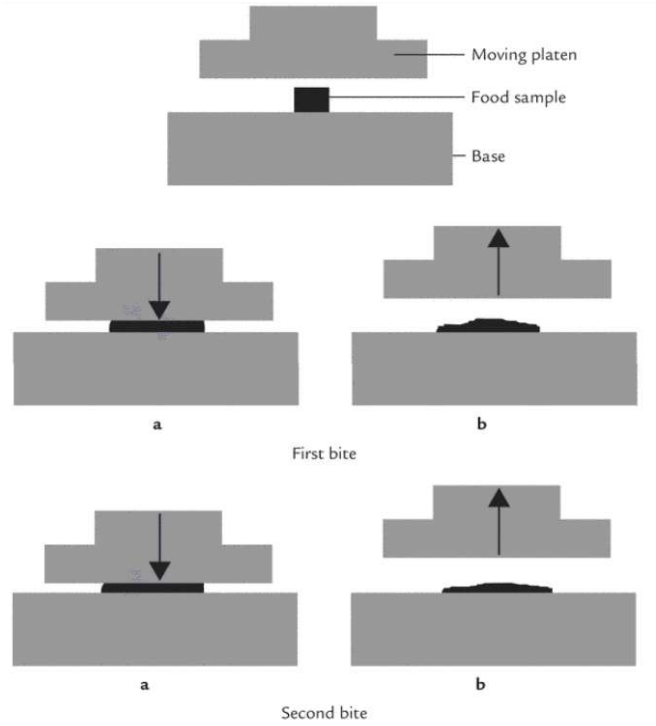

Figure 1-4. Illustration of a typical TPA test (Bourne, 2002).

The TPA method mainly measures hardness, cohesiveness, adhesiveness, elasticity (or springiness) and fracturability (originally called brittleness) but some other derivatives can also be assessed (Bourne, 2002, Rosenthal, 2010). A typical diagram of the result of a TPA test is illustrated in Figure 1-5. It can be seen in Figure 1-5 that fracturability is the main break in the first positive compression curve. The hardness is the peak force of the first compression. The ratio of the positive area under second and first compressions (A_2/A_1) is defined as cohesiveness and the area under the negative curve of the first compression test (A_3) is defined as adhesiveness or stickiness (Bourne, 2002).

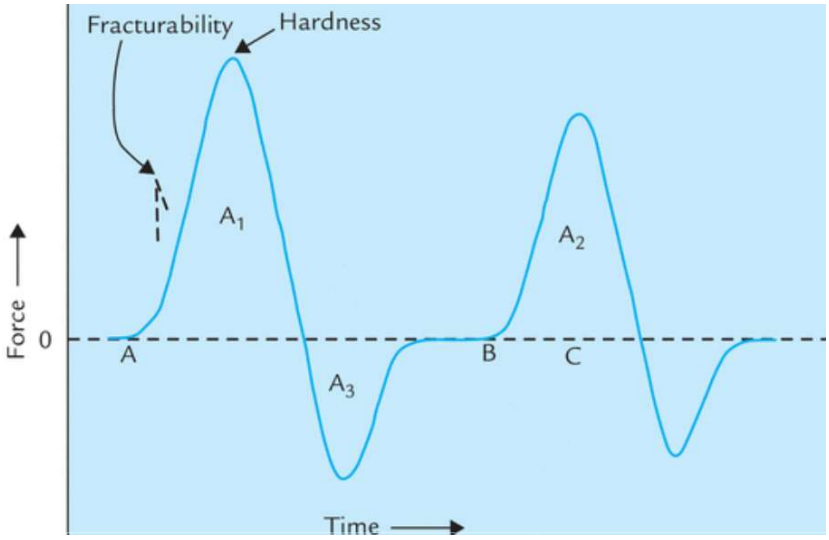

Figure 1-5. Typical graph of a TPA test (Adapted from Bourne, 2002). A: start of compression, A1: area under the first compression (positive force), A2: area under the second compression (positive force), A3: adhesiveness (negative force).

If the second compression test is not needed for data analysis, a single compression test can be performed. For example, if only hardness or adhesiveness are of interest, a single compression test would suffice. In a study by Fiszman and Damásio (2000b) a comparison of TPA and single compression tests to measure the adhesion strength of some food samples was carried out. The negative area (A3 area- Figure 1-5) and the maximum negative force (peak of the A3 area- Figure 1-5) from the first compression were employed as adhesiveness indicators. They concluded that these two parameters are strong indicators of adhesion. Due to the structural damage caused by the first compression, they also suggested that a single compression test with a low compression force provides more reliable data on adhesion. The literature also discusses that the quality of data obtained with TPA has some limitations. The effectiveness of the TPA method for measuring adhesion strength and some other textural properties was reviewed by Nishinari et al. (2019). Here, the TPA method was criticised when applied to different foods with different textural properties and a number of changes were proposed to create a reliable test method and improve the quality of the data obtained. For example, they suggested that artificial saliva should be added to foods with low water content (such as biscuits and bread) to mimic oral processing conditions. In another review,

Peleg (2019) questioned the validity of TPA altogether and suggested replacing it with “true or intensive material properties” (regardless of the size of the material). They proposed that standardised test conditions are not able to reduce the variation found when using different sample sizes. A final limitation associated with the use of TPA was highlighted by Chen and Opara (2013) as the lack of standard operating procedures. An attempt to introduce standard protocols for the use of TPA was proposed by Rosenthal (2010). He suggested being particularly cautious when researchers modify the original TPA protocol and recommended that care should be taken when deformations exceed 75% as this may lead to greater variation between replications. This suggestion could be due to the fact that the higher deformation forces lead to enormous structural damage to the product, which subsequently affects the quality of the data.

Overall, TPA is a widely accepted method that is easy to perform. The data obtained should be treated with caution as they can easily be influenced by various parameters. It should be noted that either TPA or the simple compression test can be used depending on the parameters measured.

1.1.6 Instrumental stickiness test methods

Attempting to instrumentally measure the stickiness of food materials is a complex task and its instrumental characterisation is therefore important. This may be the reason why researchers in the past have looked at instrumental measurement of stickiness from different angles, considering adhesion force, viscosity and viscoelasticity, as well as investigating the interplay between adhesion and cohesion forces (Adhikari et al., 2001).

Instrumental stickiness test methods have been reviewed by Adhikari et al. (2001) and are summarised as shear cell, sticky point temperature, optical probe, tackmeter, peeling, weighing, and glass transition temperature. The methods used should be selected specifically for the foodstuff in question, as each is designed for a particular textural characteristic. Table 1-3 lists various instrumental test methods for characterising the stickiness of foods and associated terms.

Table 1-3. Test methods for measurement of stickiness in relation to different food products.

Test method	Test products	Comment	Reference
Sieve	Stickiness of cooked rice	This method is very easy to use and gives a clear indication of the stickiness of cooked rice by placing sieves with different meshes on top of each other	(Kumar et al., 1976)
Contact angle	Adhesion on food packaging	An indication of work of adhesion	(Adhikari et al., 2007a)
Peel	Adhesion on food packaging	Widely used for dough and adhesive joints	(Adhikari et al., 2001)
Weighing	Adhesion of food to its packaging	It is suggested to use this method as a comparison for the same food in different experiments	(Michalski et al., 1997)
Tackmeter	Cereal doughs, confectionery	Measuring the tensile strength between the probe and adhesive	(Adhikari et al., 2001)
Optical probe	Food powders	Reports the flow pattern of the powder	(Boonyai et al., 2004)
Shear cell	Food powders	Reports the flow pattern of the powder	(Boonyai et al., 2004)
Sticky point temperature	Food powders	A point at which the impeller in the experimental set-up requires the maximum force to drive it	(Boonyai et al., 2004)
Glass transition temperature	Food powders	Reports on the change from rubbery to glassy physical state of food and vice versa	(Boonyai et al., 2004)
Rheology	Mainly for viscoelastic materials	Employing different rheological parameters like storage and loss modulus	(Bourne, 2002)
Compression and penetration	Surface and penetration stickiness measurements	A wide range of accessories can be connected to the texture analyser to measure various texture properties	(Fiszman and Damásio, 2000b)

Back extrusion	Mainly for liquid sample	A simple method consisting of a cylindrical plunger and a concentric annular space	(Osorio and Steffe, 1987)
Texture Profile Analysis	Varied samples	Area of negative peak	(Fiszman and Damásio, 2000b)

An important point to consider about the different test methods for stickiness is that they are designed for different food textures. For example, powdery foods should be measured with specific methods, while viscoelastic samples are measured differently. At the same time, some of the methods are applicable to several food textures, such as TPA and the measurement of glass transition temperature.

Considering the above methods, and especially when performing a compression test (such as single compression test) with a texture analyser to measure the stickiness of sticky foods, one problem with the measurement is that some of the food often sticks to the probe when it is pulled out (the probe is lifted) and does not come off. In this case the measured stickiness would be wrong because the curve on the X-axis does not reach zero (see Figure 1-5, when A3 on the time axis reaches zero). One solution is to do some preliminary tests to make sure that the sample will detach after being pulled. Another suggestion is to glue the sample to the base (e.g., with a cyanoacrylate adhesive) so that it remains attached to the base (Pons and Fiszman, 1996, Fiszman and Damásio, 2000b). Another solution is to optimise the retraction speed to ensure the detachment of the sample from the probe. These issues relate to the types of failure, which are discussed in more detail in the following section.

1.1.7 Failure mechanisms

When the adhesive and adherend (e.g., substrate) are pulled apart, two types of failure can occur: adhesive failure occurs when the specimen detaches after undergoing little or no necking (stretching of the material); cohesive failure occurs after necking of the sample (Figure 1-6) (Kilcast and

Roberts, 1998). Despite the differences between the failure modes, it is assumed that sticky behaviour is observed in both failure modes.

Figure 1-6 illustrates the typical process of cohesive and adhesive failure. Cohesive failure occurs when a sample experiences necking (c1). As a result, some residual material remains on both surfaces when the material is removed. In the case of oral processing, this residue may be on all oral surfaces, such as teeth (may also contribute to tooth packing), tongue, hard palate or lips (c2). Adhesive failure occurs with little or no necking (a1) and therefore manifests as relatively clean detachment (a2) (Kilcast and Roberts, 1998).

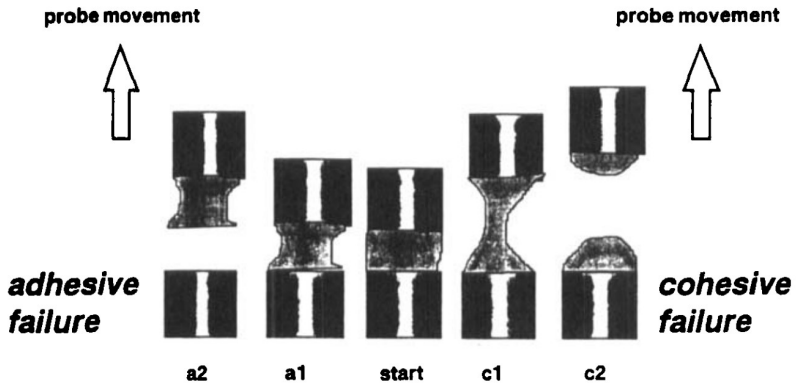

Figure 1-6. Diagram of adhesive and cohesive failure modes (Kilcast and Roberts, 1998)

Cohesive failure occurs after necking happens (c1); here, there is a failure on both sides (c2). Adhesive failure occurs when there is little or no necking (a1); (a2) is demonstrating a clean detachment.

Necking or long behaviour indicates that the surface(s) adhere to the surfaces when pulled from a sample. The necking causes a change in the shape of the sample (Figure 1-7).

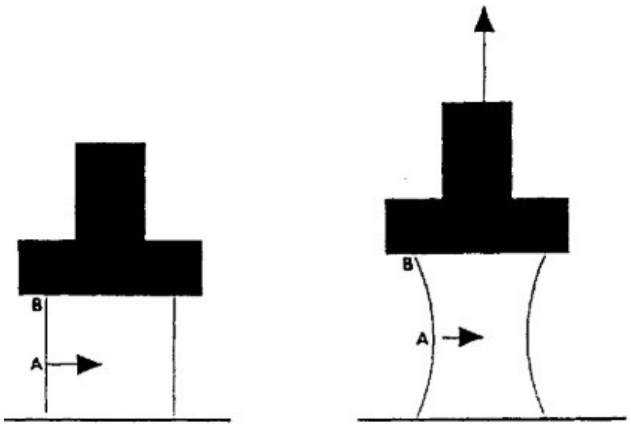

Figure 1-7. Necking of a sticky sample (Hoseney and Smewing, 1999).

As the upper surface pulled away, the sample starts to neck or stretch (in this example, sample was a bread dough).

It is important to note that necking can be seen as a function of rheology rather than being solely due to tackiness as a surface property (Hoseney and Smewing, 1999). This means that what is being measured is a combination of different rheological parameters and that stickiness is one of them. Therefore, experiments to study stickiness need to minimise necking so that more accurate data can be collected. To achieve this, optimising the test speed can significantly minimise necking and thus provide more valid data.

1.1.8 Bulk modulus

Compression of a sample leads to a reduction in intermolecular distances within the microstructure (Figura and Teixeira, 2007). Di Monaco et al. (2008) pointed out that the non-uniform treatment (as opposed to bulk properties) of a group of samples in a sensory situation could be a shortcoming of instrumental measurements. They suggested that uniform pressure on the food sample from all directions would give a more similar replication of the chewing process. The lack of regular instrumental measurements leads to the consideration of the bulk modulus phenomenon.

The bulk modulus is the measure of the change in volume of a sample when it is compressed from all sides without changing its original shape (White and

Mohsenin, 1967). Compression of a food leads to a reduction in the intermolecular distances within its molecular structure (Figura and Teixeira, 2007).

To measure the bulk modulus, the same pressure is applied from all directions and to all sides of the sample (Joyner, 2019) (Figure 1-8).

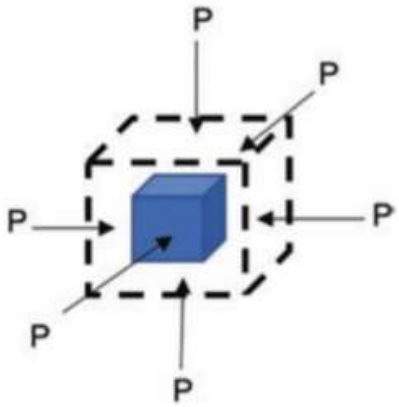

Figure 1-8. Diagram of bulk modulus (Joyner, 2019). Pressure (P) is applied from all directions which results in change in the volume.

The bulk modulus (k) is defined by the pressure applied and the change in volume divided by the original volume.

$$k = \frac{p}{\Delta v / v} \quad (1\text{-}2)$$

P= pressure (Pa)

v= volume (m^3)

Δv= volume change (m^3)

The volume of the sample is defined by the density and the mass of the sample.

$$\rho = \frac{m}{v} \quad (1\text{-}3)$$

ρ=density (kg/m^3)

m= mass (kg)

v= volume (m^3)

Another definition of bulk modulus is given by Figura and Teixeira (2007) who suggested that bulk modulus represents the firmness of a food and is related to the amount of elastic compression the material can undergo by changing the distance between molecules. Another definition of bulk modulus comes from Elfawakhry et al. (2013) who described it as a “general feeling” of the volume of wheat dough under uniform pressure.

The application of the bulk modulus was introduced primarily to measure the mechanical properties of agricultural products in their original form and to enhance the understanding of fruits and vegetables (e.g., apple, peach and potato) throughout their life cycle (harvest and transport to delivery to the customer) (Morrow and Mohsenin, 1966, Finney and Hall, 1967, Sharma and Mohsenin, 1970, Clark and Rao, 1977). At that point, the measurement of bulk modulus was used to study the relationships and responses to the applied pressure and subsequent deformation and structural changes of the product.

There are various methods of measuring bulk modulus, and hydrostatic compression has been suggested as a way of studying the viscoelastic behaviour of certain foods (Morrow and Mohsenin, 1966). The apparatus used to measure bulk compression usually consists of a sample vessel, a pressure supply and a pressure measurement system that uses a hydraulic fluid (White and Mohsenin, 1967, Clark and Rao, 1977, Figura and Teixeira, 2007). When compression causes the molecules of the material to approach each other, the material usually returns to its original volume after the pressure is released. Therefore, the bulk modulus can be described as the resistance of the structure of the material to the pressure exerted (Figura and Teixeira, 2007). It is worth noting that measuring the bulk modulus is challenging. Applying uniform pressure in a chamber is a difficult task, and the measurement is also slow. If air is present in the structure of the food being tested, this limits the application of bulk modulus measurement, as air is a compressible element in an otherwise incompressible system (Bourne, 2002). Due to the limited technical equipment and commercial devices available to measure bulk modulus, very little data is available in the food science literature.

In a study, the mechanical properties of McIntosh apples were investigated by measuring the bulk modulus, from which it was concluded that they exhibited linear viscoelastic behaviour. It was also suggested that most agricultural products (e.g., apples) exhibit time-dependent textural behaviour (Morrow and Mohsenin, 1966) and if a food has an anisotropic bulk modulus, then the changes in the dimensions x, y and z would be different (Figura and Teixeira, 2007). Güzel et al. (2007) used bulk modulus measurement methods to study the mechanism of dehulling peanuts with a shelling machine. They found that the bulk modulus of peanuts was different from that of a single peanut, which was due to the stiffness of the individual peanuts. They also reported that beyond a certain point, the deformation continued to increase, while the force value decreased, and that this represents the behaviour of a viscoelastic material. Clark and Rao (1977) designed a low-pressure bulk apparatus (0 - 3448 kPa) to measure the texture of fresh peaches. They found a positive correlation between the bulk modulus data and sensory ratings of hardness and elasticity. They also pointed out that the results may not be valid with increasing pressure, as the flesh of the peaches is damaged. In another study by Sharma and Mohsenin (1970) a pressure of less than 276 kPa was recommended to avoid possible structural damage to apples. If the food is very sensitive to damage, a low pressure chamber could be used (White and Mohsenin, 1967).

In addition to agricultural products, food gels have also been studied using bulk modulus. In an interesting research study, the bulk modulus of gels made from genetically modified (GMO) potato starch was compared to the gel of a control starch sample. The higher bulk modulus values of the GMO sample suggested that the starch was linked by more intermolecular physical interactions within the structure of the sample. These intermolecular interactions also resulted in gels with higher viscosity, melting point and stiffness of the GMO starch (Kanarskii et al., 2011). The bulk modulus of polysaccharide gels with different oil contents was studied by Ching et al. (2016). They found that the rheological behaviour of the individual microgel particles affected the bulk modulus of the suspension and concluded that increasing the oil content of the microgels lowered the bulk modulus of the

suspension due to the higher deformability of the microgels. In another study, the bulk modulus of wheat dough was characterised and used to determine the torque required for dough processing: Dough with a higher bulk modulus requires higher torque values (Steffe, 1996).

Another device that has been used to measure bulk modulus is an oedometer (Cheng et al., 2015). Normally, an oedometer is used for measuring the consolidation characteristics of soils. However, it has also been used to measure the compressibility of grains (Moya et al., 2002, Moya et al., 2006). Cheng et al. (2015) used an oedometer to measure the bulk modulus of shelled corn at different pressure levels and moisture contents. They concluded that bulk modulus increases with decreasing moisture content of shelled corn and increasing compression ratio. They also proposed an exponential model linking bulk modulus to moisture content and compression pressure.

The bulk modulus seems to have some potential for measuring texture and especially for cohesive/sticky foods. One of the major limitations for the wide application of this method is the complexity of the experimental setup. At the same time, this experimental setup is not commercially available, and its availability depends mainly on research groups developing their own experimental setup. These could be some of the main parameters that lead to the limited use of the bulk modulus by researchers. If such an experimental setup is available, the method should be applied to different food textures, especially cohesive-sticky materials, to verify whether the bulk modulus can provide reliable data or not.

1.1.9 Stress relaxation test

Rheology is a science that studies the behaviour of materials under various stimuli (e.g., applying stress or strain) (Malkin, 1994). Analysis of the rheological properties of food is an approach to measuring the texture of food. In this method, texture attributes (e.g., stickiness) are assumed to be both a surface and a rheological (internal) property of the food (Dobraszczy, 1997).

Among the numerous rheological techniques available, this thesis focuses on stress relaxation tests as a tool to characterise the mechanical behaviour of sticky materials. In a typical stress relaxation test, an instantaneous deformation (or strain) is applied to the sample and the evolution of the stress over time is measured (Norton, 2010). It is known that the relaxation process takes different times for different materials. For common viscoelastic food materials, the spectrum of relaxation shows a wide range of timescales (Mewis and Wagner, 2012, Malkin and Isayev, 2017). If the material is a viscoelastic solid, then partial relaxation occurs. In the case of a viscoelastic fluid, on the other hand, the stress relaxes completely and reaches zero stress state (Mewis and Wagner, 2012).

Prior to conducting a stress relaxation experiment, an amplitude sweep test is performed using the oscillating shear rheometry technique to determine the Linear Viscoelastic Range (LVE) of the material. All subsequent experiments are conducted within this range, where the structural changes within the sample are reversible and consistent with the linear viscoelastic model. It is noted that the use of deformations higher than the LVE is becoming popular among researchers as this could improve the quality of the data obtained and increase the correlation of rheology with oral processing data. It was also highlighted by Bhattacharya (2010) that experiments conducted beyond the LVE are more useful for handling samples (such as dough) as stress relaxation tests provide insights into microstructure as well as the behaviour of food materials.

By measuring the response of the material to the applied oscillatory shear deformation, the solid-like and fluid-like components of the viscoelastic behaviour can be discriminated. Within the LVE range, if the material has a higher loss modulus (G″) than storage modulus (G′), then its behaviour is dominated by viscous response, and if G′ is higher than G″, then material's behaviour is dominated by a solid-like response (Ozturk and Takhar, 2017, Ching et al., 2016). Typically, viscous fluids and free-flowing polymer fluids exhibit a viscosity-dominated response (G″ > G′), polymer melts and entangled polymer solutions, as well as gels and other viscoelastic solids show an elasticity-dominated response (G′ > G″).

Although there are many studies applying the stress-relaxation test in different disciplines, research in the food field is still limited and mainly applied to viscoelastic food materials. Therefore, only related literatures are discussed below.

Dobraszczy (1997) measured the relaxation time for different wheat doughs. At low relaxation times (less than 1 s), no differences were found between doughs with low and high stickiness. They therefore suggested that in production lines where stickiness is a problem, changing the speed of the line (e.g., conveyor belts) may reduce surface stickiness as the doughs do not have enough time to relax, thus largely preventing stickiness. Although this proposal seems to reduce the problem of stickiness in production lines, its practicality should also be considered. Industrial production lines are technically designed systems where any change in the individual steps affects all production processes, and it may not really be feasible to, for example increase the speed of a conveyor belt on which the dough takes its rest time on. It may be more advantageous to consider other parameters in the dough recipe.

Hydrocolloids play an important role in the production of viscoelastic food materials and many of them are known for their adhesive properties (Bosc et al., 2008, Nussinovitch, 2017, Farbo et al., 2020). The rheological properties (storage modulus and loss modulus) and surface adhesiveness (pull off test) of iota-carrageenan blends were investigated. The results showed that the storage modulus was positively correlated with the parameters from the pull off test (maximum adhesion force). However, they showed a negative correlation with the loss modulus, which is indicative of the gel structure. Increasing the content of iota-carrageenan from 1% to 2% resulted in an increase in storage modulus and consequently a decrease in loss modulus. The authors concluded that the 2% mixture was a more structured gel and that the adhesion strength caused by iota-carrageenan gels increased at a 2% concentration, indicating that a more structured gel means a higher adhesion strength (Bosc et al., 2008). Ross et al. (2019) developed and performed sensory and rheological measurements on various hydrocolloid-thickened liquids (xanthan gum, starch and carboxymethyl cellulose gum).

Strong correlations were found between the parameters of stickiness and viscosities at a shear rate of 100 s^{-1}. It has been suggested that shear rates of 90-150 s^{-1} can be used as an indicator of the stickiness of hydrocolloid thickened liquids, with the lower shear rate values corresponding to samples of higher viscosity levels. These results are consistent with those of Stokes et al. (2013), who suggested that viscosity values measured at shear rates of about 50 s^{-1} cannot be used in isolation to predict stickiness and that more complex rheological measurements are required to capture the sensory response. In another similar study by Ong et al. (2018), the highest sensory stickiness values were obtained for the carboxymethylcellulose (CMC) solutions with the lowest shear thinning behaviour, i.e., the most Newtonian-like sample. It was also reported by (Ruis et al., 2007) that the stickiness of caseinate aggregates is strongly pH-dependent, and their stickiness was described by the adhesive-hard-sphere model (AHS). This model was used to explain possible correlations between stickiness, gel properties and pH. Further analysis showed that the increase in stickiness was also related to the gelation of the aggregates.

It is obvious that hydrocolloids strongly influence the viscosity of food and consequently change the degree of stickiness. Researchers generally agree that higher viscosity is associated with stickier foods. This means that stickiness can be manipulated by both viscosity and structural changes.

Other raw materials such as starch and sugar also play an important role in stickiness and alter the rheological characteristic of foods. In a study by Iturriaga et al. (2006), the influence of the soluble and total amylose content of different starches on the degree of stickiness, measured by instrumental analysis, was evaluated. Although the samples with the lowest amounts of these amyloses had the highest stickiness, other parameters such as the interactions between soluble amylose and protein content were found to be important indicators of stickiness, at least when starch-based foods were involved. In another study, the role of amylopectin in the surface stickiness of parboiled rice was investigated. The leaching of amylopectin from the rice was found to be an important parameter for its stickiness. Parboiling the rice

gelatinises the starch, resulting in a reduction in the amount of leached amylopectin and a less sticky texture (Li et al., 2021).

Caramel as a high sugar food was the model food in a study by Wang and Hartel (2021a). They used samples of caramel made with different formulations to study different structural aspects such as rheology and surface stickiness. They performed an amplitude sweep test with a stress range of 100-30,000 Pa and an oscillation frequency of 1 Hz. They found that an increase in structural damage due to the application of external forces correlated with a decrease in viscosity and elasticity. In addition, the intermediate values of the rheological data (G", G') showed a higher peel strength (surface stickiness), which could then be related to adhesion, cohesion, and surface energy within the caramel.

This section has discussed the definition of food texture and in particular stickiness. Due to the complexity of stickiness, various theories have been developed to enhance the understanding of this phenomenon. These theories are linked to parameters that influence stickiness (e.g., compression) and how understanding the parameters involved helps to gain a broader and deeper knowledge of stickiness. One of the most important instrumental measurements of stickiness is the compression test, which is strongly influenced by the type of failure. The type of failure can be reduced by modifying the test method, for example by changing the speed at which the probe is withdrawn. Bulk modulus of food has also been discussed as a possible measurement where the texture of the food is measured in its original form, as opposed to the compression test. In addition, the rheological properties of foods can lead to a deeper understanding of their texture. It was highlighted that the stress relaxation test can define stickiness by how quickly or slowly a sample can relax. Stress relaxation is a suitable method for conducting experiments within the LVE domain to understand the underlying mechanisms of micro-level stickiness. On the contrary, by conducting experiments beyond the LVE range, stress relaxation can also help to predict food stickiness during the production process and subsequently optimise the process.

Overall, stickiness is a multidimensional texture characteristic that can only be determined with different instrumental techniques and careful experimental design.

1.2 Oral processing

Food texture is the key sensory attribute that determines consumer food preferences (Sharma et al., 2022). The texture perception emerges during food oral processing and depends on the mechanisms of food breakdown during chewing. The knowledge of specific mechanisms of food breakdown in the mouth and their links to texture perception is critical for enabling food formulation and structure design with improved consumer acceptability. This is particularly important for reformulation efforts directed at replacing high salt, fat and sugar, as well as improving portion control, and inclusion of valorised and waste stream ingredients in order to ensure a sustainable, resilient and secure supply of food.

1.2.1 Oral texture perception

Influencing food quality and nutritional parameters related to knowledge of the conditions that influence the degradation of food in the mouth is essential for comprehending food texture during chewing (Hutchings and Lillford, 1988). Since the texture is defined as a sensory characteristic of food (Szczesniak, 2002), it is of great importance to gain a deeper understanding of the oral perception of texture.

Besides the mouth's main function of chewing and crushing food as the first step of digestion by enabling the swallowing of food, it has the greatest importance for the consumer's eating experience. The eating experience is the reason for the current increased attention and research on the chewing process. The oral processing of food is closely related to the nature of food. The mouth hosts a dense network of different receptors, and these receptors provide for the perception of food in the mouth (Chen and Engelen, 2012, Haggard and de Boer, 2014). These receptors can be categorised according

to their response to mechanical (mechanoreceptors), thermal (thermoreceptors), and chemical (chemoreceptors) stimuli (Breen et al., 2019). Mechanoreceptors are responsible for the sense of touch and proprioception, i.e. self-awareness of our body position (Kandel et al., 2012). Thermal sensations, such as the temperature of food, are perceived by thermoreceptors. Finally, chemoreceptors provide gustatory perception and chemical stimuli such as the perception of taste and aroma (Engelen and van der Bilt, 2008). Although mechanoreceptors are mainly responsible for perceiving the texture of food in the mouth, their exact mechanism is poorly understood (Breen et al., 2019). They are located in the lips, tongue, palate, and teeth (Trulsson and Johansson, 2002). These receptors have two main functions; the perception of sensory stimulus and transferring the oral food perception data to the brain (Chen and Engelen, 2012). There are fast adapting (FA) and slow adapting (SA) mechanoreceptors and are different in their reaction to various stimuli (Haggard and de Boer, 2014). Slow adapting type I and type II (SA I and SA II) and fast adapting (FA I) receptors, that end in Merkel cells, Ruffini-Pacinian corpuscles, and Meissner corpuscles respectively, are present in the oral cavity. SA I and SA II are responsible for the perception of food texture and shape, and also tongue positioning within the mouth. SA II is specifically linked to the stretching of the skin (Johansson and Flanagan, 2009). FA I reacts to movement and detects the rapid changes in the texture of food during mastication (Chen and Engelen, 2012). It has been suggested that there may be a link between the fungiform papillae and mechanoreceptive perception of the tongue. Essick et al. (2003) categorise some possible reasons for this; it could be that the highest number of fungiform papillae and mechanoreceptors responsible for the tongue mucosa are located in the anterior part of the tongue or the somatosensory afferents of the fungiform papillae probably terminate in a part of the papillae responsible for mechanoreception.

After sensory data has been captured by the receptors, it is transmitted to the brain via the trigeminal network. Figure 1-9 illustrates the pathway by which sensory data is transmitted to the central nervous system via the trigeminal network (Engelen and van der Bilt, 2008).

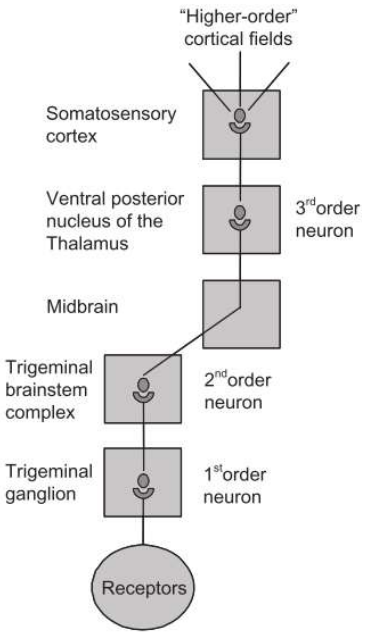

Figure 1-9. Trigeminal network for transmission of sensory data from receptors to the brain (Engelen and van der Bilt, 2008).

The transmission of sensory feedback from the mouth begins with the sensations received by the receptors. The information reaches the trigeminal nerve ganglion, whose main task is to transmit sensory data from different parts of the head (e.g., teeth, gums, tongue, lips) to the brain (Engelen and van der Bilt, 2008, Day et al., 2018). The trigeminal brainstem complex in the brain acts as a sensory processing site (Hu and Woda, 2013). Subsequently, sensory data is transmitted via the midbrain to the third level of neurons. This is the ventral posterior nucleus of the thalamus, which contains specific somatosensory nuclei (Gebhart and Schmidt, 2013). The data is then passed on to the somatosensory cortex, where it becomes the input for the "higher order" cortical fields. The interpretation of these signals by the brain areas leads to the data becoming "perception" (Engelen and van der Bilt, 2008). A possible conclusion from above explanations could be that an increase in the density of the fungiform papillae leads to an intensification of the perception of food. This could be evident in the detection of 6-n-propylthiouracil (a bitter compound) by individuals (Prescott et al., 2004).

Consumers assess the quality of food based on their touch and mouthfeel and both can be considered to be components of the texture percept. Mouthfeel is the sensory response elicited by the activation of mechano-receptors in the mouth, with the leading role played by the receptors located on the tongue (McKenna, 2003, Stokes et al., 2013). The influence of mouthfeel on a product can be illustrated by the phenomenon that foods with a high-fat content are often described more positively and acceptably by adults (Guinard and Mazzucchelli, 1996). Similarly, the rejection of reduced-sugar products (such as cakes or even drinks) where sugar is replaced by an alternative sweetener (e.g., polydextrose, aspartame) is often due to the perceived texture or after feel (change in bulk properties or to some extend viscosity), even if the level of sweetness remains the same (Stokes et al., 2013).

All these parameters suggest that the perception of texture is unique to the individual (Young et al., 2016). It has been suggested that some people have 'better oral acuity', which may be related to their higher ability to discriminate foods based on their texture (Breen et al., 2019). For example, it has been suggested that people over the age of 40 have fewer fungiform papillae on the tongue and a higher lingual tactility response threshold. Although 48 assessors were recruited for this study, extensive testing would need to be done with more assessors to draw solid conclusions. This study also proposed a method called 'bottom-up' to investigate peripheral sensory processes, as opposed to the more conventional 'top-down' method, which focuses more on the properties of foods than on the underlying mechanisms in the oral cavity (Bangcuyo and Simons, 2017).

1.2.2 Bolus formation and swallowing mechanism

The mastication process (generally 10 to 40 chewing cycles) transforms the food into a ready-to-swallow mass called a bolus (Woda et al., 2006b, Chen, 2009). The characteristics of a bolus have been defined using measurement techniques such as rheology (e.g., viscosity), texture profile analysis, hydration, and particle size (Chen and Lolivret, 2011, Peyron et al., 2011,

Panouillé et al., 2014, Young et al., 2016). For example, the viscosity of the bolus is an important parameter that determines the time of swallowing as a higher bolus viscosity leads to a delay in swallowing time (Dantas et al., 1990). A bolus is formed by a series of dynamic structural changes in the food that begin with the initial chewing and continue as saliva mixes with the food until a bolus of the correct size, shape, and texture is formed. This is then delivered to the back of the mouth for swallowing (Bourne, 2002, Chen, 2009, Koç et al., 2013). In a research study by Aguayo-Mendoza et al. (2019), the rheological properties, particularly viscosity, of different foods with liquid/drinkable (e.g., water) and semi-solid/spoonable (e.g., pudding) textures were highlighted as key parameters for oral food processing. Parameters mentioned for oral processing included duration of consumption, speed of consumption, and size of bites. It was also pointed out that the rheological properties of food have a greater influence on oral processing than parameters such as taste and frequency of consumption. That was a reason to suggest adhesiveness and cohesiveness as key factors in bolus formation (Koç et al., 2013). Evidence for this hypothesis is the increased values for these parameters measured at the end of chewing (Shiozawa et al., 2003, Young et al., 2013, Young et al., 2016, Gao et al., 2017). It has been suggested that the perception of the stickiness of the bolus is a possible trigger for swallowing. However, these experiments were mainly conducted with different types of breakfast cereals where the initial low water content limits the validity of this conclusion to other food products (Adhikari et al., 2001, Loret et al., 2011, Peyron et al., 2011). Stickiness was also perceived as dominant in various bread products at the end of the chewing process (Panouillé et al., 2014). The dominance of the sticky sensation at the end of chewing is related to the amount of saliva ingested with a positive correlation with continuous chewing, adhesion and cohesion forces above a certain level, and reduction of yield stress and peak force below a certain level (Loret et al., 2011, Devezeaux de Lavergne et al., 2017, Lorieau et al., 2018). In contrast, stickiness of emulsion-filled gels with high elongation was found to be dominant during oral processing. This was thought to be due to the higher gelatin content of the gels (Devezeaux de Lavergne et al., 2015b). It should also be considered that physiological conditions (e.g., dentition)

might be related to the differences in bolus characteristics (Hutchings et al., 2014). It was also hypothesised by James et al. (2011) that the initial texture of the samples (fracture stress) was related to the degree of stickiness upon swallowing. The study of biscuit samples with different sugar-to-fat ratios of the initial recipe showed that there was a positive correlation between adhesiveness on swallowing and sugar-to-fat ratio. However, the stickiness of the bread samples was found to be related to chewing and bolus manipulation rather than initial texture properties (Jourdren et al., 2016). In another study, Young et al. (2016) measured physical parameters such as yield stress, peak force, adhesion force, and cohesion force of boli from different biscuit formulations. These parameters were examined at the beginning and middle of chewing and at the time of swallowing. No consensus was found for the whole group and the results were different for each assessor. Therefore, it was suspected that individuals have different and unique chewing and swallowing processes, which called into question the existence of a generally applicable swallowing threshold based on individual parameters compared to a group of parameters.

In summary, bolus formation is influenced by various structural characteristics such as adhesiveness and cohesiveness, as well as by people's physiological conditions. It should also be kept in mind that perceived bolus adhesion is a result of both the initial moisture content of the food and oral processing parameters such as salivary secretion.

1.2.3 Stickiness perception

As noted in section 1.1, the perception of the texture of food is a process involving several parameters. Stickiness appears to be one of the most complex of these texture features, and the isolation of stickiness from the overall perception of texture is not well studied in the literature. Expectations of the sensory stickiness of different foods are influenced by geographical differences and cultural values. For example, sticky table rice is disliked by Indian, Bangladeshi, and Malaysian consumers, while it is popular in Japan and Korea (Kumar et al., 1976). However, there are certain health conditions,

e.g., dysphagia, where people have a serious problem with sticky foods because they cannot swallow them safely (Aslam and Vaezi, 2013).

The perception of stickiness has been linked to food viscosity, as viscous and thick foods are perceived as stickier compared to thin and liquid foods (Nussinovitch, 2017). It is therefore suspected that other textural properties may also lead to an increased or decreased perception of stickiness. For example, the ability of emulsions containing maltodextrin to coat the mouth can be perceived as creaminess or thickness, and both of these properties have been linked to the perception of stickiness. Perhaps in this study with maltodextrin, parameters such as food interactions in the mouth and the perceptions caused by them, as well as the presence of saliva, should be added as further non-rheological parameters of stickiness perception (Akhtar et al., 2006).

We know that chewing a sticky product requires more activity of the jaw from the point of view of muscle activity. The receptors in the jaw can be seen as a way of assessing the perception of stickiness. However, current knowledge of oral processing does not identify specific receptors for individual texture perceptions. The paper by Foegeding et al. (2015) discusses some possible reasons for the insufficient understanding of texture perception in the mouth. For example, each mechanoreceptor only picks up a basic part of the stimulus. However, understanding how the arrangement or network of mechanoreceptors subsequently encodes perception requires advanced modelling and computational techniques.

The tactile sense of stickiness was the subject of a research study by Yeon et al. (2017), in which the neural activity of tactile perception of stickiness was investigated using fMRI. Although the participants in the experiment only touched the sticky materials with their index fingers, the results showed the activation of several brain regions. These active regions were the contralateral basal ganglia region, the ipsilateral basal ganglia region, the insula, and the superior and middle temporal cortices. These results suggest that stickiness is perceived through activation of both the somatosensory cortex and other cognitive processes (such as ipsilateral dorsolateral

prefrontal cortex (DLPFC)). At the same time, activation of subcortical regions (including pallidum, putamen, caudate, and thalamus) is responsible for the perception of different degrees of stickiness. A follow-up study by the same researchers (Kim et al., 2017) found that patterns of neural activity can correlate with groupings of tactile perception. This means that as the strength of the sticky stimuli increases, higher neural activities are involved, and also different neural activity patterns are associated with different intensities of tactile stickiness. The results of the above work establish a link between the intensity of stickiness and how the human brain is activated in response to that intensity. Since these studies used fMRI, which was limited to tactile stickiness, further research should be done to investigate how brain activates during oral processing of sticky materials.

Looking at stickiness from a different perspective and in the field of virtual reality, participants perceived hard-appearing objects as significantly less sticky than objects that appeared to be soft, based on their visual feedback. In this study, the perception of stickiness was elicited by electrical stimulation of fingertip mechanoreceptors through multisensory integration (Yem et al., 2018). These results of oral processing of sticky materials are consistent with previous discussions that the softer foods are perceived as stickier compared to hard and elastic materials.

1.2.4 Sensory evaluation techniques

Empirical sensory evaluation of food is an innate trait of human nature. Consumers prefer or reject foods based on their perception without understanding the underlying processes or mechanisms. Scientific experimentation began in the mid-20th century with the US government's efforts to provide suitable food for its soldiers (Meilgaard, 2016). Since then, sensory evaluation techniques and methods have continued to evolve.

Sensory testing is divided into three main methods: discriminative, descriptive, and hedonic testing (Lawless and Heymann, 2010). Discriminative testing methods are used to determine if there are differences between samples. If the differences are statistically confirmed, descriptive

methods are used to understand the reason for the difference or intensity of the parameters. Finally, the hedonic test refers to the acceptability of the samples and how much the products are liked by the consumers (Kemp et al., 2009).

The Time Intensity Technique (TI) is a descriptive sensory assessment method developed over decades. TI is used to continuously monitor short-term changes in specific sensory attributes over time, from the onset of chewing or perception to swallowing or fading (Cliff and Heymann, 1993, Meilgaard, 2016). TI was originally used to study the function of the flavour release (Dijksterhuis and Piggott, 2000). It is known that oral food perception is often related to changes in the bolus during chewing, which limits sensory perception to the temporal aspects. Therefore, due to the chewing process, methods that use a dynamic rather than a static process may provide more realistic data (Dijksterhuis and Piggott, 2000, Meilgaard, 2016, Hort et al., 2017). Time-intensity techniques are more beneficial when assessing products with sensory characteristics that change over time and also products that have long mastication or eating time (Kemp et al., 2009). The wide range of TI applications was discussed by Hort et al. (2017). They emphasised that TI can be used in most food products and gave examples such as beverages, meat products, chocolate products, and olive oil. The TI method includes both continuous and temporal point measurements (Meilgaard, 2016). Continuous Time Intensity (CTI) is widely used among researchers. CTI has traditionally been used to measure one attribute at a time, but other methods have been developed to measure more than one attribute. This technique can be used to conduct time-consuming studies (e.g., how a cream lotion works) or to make measurements that require more time (e.g., assessing texture).The time for long and short techniques can vary from hours to a few minutes (Meilgaard, 2016). Compared to the traditional sensory method, CTI can provide a dynamic understanding of attributes, whereas most sensory methods only provide an overview or average sensory perception (Hort et al., 2017). From a general perspective, CTI is most useful when it comes to specific attributes.

Besides CTI, the Discrete Time Intensity (DTI) technique also belongs to the category of fixed time. In the DTI technique, data are recorded at predetermined time points (e.g., every 5 seconds) during sensory evaluation and assessors evaluate the same sensory attribute(s) over time (Kemp et al., 2009, Hort et al., 2017). Although DTI can be a time-consuming technique, it provides detailed data on the sensory attributes under investigation. DTI has a wide range of applications, from food and beverages to pharmaceuticals and household products (Hort et al., 2017). In a study by Sudre et al. (2012), various wheat flakes were evaluated using both a standard general procedure and a DTI method consisting of four predetermined time points to assess the taste of the samples during chewing. The results were compared and both methods provided similar conclusions regarding the differences between the products. For the DTI method, it was also found that the differences in preference between the samples decreased as the chewing progressed. This result highlights that the specific choice of evaluation methods is based on careful consideration of the research questions. It should be noted that the use of fixed time points compared to continuous measurement simplifies the assessment procedure by alleviating the degree of training and cognitive load (Galmarini et al., 2016). This means that the use of less trained assessors might be more practical when using DTI compared to the CTI method.

Among many aspects of chewing, the side of the mouth most used to manipulate the food is of great importance. This can be used by researchers to select a side of chewing (even a standardised protocol) or even the habitual chewing of subjects. Various methods such as EMG can be used to determine the preferred chewing side of subjects. The simplest method to determine the preferred side of chewing is to select the side of the mouth (right or left) where the first chewing occurs. However, it has been further explored that this may not be the true preferred side as the texture of the food during chewing may influence this. For this reason, EMG can be used to study the whole chewing process and eventually determine the preferred side (Varela et al., 2003). Although habitual chewing does not mean that individuals have a preferred chewing side, the texture of the food can

influence the preferred chewing side and makes it very difficult to determine (Paphangkorakit et al., 2006). Literature reports that influencing the chewing habits of individuals affects food intake. Standardised chewing is mainly used to study how the food intake or energy intake of individuals is influenced (Zandian et al., 2009, Li et al., 2011, Higgs and Jones, 2013). It is important to acknowledge that standardized eating procedures can influence the performance of assessors and impose restrictions on their natural eating behaviour. While the use of standardized eating protocols can facilitate comparisons between different assessors, it also leads to deviations from their typical eating behaviour, consequently impacting how they manipulate food during oral processing. Alternatively, standardisation can be achieved through computation, e.g., by using normalised time. If a study aims to measure habitual eating behaviour, it is important that chewing is not restricted. Therefore, it is important to choose one of the two methods, standardised or non-standardised chewing, depending on the aim of the study.

In summary, the selection of sensory methods should be based on the objectives of the sensory evaluation. Among the many methods, TI and especially DTI is a widely used technique for evaluating foods whose texture changes during oral processing. Although the time intensity method is time-consuming and costly compared to other sensory methods, it can at the same time provide a large amount of detailed data about each assessor and their perception of the attribute in question.

1.2.5 Parameters affecting sensory evaluation

When conducting sensory evaluation, there are psychological and physiological factors that may influence the assessors' perception of the samples. These parameters are discussed below:

1.2.5.1 Psychological factors

Expectation error: Assessors usually find what they expect to find. If assessors know the sensory samples they are to evaluate, they can anticipate their reactions before the samples are presented to them. To avoid

the expectation error, it is very important not to reveal any information about the test or the samples before data collection. In addition, coding the samples and presenting them randomly has been shown to reduce the influence of the expectation error (Kemp et al., 2009, Meilgaard, 2016).

The error of habituation: If assessors are presented with samples of similar characteristics every day, they may give the same answers every day and overlook the actual variance in characteristics, e.g., in the daily quality checks. To avoid the error of habituation, samples with very different characteristics should be presented from time to time (Kemp et al., 2009, Meilgaard, 2016).

Stimulus error: This error is important when assessors use other information or criteria to evaluate and score samples. An obvious example is the differences in colour between samples, where assessors tend to rate the samples with deeper colour as tastier (Kemp et al., 2009, Meilgaard, 2016).

Halo effect: When assessors (mainly untrained consumers) are asked to rate more than one attribute in a sample, they tend to give similar ratings for all rated attributes. Possible ways to reduce the halo effect are to reduce the number of parameters rated or to use trained assessors (Kemp et al., 2009, Meilgaard, 2016). A sweeter product might be more attractive to assessors than a product that has a similar texture but is less sweet. As a result, sweetness may influence their judgement of a food product, even if this factor is not the attribute, they are interested in. This is known as the dumping effect, where a strong attribute of the food that is not intended to be the target of the study influences assessors' perceptions and rankings (Lawless and Heymann, 2010). It has been suggested that the mouthwash of Gymnema sylvestre is able to block the perception of sweetness for a short period of time, suggesting that it can be used for sensory evaluation purposes (see Appendix 6.1 for more information).

In addition to the above parameters, there are other psychological factors that can influence sensory evaluation, such as the motivation of the assessor and the effect of external distractions. These factors must also be taken into

account when planning and interpreting the results of sensory evaluations (Kemp et al., 2009).

1.2.5.2 Physiological Factors

Adaptation: This error occurs when assessors evaluate certain characteristics of samples over a long period of time. This can happen in a sensory evaluation when a large number of samples are presented. In this scenario, the assessors' perceptions adapt to the attribute, and they give ratings based on how much they have adapted to the samples rather than their actual characteristics. To reduce the occurrence of adaptation errors, it is recommended to limit the number of samples, to provide sufficient rest time between the presentation of the different samples, and to provide sufficient number and duration of breaks during sensory evaluation (Kemp et al., 2009). In the study by González et al. (2001a), adaptation error was only found in untrained assessors and not in trained assessors.

Perceptual interactions between stimuli: The presence of certain features in samples may cause one of the following interactions: Enhancement that increases the perceived intensity, which is the opposite of the suppression interaction. It is also possible that a synergistic interaction occurs, where the perceived intensity in a mixture is higher than that of the individual substances (Kemp et al., 2009, Meilgaard, 2016).

1.2.6 Controlling the sensory environment

Location of sensory room: It is very important that the room in which the sensory evaluation is carried out is not near any facilities that might interfere with the senses, such as a smokehouse in a meat processing plant. It is also important that participants enter and leave the sensory centre without seeing anything that could affect their sensory evaluation, such as a place or a sign with information (Lawless and Heymann, 2010).

Control of colour and lighting: Colour is a physical and psychological characteristic of food. It is generally accepted that differences in the appearance of samples in a sensory experiment can influence the judgement

of assessors (Meilgaard, 2016) and affect the perception of texture, taste, and flavour (Calvo et al., 2001, Engelen and van der Bilt, 2008). In order to reduce the perceptual biases due to visual colour variations, various methods are used by researchers; Meilgaard (2016) suggested using coloured serving containers or coloured illumination (red), and Morrot et al Morrot et al. (2001) recommended adding colourants to food, to name a few examples. However, strong levels of coloured lights can affect the assessor's perception of the food. This should be taken into account when planning a sensory evaluation and interpreting the results (Meilgaard, 2016).

Control of the environment: The test environment should be controlled and kept constant as much as possible. For example, the room temperature must be monitored and kept at about 22-23°C throughout the test. Other parameters such as noise, separation of test subjects, and accessibility of the test centre should also be considered (Lawless and Heymann, 2010).

In summary, it is not easy to conduct a sensory evaluation by simply presenting samples to assessors and recording their responses. There are a variety of factors and parameters (e.g., physiological factors) that influence the subjects' perceptions, so the quality of the data obtained could become questionable. Therefore, when designing the sensory evaluation, the parameters that influence the assessors' judgement should be carefully considered.

1.2.7 Electromyography (EMG) and food texture studies

1.2.7.1 Introduction to EMG

Surface electromyography (sEMG or EMG) is a non-invasive method that plays an important role in the study of human muscle activity and has been used since the 18th century. EMG is considered the "gold standard" for measuring muscle movements during mastication. The EMG technique involves recording the electrical responses of active muscles during a specific physical activity. Chewing is a complex and dynamic process that takes place in the mouth and involves the simultaneous activities of the

masticatory muscles and the tongue, which are hidden from our view. Therefore, the EMG technique provides important in vivo measurements to further understanding in this challenging area of research (Brown et al., 1998a, Merletti and Farina, 2008, Chen and Engelen, 2012, Kemsley and Defernez, 2012, Kazamel and Warren, 2017). EMG has various applications in clinical practice (Katirji, 2007), including, dentistry (Nishi et al., 2016), neurology (Zwarts et al., 2005), and food texture evaluation (Boyar and Kilcast, 1986, Jack et al., 1993, Le Reverend et al., 2016). The nature of the mastication process and the continuous manipulation of food texture during this time make the EMG technique a powerful method for food texture analysis.

1.2.7.2 Muscles of mastication (Muscles motor and their activation)

Several muscles are active during the chewing process. However, the masticatory muscles normally used for EMG studies are the left and right masseter muscles and the temporalis muscles, which are located on either side of the face (González et al., 2001b). These two muscle groups are essential for lifting and closing the jaw (Jack et al., 1993). The masseter muscle is considered the most important masticatory muscle (Boyar and Kilcast, 1986). It is a large muscle that has three layers: a superficial layer, a deep layer, and a middle layer, with the superficial layer being used in EMG recordings as it is easy to identify (Chen and Engelen, 2012, Stepp, 2012). The temporalis muscle is another superficial muscle commonly used in EMG studies as it is responsible for lifting and retracting the mandible (Hanasono et al., 2001, Chen and Engelen, 2012, Foegeding et al., 2015). The temporalis is a wide and large muscle located on both sides of the skull. As the access to the temporalis is relatively easy, it is often used together with the masseter muscle to study mastication (Wang et al., 2018). Figure 1-10 illustrates the location of the masseter and temporalis muscles (Chen and Engelen, 2012).

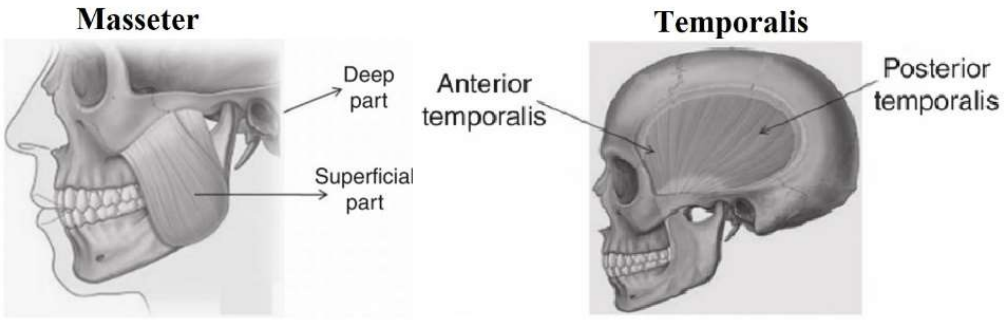

Figure 1-10. Muscles of mastication (left; masseter, right; temporalis) (Chen and Engelen, 2012).

It is not practical to measure the deep part of the masseter with surface EMG. To measure the activity of the posterior temporal muscle, the hair on the side of the subject's head must be shaved. This is often not desired by the assessors and must therefore be taken into account in the planning.

1.2.8 EMG applications in food texture evaluation

EMG measurement of muscles during mastication is a promising method to combine sensory assessment with physiological data (Jack et al., 1993). A major advantage of EMG for the study of food texture compared to other methods is that it does not interfere with the normal chewing process (Mioche and Martin, 1998, González et al., 2001b), while one of the limitations of EMG can be the interference of noise in the recorded data which can affect the quality of the data obtained. This noise can be managed by using a reference electrode to record the baseline activity and subsequently use it to eliminate the noise from the muscle activity data.

1.2.8.1 EMG parameters

There are a number of different parameters that can be extracted from EMG data. The purpose of the study influences which EMG parameters should be extracted and used. Parameters such as total sequence duration, main chewing sequence time, number of chews, clearance time, and masticatory frequency can be used (Chen and Engelen, 2012). Table 1-4 provides a summary of the most commonly used EMG parameters in food chewing

research. It is important to note that each parameter may have different names used by researchers; some examples can be found in Table 1-4.

It is also common to use the ratio of different parameters, depending on the research objectives. For example, Kohyama et al. (2005) used the ratio of muscle activity and the ratio of amplitude in their study. Numerous studies that have been published in the literature either combine different parameters or alter common ones. For example, the sum of muscle activities (Sakamoto et al., 1989, González et al., 2002, Peyron et al., 2002, Kohyama et al., 2007a) and the segmentation of the mastication process (e.g., the total chewing sequence, the first chew, the last 5 chews) (Shiozawa et al., 1999, Kohyama et al., 2008, Kohyama et al., 2016c).

Table 1-4. EMG parameters for studying the mastication of food products.

EMG parameter	Explanation	Publication
Mean chewing frequency (chew rate)	Total chewing cycles over a specific time duration divided by the time (in this study 2 minutes)	(Plesh et al., 1986)
Cycle-by-cycle duration	Parameters such as the mean durations of opening, closing, and occlusal phases, the total duration of a chewing cycle, and the maximal opening distances	
Total number of chewing cycles	The total number of chewing cycles across the whole chewing sequence	(Sakamoto et al., 1989)
Chewing energy (other names: chew work, muscle work)	The area under each burst of chewing as measured by the EMG, compared to the same parameter that was measured whilst chewing the control sample	
Total chewing energies (total chew work)	Sum of all chewing energies of the single bursts	
Number of swallows	Analysis of mastication sequence based on the swallowing times	(Brown, 1994)
Chew work rate	Calculation of chew work/chew time for single chewing cycles or the whole mastication event	(Brown et al., 1998a)
Sum of a specific group of chew works	Calculation of sum of each 5-chew works, e.g., 1-5, 6-10	(Brown et al., 1998b)
Maximum (peak to peak amplitude) and mean voltage	Calculation of EMG parameters for a single burst and total bursts of chewing Maximum voltage indicates the highest peak.	(Mioche et al., 1999)
The interval between single cycles	Measurement of the time between the activity of masseter (discharge) cycles	(Shiozawa et al., 1999)
The amplitude of EMG activity	Measurement of the height of integrated EMG data for individual cycles	
Fourier transform data	Calculation of frequency domain data by doing Fourier transform, as a novel method of EMG data analysis	(kemsley et al., 2002)
Clearance duration	Indicated as the time between the end of the last rhythmical chew to the rest position point of mastication muscles	(Kohyama et al., 2002)

EMG studies can be divided into three categories of questions. The first area is about clinical professionals speculating on how dental treatments affect chewing and masticatory habits. The second area is concerned with how food affects the chewing pattern, while the third group is more interested in the relationships between sensory feedback and the evaluation of food texture (Peyron et al., 2002). Therefore, based on the second and third question groups the EMG applications in the food-related studies are discussed below:

1.2.8.2 The effect of food products on EMG parameters

Muscle activity and consequently EMG characteristics can provide information about the textural properties of food. Some researchers have even attempted to categorise foods based on EMG characteristics rather than sensory evaluation. In an early EMG study, Sakamoto et al. (1989), categorised foods based on their chewing energy patterns rather than their sensory scores. In this study, foods with different texture characteristics (e.g., rice crackers, toffee, peanut and surimi sausage) were used by measuring the activities of the masseter and digastric muscles, and the term "chewing energy" was introduced as a distinctive EMG parameter for different foods. The use of this parameter provided a convenient method for indirect measurement of foods with different textures.

Multiple parameters affect the EMG parameters during food oral processing. Diaz-Tay et al. (1991) reported that when the initial particle size (9.2 and 2.4 mm) and volume of peanuts presented to the subjects were changed, the masseter muscles were affected more than the anterior temporal muscles. They hypothesised that these effects were mainly due to the weight of the sample and its volume rather than the particle size. According to Le Reverend et al. (2016), saliva incorporation and mechanical action are the main parameters in food structure manipulation during chewing. They pointed out that model foods with a lower water content require less chewing effort and the work required to open the muscles varies during chewing, which is due to structural changes of the samples from a solid-brittle to a pasty-sticky texture. Previous research has shown that a higher amount of rice cake samples (9 g compared to 3 g) increased the values of the swallowing parameters (peak pressure and sound). In contrast, a decrease in the weight of the samples reduced the values

of the chewing parameters (such as chewing cycles and total chewing time). The opposite was found when a 9 g elderly-designed sample was compared with a 3 g standard sample. The results showed that using elderly-designed samples reduced chewing effort while maintaining the higher sample sizes (9 g). This study highlights the importance of carefully developing foods with the required muscle activity in mind. When developing food for older people, it is important that the food requires less muscle activity to prepare for swallowing and that the eating experience is also pleasant for older people (Kohyama et al., 2007b).

Human masticatory behaviour was studied by Kohyama et al. (2008) who reported that food must undergo catastrophic compression (high and very high) to affect human chewing behaviour. In their research they initially selected 63 masticatory parameters and then extracted three principal components related to EMG parameters by applying various statistical analyses. They also analysed different time intervals of chewing (early, middle and late) with varying degrees of compression-deformation (from low to very high). By doing EMG studies using side-imposed chewing, muscle activities can be greatly influenced. Mioche et al. (1999) reported that total muscle activity was reduced in about 75% of subjects when they were instructed to follow their habitual chewing style compared with side-imposed chewing (chewing only on the left or right side of the mouth). It was also reported that the activity of the masseter muscle was less sensitive to differences in the nature of food compared to the temporalis muscle. They indicated that the stickiness of the toffee samples might slow down the opening phase of each cycle, which could be an explanation for the lengthening of the sequence duration. Kohyama et al. (2016a) suggested that habitual chewing is deemed to be a superior approach compared to side-imposed chewing protocols. They analysed muscle activity during chewing of hydrocolloid gels of different concentrations. They concluded that as the mass of the gels increased, EMG parameters for the number of chews, chewing time and chewing effort increased. In addition to physiological differences, Mioche and Martin (1998) questioned whether or not training of assessors might affect EMG parameters. They used both trained and untrained assessors to assess the texture of beef samples. They concluded that trained assessors might develop an analytical mechanism in their masticatory muscle activity that allows them to assess the tenderness of beef more efficiently.

It is also important to note that the jaw opening muscles may also play an important role in the perception of stickiness. Kohyama et al. (2005) reported that an increase in the stickiness of cooked rice resulted in higher jaw opening muscle activity. They also found strong correlations between their EMG results and a two-step compression test using a texture analyser. The higher stickiness of cooked rice was related to its higher water content.

EMG parameters are not only influenced by the nature of food, but also by the development of masticatory behaviour. A recent systematic review by Almotairy et al. (2018) examined the maturation of sensorimotor control of the jaw and masticatory behaviour in children. As children grow and their dentition get complete (usually after the age of six), their EMG parameters and bite force improve. They found that children aged 10-14 years develop similar chewing and muscle activity behaviour to adults (e.g., improved chewing parameters). This means that researchers need to be aware that children under this age can show significant inconsistencies in their chewing patterns.

Among texture features, tenderness and hardness are two of the most commonly used by researchers (González et al., 2001b). Plesh et al. (1986) studied the effects of chewing gum hardness on chewing patterns. Analysis of EMG data showed that increasing masticatory muscle activity was related to harder gums and that the opening and occlusal phases of chewing played the main role in prolonging oral processing time. In another study, Shiozawa et al. (1999) found that peanuts require a relatively long chewing time as well as more chewing cycles to reach a safe point for swallowing, i.e. a so-called "swallowable consistency". The reduction in the amplitude of the masticatory muscles also correlated with the reduction in hardness (when comparing peanuts, gummi candy, and rice cakes) when chewing was continued. It was also suggested that the reduction in the amplitude of the digastric muscles could be a response to the reduced particle size or volume of the sample, which normally occurs at the end of chewing. In another study by Peyron et al. (2002) , the strongest manipulations of the hardness of gelatin-based model foods were found during the first five chews. It was hypothesised that the higher the hardness level, the more chews were required. As hardness was significantly correlated with EMG parameters (e.g., total muscle work and cycle muscular work), it was suggested that a single predictive parameter should be chosen to match the aim

of the study. Another study by Foster et al. (2006) found that higher masseter and temporalis activity was associated with higher hardness regardless of the type of model food. They studied different model foods with elastic and plastic properties and different degrees of hardness. The chewing apparatus manipulates the number of chewing cycles and total effort as a function of hardness. Although hardness plays an important role in the perception of oral texture, it was reported by Taniguchi et al. (2008) that increased viscosity, even with the same degree of hardness, delays the transit time of the bolus in the pharynx. In this study, the effects of hardness and viscosity of food on swallowing time and pharyngeal phase of swallowing were investigated. They suggested that total swallowing time was most affected by the increase in hardness.

Duizer et al. (1996) investigated the possible relationship between beef tenderness using sensory time intensity, EMG, and instrumental TPA. Their results suggest that the perceived sensory time intensity of beef tenderness is due to the chewing rates of the trained assessors, but they noted that the high variation in the data means that the conclusions need further consideration and possibly re-examination. By using the EMG and dividing the chewing process into initial, middle, and late phases, the authors found that tenderness was mainly associated with the initial phase of chewing with fewer mechanical changes. In contrast, Mathoniere et al. (2000) found that the tenderness of meat samples was more related to the middle and late stages of mastication, where the food undergoes catastrophic mechanical changes after chewing. They found that although the total number of chews varied greatly from assessor to assessor, it was the prominent predictor of the overall tenderness measure. A couple of papers by Carson et al (Carson et al., 2002a, Carson et al., 2002b), examined the firmness (or hardness) and cohesion of a range of model foods and food products. They used an Electronic Sensing System (ESS) which had similar functionality to the EMG but used a different analysis programme. They reported that the highest correlation of firmness of all samples was obtained by ESS parameters of total energy, peak energy, and Fourier power, while descending energy was a more reliable predictor of cohesive force. They suggested that ESS should be used to determine the firmness or hardness of samples with similar textural attributes rather than when there are large differences.

It can be further discussed that stickiness compared to hardness has some significant differences in terms of activation of the jaw muscles. Hardness is considered to be the force required by the masseter and temporal muscles to crush or deform food during initial chewing, whereas stickiness is reflected in muscle activity as the force required to open the mouth after compression by the mandible and maxilla. Stickiness can be measured not only from a few chews but from the whole chewing process.

1.2.8.3 The application of EMG in studying sensory and texture perception of food

Dividing the chewing process seems to be an effective method to gain a deeper understanding of the development of food texture during oral processing. Shiozawa and Kohyama (2011) studied the adhesiveness of biscuits and rice cakes in three stages during chewing (early, mid and before swallowing). The total number of chews, based on the activity of the masseter muscle, showed that adding water to the samples reduced the number of chews. However, instrumental measurements indicated that the adhesive force of the biscuit bolus increased from the middle to the last stage of chewing, while it was the opposite for the rice cake bolus. It was also suggested that the addition of water to the samples during chewing increased the ease of bolus formation for both model foods. The influence of water content on adhesion was highlighted by Kohyama et al. (2005), who reported that increasing the water content of the cooked rice samples correlated adhesion force and cohesion force with the mouth opening muscle groups as their activity increased. However, the opposite was true for the firmness of the rice: as the water content increased, the structure of the samples became softer, requiring less bite force and muscle activity.

Ingredients are the components of food that have a major impact on their texture and thus on the recorded EMG data. Kohyama et al. (2016c) conducted a study investigating the perceived texture of cooked rice, where the content of amylose varied as the most important parameter for the texture of the rice. EMG recordings of both masticatory muscles showed that the number of chews, total muscle effort and chewing time correlated positively with higher amylose content, while stickiness and time between chews (interburst duration) showed a negative correlation. Another

finding was that although amylose content was the main factor influencing the texture of the cooked rice, the influence of amylose in the late stages of chewing was lower and all samples behaved similarly.

Another group of researchers investigated that the effects of processing can also strongly influence perceived texture, even when ingredients remain constant. For example in a recent study by Gao et al. (2018) the effects of different mixing methods of bread dough and baking on the final products were investigated. The vacuum mixing of the dough followed by standard baking increased the density of the bread, which resulted in higher muscle activity and longer chewing time during consumption. The EMG parameters of chewing time and burst time were defined as key parameters for these products.

To summarise, EMG records changes of food texture during chewing and multiple parameters such as ingredients, processing and physiological characteristics of individuals affects the results obtained. As the muscle activity is related to the food texture, dominant textural attributes are easier to define (such as hardness) compared to stickiness as a complex attribute.

It should always be remembered that EMG has some limitations, and that extrapolation of data should be done with caution. Although EMG provides a large amount of data, analysing the data obtained can be challenging which is discussed in the next section . Therefore, it is necessary for food scientists to work with data scientists who have the appropriate knowledge of EMG data and its nature.

1.2.8.4 Parameters affecting EMG data collection

Several parameters influence the acquisition and interpretation of EMG data, and these parameters can significantly affect EMG values. Some of the most important parameters are explained below:

Influence of gender on EMG: Several studies have found gender differences in chewing behaviour (Woda et al., 2006a, Alsanei and Chen, 2014, Kohyama et al., 2016b, Almotairy et al., 2018). A possible reason was discussed by Alsanei and Chen (2014) by reporting that female subjects have a smaller mouth volume compared to male subjects. This could be the reason why they often prefer a smaller

bolus size when chewing. On the other hand, Woda et al. (2006a) reported that male assessors have higher vertical amplitude and higher masticatory and EMG activities per sequence, which could mean that female assessors chew more slowly compared to male subjects (Kohyama et al., 2016b).

Influence of age on EMG: The process of chewing develops from birth until about 14 years of age when chewing becomes an adult-like behaviour (Almotairy et al., 2018). It has been reported that mandibular vertical displacement or muscle strength decreases with age (average age of 80 years) (Karlsson and Carlsson, 1990). It was also found that with increasing age (from an average age of 29.4 to 67.7 years), the number of chews and total chewing time increased significantly. This was thought to be related to the decreasing muscle activity of the tongue and jaw elevator muscles with age (Kohyama et al., 2002). Another review study highlighted that there was a gender difference in EMG parameters in children under 12 years of age (Almotairy et al., 2018). As mentioned above, the results of age-related EMG studies should be interpreted with caution, especially when children are used as subjects. It is possible that their chewing behaviour is not yet fully developed. Consequently, the EMG results would be more affected by age-related physiological aspects rather than the food texture. A review manuscript by Mioche et al. (2004) indicates that chewing behaviour is affected in healthy aging and consequently bolus properties change, texture perception remains almost consistent. This suggests that the role of texture becomes more dominant with ageing. This could be related to the relatively superior preservation of texture perception receptors compared to chemoreceptors. These parameters of chewing behaviour and bolus properties are significantly affected by age-related parameters such as dental and health problems.

Influence of dental state on EMG: It has been stated that subjects recruited for an EMG study must not have any dysfunction of the temporomandibular joint (Braxton et al., 1996). Van der Bilt et al. (1994) reported that even with 2-3 teeth missing, the masticatory process is similar to subjects who are not missing teeth. However, some research has found that it is important that assessors should have at least 28 teeth for a sensory study (Chen and Engelen, 2012). As a screening criterion, the assessor should not have gum or periodontal disease and should not use a removable prosthesis (Gao et al., 2018). It should also be added that handling extremely stickiness materials may pose some risks even for a person with generally

optimal dental health. During some pre-testing of model foods for the current thesis, one of the supervisors lost a dental crown. While this was not a pleasant experience, it made it clear that extreme stickiness can significantly affect both EMG data and teeth!

Influence of general health conditions on EMG: There are different approaches to the criteria of a "healthy individual" for the selection of EMG subjects. The most relevant approach for the study of mastication is that subjects should be individuals with a healthy EMG signal in the time domain, who have no known neurological disorders, no masticatory or swallowing disorders or diseases of the head or neck (Lapatki et al., 2003, Chen and Engelen, 2012, Alsanei and Chen, 2014, Sadikoglu et al., 2017).

All of the above parameters are key factors in selecting human subjects for EMG studies. Some other parameters such as body mass index (BMI) and facial morphology are also considered important (Chen and Engelen, 2012). These parameters should be used as selection criteria for assessors.

1.2.8.1 Limitations of EMG

Although EMG can provide detailed data on the masticatory process from initial chewing to swallowing, it has some limitations that need to be considered.

Because EMG records the surface activity of muscles, it provides the average activity of a group of motor units in the muscle. Other EMG methods (e.g., using needles instead of surface electrodes) may reduce this effect, but could pose some serious problems in oral processing studies such as affecting habitual chewing patterns. Another limitation of the EMG method may be related to where the electrodes are placed on the subject's face, which can have a negative impact on reproducibility. This problem can be greatly reduced by placing the electrodes in the same location using physical landmarks. From an application perspective, the person responsible for setting up and conducting the EMG study must be well trained to attach the electrodes correctly and ensure that the signal is recorded with a minimal noise. It is also important to limit the number of samples in each session of EMG data collection to minimise fatigue, which in turn can affect data quality (Kohyama et al., 2015).

Another method to reduce variability in EMG data is to normalise the data as an effective component of data analysis (Szyszka-Sommerfeld et al., 2020). To obtain reliable data from EMG, careful experimental designs and standardisation approaches should be chosen. These are important for individual studies, but it can be challenging to compare data from different studies when different methods have been used. This can be mitigated by looking at overall trends rather than absolute values (Almotairy et al., 2018).

In summary, EMG is a technique that can provide very useful insights into the masticatory process by recording muscle activity. This method differentiates the textural properties of food, which offers valuable insights into the underlying parameters of mastication. It should also be carefully considered that EMG can lead to unreliable data collection if the correct methods are not followed. Thus, the design of EMG experiments is of great importance to obtain reliable data.

2 Materials and Methods

2.1 Model food

2.1.1 Development of the model food formulation

The original idea for the preparation of the model food was derived from Turkish Delight. There were three main reasons for this choice: firstly, the dominant stickiness that discriminates it from similar sweets. Secondly, Turkish Delight has a semi-solid to a solid structure, which makes it suitable for studying the stickiness of semi-solid foods. Last but not least, Turkish Delight was one of the stickiest foods known to the research team.

Sugar (sucrose), a low molecular weight compound, plays the main role in the stickiness of the model foods. The sugar content was varied in the model systems to alter the stickiness. Another important ingredient was native wheat starch, which is mainly responsible for the jelly-like texture of Turkish Delight. Citric acid prevented recrystallisation of the sugar during the production process and storage. Without citric acid, many sugar crystals form, especially at higher sugar contents, resulting in a dramatic loss of quality. In order to develop the model food with different degrees of stickiness, different approaches were followed before finalising the formula. The development process of the model foods is explained below.

To begin with, starch and citric acid were mixed with water. After they were mixed to the point where there were no lumps or non-dispersed particles, they were heated in a pan until they had a paste-like consistency. The reason the paste was made first was to gelatinise the starch. Then the sugar was added slowly while stirring and heating. Adding the sugar at the end presented two problems: It was a challenge to dissolve the sugar as the water was consumed by the starch. The other problem was phase separation after cooling the model food. It was obvious that the main cause of both problems was that the sugar did not dissolve completely, which was solved by adding the sugar earlier when making the starch paste.

Producing a reproducible model system is crucial, so careful consideration was required in the processing. First, a Thermomixer (Thermomix TM31, Vorwerk, Germany) was used: This appliance has some advantages, e.g., constant mixing speed, even heating and precise control of the process due to the closed container. This appliance was suitable for recipes with low sugar content, but some problems occurred with recipes with higher sugar content. Although the Thermomixer offers a constant mixing speed, mixing was not reliable as some parts of the paste did not mix properly with the rest of the recipe and some lumps were observed. In addition, the advantage of a closed vessel kept water vapour in the mixture, which caused problems as traditional Turkish delight recipes allow water evaporation. The final limitation of the Thermomixer was its limited heating capability. It was necessary to reach the boiling point, which the Thermomixer could not do.

Saucepans and manual mixing were used to solve some of the problems with the Thermomixer. Gelatin was an ingredient that increased reproducibility and gave a self-standing, sliceable and chewable texture. However, the role of gelatin was questioned after some initial informal sensory trials. Since the melting point of the gelatin solution is below body temperature, it results in a texture that can be chewed and is self-standing at the same time (Choi and Regenstein, 2000), but the model food began to soften at ambient temperature (20°C), and later in the sensory evaluation, the structure of the model food changed due to mouth temperature (around 37°C) during the first few chews. For these reasons, gelatin was excluded from the final model food formulations.

2.1.2 Final model food formulations

The first step in preparing the model was to weigh all the ingredients (Table 2-1). It should be noted that when sugar is used, this refers to sucrose. All the model foods were formulated with white granulated beet sugar (Sainsbury's, UK), native wheat starch (Foo Lung Ching Kee, Hong Kong), citric acid (Sigma Aldrich, Dorset, UK) and tap water (Table 2-1).

Table 2-1. Model food formulations. Sample codes are made up of the sugar concentration followed by the heating time (e.g., 65-120: 65 % sugar, 120 minutes heating time).

Sample code	10-90	35-90	50-75	65-75	50-120	65-120
Sugar (g)	10	35	50	65	50	65
Native wheat Starch (g)	8.5	8.5	8.5	8.5	8.5	8.5
Citric acid (g)	0.3	0.3	0.3	0.3	0.3	0.3
Water (mL)	81.2	56.2	41.2	26.2	41.2	26.2
Desired temp. (°C)	79	84	99	108	99	108
Final heating time (min)	90	90	75	75	120	120

It should be mentioned that waxy maize starch was also considered for the model foods. Although waxy maize starch has a high content of amylopectin, it was not able to give a semi-solid (self-standing) texture even in formulations with high sugar content. This could be due to the fact that waxy starches are not able to form gel structures (Iturriaga et al., 2006). This was the reason why this starch was removed from the recipes.

In the preparation of each model food, the total amount of sugar and 75% of water were placed in a saucepan and heated at medium intensity on an induction hob to achieve the desired temperature (Table 2-1). These temperatures were the point at which the sugar visually dissolved in water.

Then, while stirring with a spatula, citric acid was gradually added to the pot. The solutions were then heated for either 40 minutes (model foods with 10, 35, 50% sugar) or 60 minutes (model foods with 65% sugar content). Throughout the cooking process, the temperature was monitored regularly to ensure even heating.

The remaining water and starch were mixed separately and added to the saucepan. The final cooking time was chosen so that the model foods had different stickiness (Table 2-1). The mixture of model food samples was poured into 30 mL plastic containers and tubes. The 30 mL containers (with lids) were used for analysis with the texture analyser and the tubes (5 mL) were used for sensory evaluation (the tubes were the bulb part of a plastic pipette (Pipette Pasteur, PIP4206, SLS Select), which were carefully cut with sharp scissors). The model food samples were stored overnight in a cabinet at room temperature (approx. 20 °C) to harden.

Table 2-2 provides the values of sugar and water after cooking and in the final model foods.

Table 2-2. Water content and sugar levels of different model foods, before and after cooking. Values (except for water content before cooking) are presented as mean (Standard Deviation).

Model food	10-90	35-90	50-75	50-120	65-75	65-120
Sugar content after cooking (%)*	16.92 (0.23)	55.41 (1.55)	67.40 (1.15)	72.21 (0.77)	76.60 (0.86)	79.01 (0.37)
Water content before cooking (%)	81.20	56.20	41.20	41.20	26.20	26.20
Water content after cooking (%)**	68.19 (0.43)	30.43 (1.93)	20.74 (1.35)	15.08 (0.91)	13.03 (0.98)	10.51 (0.27)

* Sugar content after cooking was calculated by adding the loss weight after cooking to the sugar amount or in other words the total percentage of sugar was kept constant after the cooking

** Water content after cooking was calculated by subtracting the total weight of ingredient and saucepan before cooking from their weight after cooking

2.2 Sensory evaluation

Ethical approval (Ethics reference No.: 270-1803) for the sensory evaluation study was obtained from the Research Ethics Committee, Faculty of Medicine and Health Sciences, University of Nottingham (the copy of approval letter is provided in Section 6.2). Assessors signed to give their consent before taking part in the study.

The principal researcher was vaccinated against hepatitis B as a precaution before the start of the study.

2.2.1 Preliminary sensory testing of sticky model foods

A pilot test was conducted to ensure that the different degrees of stickiness of the model food samples were perceptible and distinguishable in the mouth. This test was conducted in two replicates by six untrained assessors comprised of students from the Food Science department (four females, two males). Stickiness was defined as the effort required to separate the teeth, while the model food was in the mouth. The

definition of stickiness was presented to each assessor before they discussed the stickiness of the model foods. After the presentation of the model foods, assessors were asked to explain which foods they found sticky and which they did not. In this way, a general consensus was reached that assessors understood the term stickiness. Samples were coded with random three-digit numbers and presented in a random order. Each panel member was presented with a sample and asked to rate the stickiness on an unstructured line scale (non- sticky to very- sticky). The weight of all samples used in the preliminary sensory sessions was 5.08 g (SD: 0.4) and they were served at room temperature using plastic cups in 30 mL cups.

This pilot evaluation provided valuable data by ensuring that the model foods were significantly different from each other and to make the selection for the main sensory study (see Appendix 6.3).

2.2.2 Assessors for the main sensory study

Ten assessors (4 females, 6 males) aged 21-27 years were recruited from students at the University of Nottingham and participated in two training sessions and two data collection sessions (four sessions in total). There were four replicates of each model food. Assessors were paid £15 (2 hours) for the two sessions at the Sensory Science Centre (SSC), Sutton Bonington Campus, University of Nottingham, and £20 for the two sessions at the Clinical Sciences Building, City Hospital Campus (an additional £5 to reimburse transport costs to the City Hospital Campus).

Assessors were selected from healthy individuals (self-reported) with at least 28 natural teeth. Exclusion criteria were participants who wore dentures and crowns. Assessors were instructed not to smoke or drink coffee or other strong drinks for at least 2 hours before the session.

2.2.3 Training sessions

There were two training sessions for each participant, held at SSC. The training sessions started with a definition of stickiness. In order to obtain specific descriptions of stickiness, five terms were used, which are listed in Table 2-3. The 5 terms selected in the current study were perceived as descriptive of stickiness and they

were used as a unifying term for stickiness perception which formed a comprehensive definition of stickiness. To make it easier for the assessors, only the definitions were given to them, and the actual terms were not mentioned.

During the training sessions, one reference sample was selected for each definition (except for 'cohesive' where two were selected). The selection of reference samples for each definition started with the purchase of different confectionery products on the UK market, each of which the principal researcher tested to decide which term was the predominant characteristic for each sample. The reference examples for cohesion represented the lowest and highest scales of that term to enhance the comprehension of cohesion. The terms, definitions and reference examples are listed in Table 2-3.

In the training sessions, the examiners were instructed to put each reference sample in their mouths and chew it, in their habitual manner, until it was swallowed. When the assessors were given further definitions of the terms, they were asked to think of all the definitions while chewing. They were also asked to compare different reference samples with the other samples. Although standardised chewing is able to reduce the variations among assessors in sensory studies, it has been highlighted in EMG studies that habitual chewing is a preferred method to measure muscle activity. Standardised chewing impacts the natural muscle activities (Mioche et al., 1999, Kohyama et al., 2016a).

Table 2-3. Terms, definitions and reference samples used in training sessions.

Term	Definition	Reference sample
Enveloping	Leaves residual material on side surfaces of teeth	Werther's original. Creamy toffees (August Storck KG, Germany)
Stringy	Forms strings as you pull teeth apart	
Tacky	Adheres to teeth, resists separation	Rowntrees; Fruit gums (Nestle, UK)
Cohesive	Pieces reform together	Maoam; Joystixx Swizzles (Dunhills, UK), Drumstick Squashies (Swizzels, UK)
Tooth packing	Packs in teeth - related to quantity that packs	Rowntrees; Fruit pastilles (Nestle, UK)

Data collection during the training sessions was recorded as follows: An unstructured line scale was used in Compusense (Compusense Inc., Canada) to score the model food samples (additional information in Section 2.3.1). The left end of the line was designated as non- sticky or as minimally perceptible stickiness, while the right end of the line was described as very sticky or as maximally sticky. Assessors were asked to tap the iPad screen to mark the line as a measure of perceptible stickiness. The assessors were advised to use both extremes of the scale for very sticky or non-sticky samples. To practise this during the training sessions, the assessors were given low-sticky (10-90) and very-sticky (65-120) model food samples (two replicates) and instructed to rate each at the extreme left and right ends of the scale, respectively. Assessors squeezed the model food samples from the tube onto the top of their tongue. They also used crackers as palate cleaners between samples.

2.2.4 Mouthwash preparation

To ensure that there was no interference between stickiness and sweetness perception it was necessary to inhibit assessors' sweetness perception. To do that, a mouthwash was prepared based on *Gymnema sylvestre* leaves to block sweetness perception. As sweeter samples are also stickier, the aim of the mouthwash was to decouple the two properties to minimise the stimulus error. To prepare the mouthwash, 7.5 g of dried leaves of *Gymnema sylvestre* (Eldira, Bulgaria) were added to 150 g of distilled water and heated to 90 °C for 60 minutes at the lowest mixing speed (40 rpm; TM31 Thermomixer (Vorwerk, UK)). The mixture of leaves and water was filtered, cooled and stored at 4 °C for up to 48 hrs before the sensory tests (Meiselman and Halpern, 1970). Before each sensory session, mouthwash was taken out of the fridge at least one hour before use to warm up to the ambient temperature.

The activity of mouthwash was verified using six student volunteers in Food Science Department. The volunteers were asked to rinse their mouths with the mouthwash as detailed in Section 2.2.5, then they were asked to put some granulated sugar (sucrose) in their mouths and indicate when the sensation of sweetness resumes. The use of the mouthwash resulted in a loss of sweetness perception and its gradual return. The average time of perception recovery was 20 minutes, with a range

between 18 and 24 minutes. Based on these results, 15 minutes was determined as a period of time where inhibition of sweetness perception was effective. For the sensory tests in the main study, the mouthwash was applied twice to ensure that sweetness perception was effectively inhibited for the entire duration of the sensory tests.

2.2.5 Mouthwash application procedure

The assessors' normal perception of sweet taste was first tested by asking them to indicate whether they perceived the sweetness of granulated sugar. If so, they were found to be able to perceive the sweetness. To eliminate the subjects' perception of sweetness, they were asked to rinse their mouths with *Gymnema sylvestre* mouthwash (10 mL, served in a 30 mL plastic container). The assessors were asked to swirl the mouthwash around in their mouth for 60 seconds in order to ensure that every part of the mouth responsible for the perception of sweetness was in contact with the mouthwash. Then they were instructed to expectorate the mouthwash into a 50 mL container and close the lid tightly. Then, they were asked to rinse their mouths with water and wait for three minutes. To check the effectiveness of the mouthwash, the assessors were given sugar granules (5 grams presented in 30 mL plastic cups). All assessors confirmed that the sugar felt like sand in their mouths and had no taste, indicating that they could not perceive its sweetness.

The expectorated mouthwash in the 50 mL containers was treated as biological waste. At the end of each session, the containers were placed in a box labelled "Biohazard" in the sample preparation room of the City Hospital and the lid of the box was carefully sealed with tape. The box was then placed in a designated area. This procedure was performed after each session. After all sessions, a 3% Virkon solution was prepared and added to the expectorated mouthwash in each container in approximately a 1:1 ratio. This was left for 30 minutes to allow the Virkon solution to completely disinfect the expectorated material. Finally, the treated mouthwash material was poured into the sink with running water from the taps. The plastic containers were disposed of using the standard waste disposal procedure.

2.2.6 Masking the colour differences of model foods

Colour is one of the key sensory attributes. Although colour is not affecting stickiness perception in a direct way, it has an effect through so-called multimodal integration

mechanisms. These mechanisms would suggest that more intense deeper colours would be more likely to be associated with higher stickiness in comparison with lighter or pale colours that would be associable with more watery textures. To minimise the effect of the colour difference between the model foods on assessors' perception of stickiness, the room in the Clinical Sciences Building on the City Hospital Campus was adapted to enable sensory data collections. This was done to enable sensory test to be run in parallel with the EMG experiments, the equipment for which was stationed in the same building. To transform the room, the windows of the room were covered with wrapping paper and the room was darkened. Then two 60 W red fire glow lamps (British Electric Lamps Ltd., Bell, UK) were connected to clip-on lamp holders (Wilko 2 m mini clip-on light) as shown in Figure 2-1. Prior to the session, three different light bulbs were tested to find out which one effectively cover the colour differences in the model food samples.

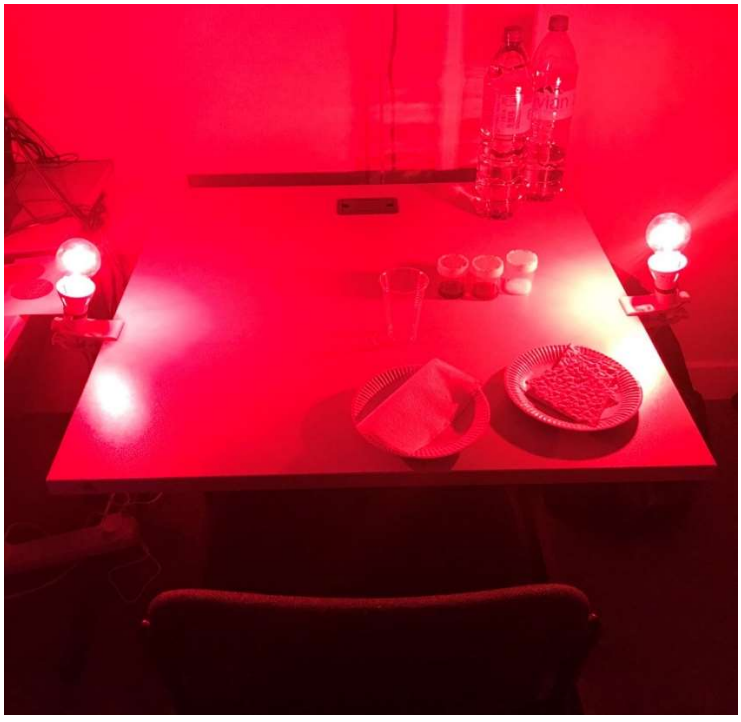

Figure 2-1. The set-up to cover the colour difference between samples and EMG set up used for collecting data at City Hospital campus.

Figure 2-2 shows the colour difference of the model foods with and without the use of the red light. It can be seen that the red light clearly minimises the colour differences between the model foods and subsequently reducing the expectation error by the assessors.

Figure 2-2. Using red light to minimise the colour difference among model foods. Top: before applying the red light, bottom: after applying the red light.

2.3 Surface Electromyography (sEMG)

The sEMG method was used to measure the electrical activities of the muscles responsible for the mastication process. The activity of the following muscles was recorded in this study: left and right masseter, left and right temporalis, and digastric muscles (see section 1.2.7.2). A Noraxon TeleMyo transmitter (TeleMyo 2400T G2) system was used for recording the muscle activities. Self-adhesive dual electrodes (Duotrodes #6145, Myotronics, USA) with a centre-to-centre distance of 19 mm were employed.

The principal researcher was trained by the EMG expert at the city hospital to place the electrodes by palpating the muscles before placing them on the skin of the assessors. To palpate the temporalis and masseter muscles, the subjects were asked to clench their teeth, and for the digastric muscles, assessors were asked to swallow some water. This allowed the principal researcher to identify the exact location of each muscle group where the electrodes should be placed. To ensure consistent placement of the electrodes, anatomical landmarks were selected and recorded for each subject, which were used in the EMG data selection sessions.

Before attaching the electrodes and to ensure complete contact between the electrodes and the skin, the assessor's skin was carefully rubbed and cleaned by applying a small amount of an alcohol-based hand lotion (Softalind pure, B. Braun, UK) to a tissue to remove the dead skin and any other residues that might interfere with high quality recording of the EMG signals. The skin was allowed to dry before attaching the self-adhesive electrodes.

The reference electrode was placed on the forehead of the assessors in order to have the baseline of noise for the recorded EMG signal. The reason for choosing the forehead was that the location of the reference electrode should be on a part of the body that is electrically neutral (Chen and Engelen, 2012).

The male participants were asked to be clean-shaven before the EMG sessions so that the EMG probes could be placed on the skin.

2.3.1 Data collection method and Compusense cloud

The time-intensity method with discrete-time points was used to collect the data.

Compusense cloud is sensory evaluation software that is used for collecting data by the Sensory Science Centre (SSC) at the University of Nottingham. Each assessor rated the samples in Compusense using an iPad (Apple Inc., USA). The Compusense software randomly assigns 3-digit codes to each sample type and also randomly assigns the order that each assessor will receive the samples.

Instructions (stages) that were given to assessors in this software were as follows:

1. Pour the mouthwash into their mouth and tap on the start button
2. Keep the mouthwash in their mouth and move it around their mouth for one minute without swallowing
3. Spit the mouthwash into a 50 mL pot and close the lid tightly. Wait for 3 minutes
4. The model food sample was presented in a tube and served in a 30 mL container with a closed lid (in order to eliminate any contamination). The containers were labelled with 3-digit numbers produced by Compusense

5. Squeeze the model food sample from the tube onto the top of their tongue
6. To rate the stickiness, an unstructured line scale from non-sticky (minimum) to very-sticky (maximum) was used
7. Tap the start button and start to chew the model food sample. Every five seconds after the start point, measure the stickiness of the sample until the point of swallowing
8. As swallowing occurs, tap the finished button and finally rate the overall perception of stickiness of the sample, using the same unstructured line scale
9. Two-minute break, to eat some crackers and water in order to clean their mouth from any possible residues of the model food
10. Repeat for all model food samples (stages 4 to 9)
11. Rest for ten minutes
12. Steps 1 to 9 were repeated for all the model food samples in a different random order

At each session, assessors were given 12 samples. The mouthwash was given twice to the assessors to ensure that the perception of sweetness was blocked throughout the session and to avoid gradual recovery. Six samples were prepared for the evaluation, so that each assessor received each sample twice: once in the first part and once in the second part. Each set of six samples was randomly ordered by Compusense and the order was different in each part of the session.

The following sections (2.3.2 and 2.3.3) were written with the help and supervision of Daniel Prado de Campos, who was the EMG expert involved in the EMG data analysis of the thesis.

2.3.2 Signal processing of EMG data

The EMG signal was digitally filtered in the range 5-500 Hz with a 4th order Butterworth filter to select the signal band of interest. The power line noise (50~Hz) and its harmonics (2nd, 3rd, 4th and 5th) were removed by a notch filter (Q = 30). The signal was rectified and smoothed using a 2nd order low-pass Butterworth filter with a cut-off frequency of 40~Hz. The four channels were summed to a single signal vector.

Figure 2-3 shows an example of EMG raw signal for a female assessor during mastication of model food 65-75. Muscle activities from left and right Temporalis muscles, left and right Masseter muscles and left and right Digastric muscles are presented. The Signal to Ratio (SNR) of the muscles was used as a validation step. The digastric muscle showed a low SNR which can lead to false detections and errors. For this reason, digastric muscle was excluded from further analysis.

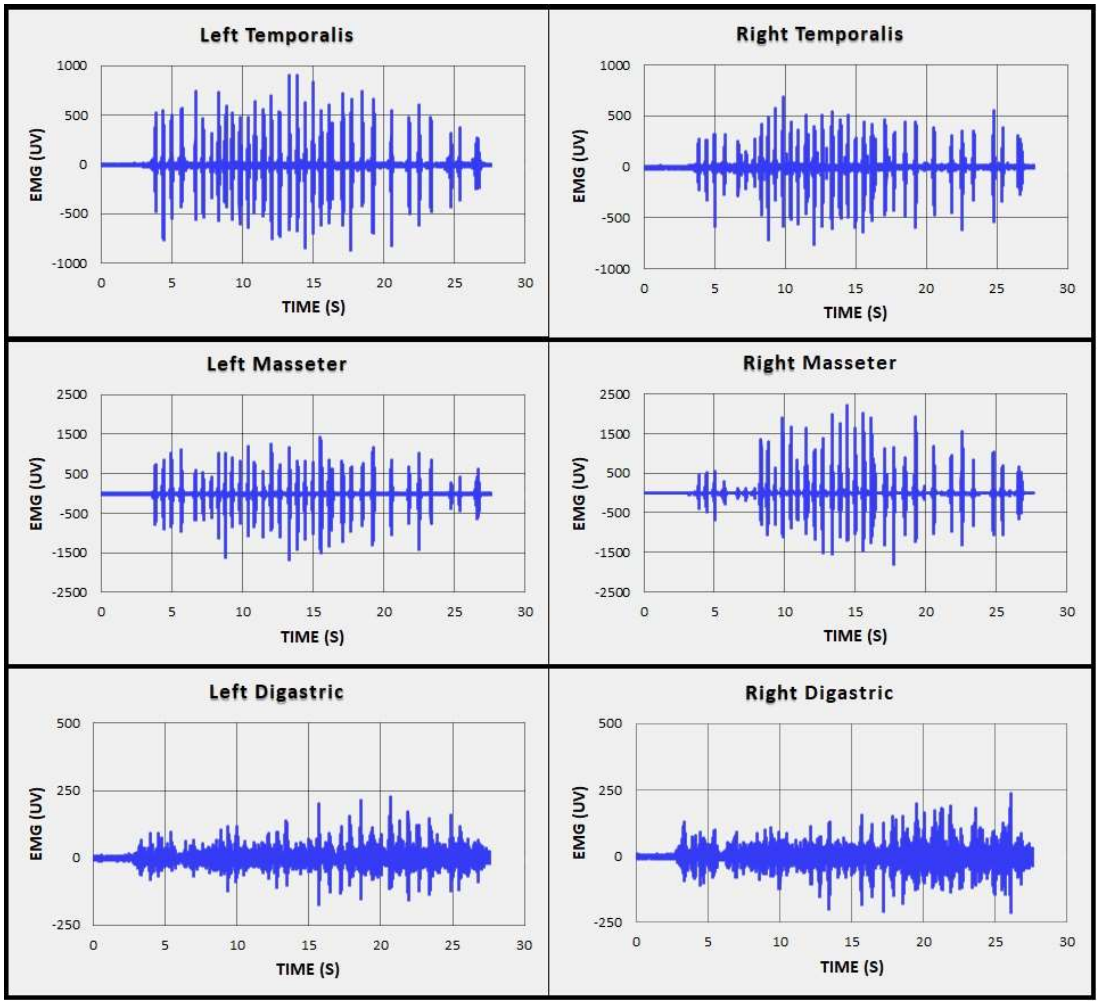

Figure 2-3. Example of EMG raw signal. Muscle activities from left and right Temporalis muscles, left and right Masseter muscles, left and right Digastric muscles of a female subject during mastication of model food 65-75.

The Double Threshold Onset Segmentation (DTOS) algorithm was used to divide each chew into different segments. The segmentation consisted of two steps; First step was to define the onset (beginning) and offset (end) from the threshold of the noise baseline. The threshold (th) can be calculated by the following equation:

$$th(BL) = \mu_{BL} + \kappa * \sigma_{BL} \quad (2\text{-}1)$$

BL: baseline noise extracted from the signal vector

μ_{BL}: mean of the baseline

σ_{BL}: standard deviation of the baseline

κ: predefined factor

Then, the threshold was used for detection of chewing. On the other hand, when signal under passed the threshold, the offset was detected. Therefore, the time between the onset and offset was defined as a signal window.

The next step was to validate whether the window length (W) was over a predetermined critical value (W_{crit}). The reason behind this verification step was due to the possible implication of signal artefacts for short signals that may cause false detection of them instead of a chew signal.

Therefore, if onset time (start of chew) is (t_{on}) and offset time (end time of chew) is (t_{off}), then a segment is valid if:

$$W = t_{off} - t_{on} > W_{crit} \quad (2\text{-}2)$$

The segmentation steps are presented in Figure 2-4. The EMG signal is from the left Temporalis muscle of a female assessor and the data was rectified. It can be seen that the threshold defines the base line and subsequently the onset and offset of individual chew (burst) are detected. Then, the time between the onset of one step and offset of the next step defines the signal window which is used for feature extraction.

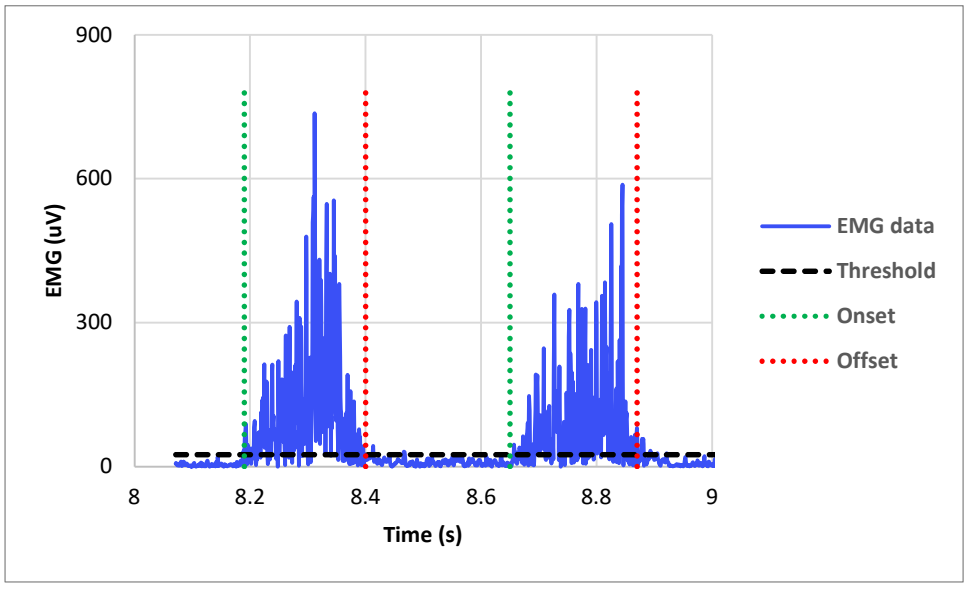

Figure 2-4. EMG signal segmentation steps. The threshold defines the baseline and then onset and offset are detected. The time from each onset to the following offset is a signal window. The EMG signal is from the left temporalis muscle of a female assessor and the data was rectified.

2.3.3 Feature extraction

The extracted features in the present study are more of temporal aspect of EMG bursts such as the Interchew time, the cycle duration and the burst duration. Signal window is based on the relation of the area of each burst to muscle activity (Chew Work). Figure 2-5 displays the EMG features of Chew Work, Burst Duration, Cycle Duration and Interchew Time taken from each cycle.

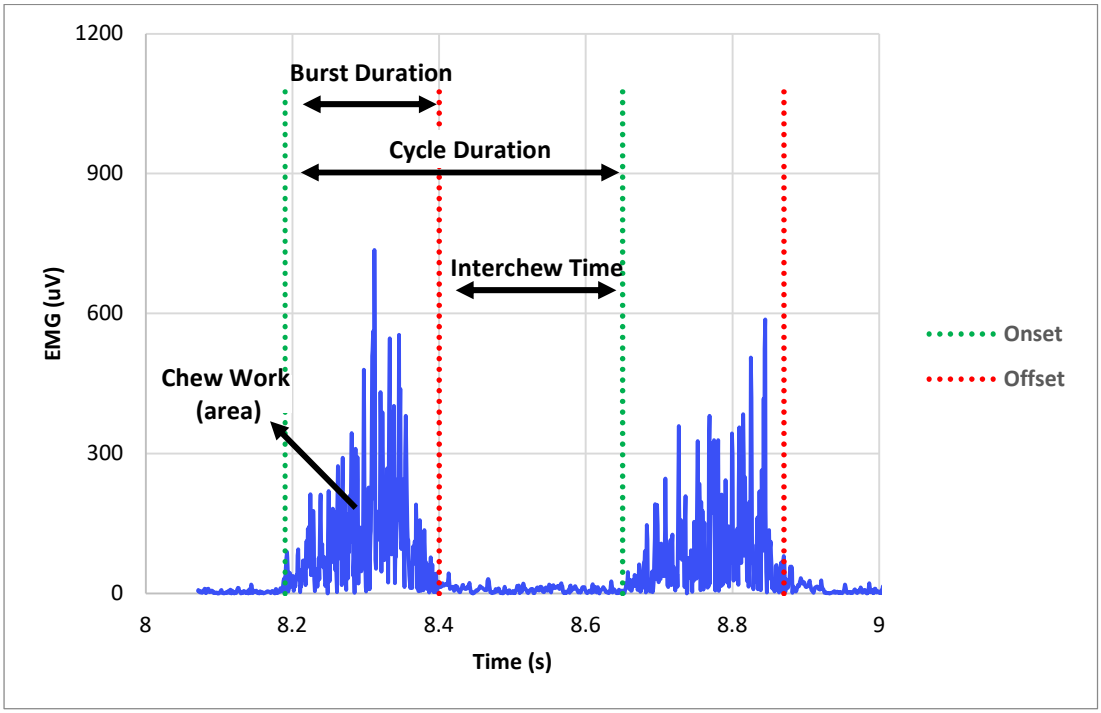

Figure 2-5. EMG features taken from each cycle. Chew Work: the area under of EMG data between each consecutive pair of onsets and offset, Burst Duration: the time between each consecutive pair of onsets and offset, Cycle Duration: The time between the onset of a chew to the next onset, Interchew Time: the time from the offset of a chew to the next onset.

In the following part, the onset and offset times are presented as t_{on}^{i} and t_{off}^{i}, respectively. Considering these, the Burst Duration of the i-th chew (a single chew during mastication) (BD_i) is calculated as:

$$BD_i = t_{off}^{i} - t_{on}^{i} \quad (2\text{-}3)$$

Similarly, the Cycle Duration of the i-th chew (CD_i) is defined as the time period from an onset to the following one:

$$CD_i = t_{on}^{i+1} - t_{on}^{i} \quad (2\text{-}4)$$

The next extracted parameter is the Interchew Time of the i-th chew ($ICh T_i$). It is the period between an offset to the beginning of the next consecutive onset:

$$ICh T_i = t_{on}^{i+1} - t_{off}^{i} = CD_i - BD_i \quad (2\text{-}5)$$

It can be seen from Figure 2-5 that $ICh T_i$ can be calculated from the difference of CD_i and BD_i.

The total time of the mastication (T) is the time period from the first onset to the last offset, as follows:

$$T = t_{off}^{M} - t_{on}^{1} \quad (2\text{-}6)$$

M: the number of total chews.

The Chew Work ($IEMG_i$), is the area of the EMG signal for each burst during the i-th chew and it is equal to the integration of the EMG signal (sum of each amplitude within the section). Then, N_i is equal to the length of i-th chew and also the signal is represented by $X_i = \{X_{i,1}, \dots, X_{i,k}, \dots, X_{i,N_i}\}$. Considering all these, the chew work is analysed as:

$$IEMGi = \sum_{k=1}^{N_i} |X_{i,k}| \quad (2\text{-}7)$$

The above features are descriptors of individual chews during the mastication process. Furthermore, additional features can be derived from the aforementioned metrics, contributing to a more thorough understanding of the complete mastication process. These additional features provide valuable insights into various aspects of mastication, enriching our knowledge of this intricate physiological process.

In order to reduce the inter individual bias, the EMG data was normalised in the range of [0:1] by using the maximum values of individual assessors. This also kept the range of signal constant throughout the analysis.

To have an accurate representative of mastication process, the signal values of each muscle (left and right Masseter and left and right Temporalis) were added together to make a single signal.

By summing up all the values of $IEMG$ (Chew Work), the parameter of total Chew Work (ChW) as an indication of all the chewing process is produced. Due to different characteristics of the first chew from the rest of the chews, it was excluded from the data. Accordingly, if the mastication has M chews the ChW feature can be calculated as follows:

$$ChW = \sum_{i=2}^{M} IEMG_i \tag{2-8}$$

The next feature is the Work Rate of the i-th chew (WR_i) which is the work within a section:

$$WR_i = \frac{IEMG_i}{CD_i} \tag{2-9}$$

By averaging this value ($\overline{WR_i}$), the Chew Work Rate (WR) can be calculated as below:

$$WR = \overline{WR_i} = \frac{1}{M-1}\sum_{i=2}^{M} \frac{IEMG_i}{CD_i} \tag{2-10}$$

It should be noted that the over line represents the average of all bites, excluding the first one. The Chew Work Rate is the effort per time during the bite and it is apparent that it is time dependant. It means that the same Chew Work in different times can result in greater or weaker WR values. The Chew Work Rate provides insights into the chewing over time, while the total Chew Work represents the total mastication.

The next extracted EMG parameter in the Proportional Work (pW). It represents the muscle work within section ($IEMG_i$) over the total Chew Work (ChW). It is a normalised metric of mean Chew Work and describes how it changes over time. The pW feature can be averaged to represent the mastication process as follows:

$$pW = \frac{1}{M-1}\sum_{i=2}^{M} \frac{IEMG_i}{ChW} \tag{2-11}$$

The mean of the Chew Cycle, can also be used as a feature, representing how fast the chews are within a mastication process. Defining this feature as Average Duration of Chews (ACh), it can be calculated from:

$$ACh = \overline{CD_i} = \frac{1}{M-1}\sum_{i=2}^{M} CD_i \quad (2\text{-}12)$$

In the same way, the mean Interchew Time ($IChT$) and the mean Burst Duration (BD) can be defined from $\overline{IChT_i}$ and $\overline{BD_i}$, respectively.

The Number of Chews (NCh) is the feature that represents how many chews are performed before the sample is completely swallowed.

It can directly be derived as; $NCh = card\,(t_{on})$

The total Chew Time (ChT) is the amount of time where chews are observed. It is equivalent to the total time (T) mentioned previously.

The Chew Rate (ChR) is the mastication rate or the frequency of chews, which is the ratio between the total Number of Chews (NCh) and the total Chew Time (ChT), therefore:

$$ChR = \frac{NCh}{ChT} \quad (2\text{-}13)$$

Regarding the EMG signal within the section, a vast list of features could be extracted. In studies involving chewing feature extraction, peak amplitude information is frequently used. The peak of the i-th chew (pk_i) can be calculated as below:

$$pk_i = max\{x_i\} = max\{X_{i,1}, \dots, X_{i,k}, \dots, X_{i,N_i}\} \quad (2\text{-}14)$$

Thus, a vector of peaks (pk) from all M windows during a mastication process can be expressed by:

$$pk = \{pk_1, \dots, pk_i, \dots, pk_M\} \quad (2\text{-}15)$$

Therefore, the Maximum Voltage Peak Amplitude (MV) is defined as the highest peak observed from the peaks of all windows:

$$MV = max\{pk\} = \underset{i=2:M}{max}\left\{\underset{k=i:N_i}{max}\{x_i\}\right\} \quad (2\text{-}16)$$

and the Average Voltage Peak Amplitude (AV) is defined as the average of the observed peaks:

$$AV = \overline{pk} = \frac{1}{M-1}\sum_{i=2}^{M} \underset{k\,=\,i:N_i}{max}\{x_i\} \quad (2\text{-}17)$$

Last but not least, the Median Frequency Shift (MFS) which is an indication of muscle fatigue can be obtained as follows:

$$MFS_i = MF_i - MF_1 \quad (2\text{-}18)$$

MF: Median Frequency

It should be noted that MFS is calculated for i-th chew in relation to the first chew.

It is important to note that the features extracted from the EMG are related to the motor unit recruitment and, therefore, the effort required to perform the chew.

The extracted EMG features are summarised in Table 2-4.

Table 2-4. Extracted EMG features and their abbreviations.

Feature full name	Abbreviation	Unit	Definition
Total Chew Work	ChW	µV *	Sum up all the values of chew work
Chew Work Rate	WR	Numerical values	Effort per time during the bite
Proportional Work	pW	µV	Muscle work within section chew work over the total Chew Work
Average Duration of chews	ACh	s**	Duration of chews within a mastication process
Number of Chews	NCh	Numerical values	Total number of chews during mastication
Chew Time	ChT	s	The time from the onset of a chew to the next onset
Chew Rate	ChR	Numerical values	Frequency of chews
Interchew Time	$IChT$	s	The time from the offset of a chew to the next onset
Burst Duration	BD	s	The time between each consecutive pair of onsets and offset
Maximum Voltage Peak Amplitude	MV	µV	Highest peak observed from the peaks of all windows
Average Voltage Peak Amplitude	AV	µV	The average of the observed peaks

Median frequency shift	MFS	Hz***	An indication of muscle fatigue

- * Micro Volt
- ** Second
- *** Hertz

2.4 Instrumental measurement

2.4.1 Texture analyser measurement

A texture analyser (TA) HD Plus (Stable Micro Systems, UK) with a flat-ended cylindrical probe of 6 mm diameter and a load cell of 5 kg was used to perform a single compression test on the model food samples. The experimental protocol for the compression test is shown in Table 2-5.

Table 2-5. Texture analyser experimental protocol.

No.	Stage	Comment
1	Move probe at 1 mm/sec to the surface of the sample	
2	As soon as probe touches the surface, move to force 1 g at 1 mm/sec (Holding step)	In order to have full contact between probe and model foods
3	Set data capture ON	
4	Move down 2 mm at 1 mm/sec (First compression step)	In order to have full contact between probe and model foods
5	Move to force 10 g at 1 mm/sec (Second compression step)	These two parameters were crucial for minimising necking and preventing the penetration of the probe into the model food
6	Move up 70 mm at 10 mm/sec (Separation or withdrawal step)	
7	Wait for the target condition	
8	End test	

In general, a compression test is mainly concerned with the value of hardness, i.e., the maximum positive force for a given displacement exerted by the probe pressing on the surface of a material. The adhesion or pull-off behaviour is measured during pull-off in a compression test. A combination of the indentation and separation branches of the force-vs-distance curve is called a "compression-separation test", the term used throughout this thesis.

The experimental protocol was designed using Test maker software version 7.0.0.0. (Stable Micro Systems, Godalming, UK). Instrumental data from the texture analyser was recorded using Exponent software version 6.1.16.0. The reported data are the mean of triplicates, and all the measurements were carried out at room temperature (about 20 °C).

The experimental setup of the texture analyser is shown in Figure 2-6. The annotations in the figure are as follows: A- a flat-ended cylindrical probe with a diameter of 6 mm attached to a 5 kg load cell. B- the 30 mL plastic container with the model food sample attached in a hollow metal cylinder. The assembly was attached to the heavy-duty platform of the texture analyser with a 3-point rig (C). No attempt was made to fix the cup as it was held by the hollow metal cylinder. The photographic images were taken with an iPhone 7 Plus (Apple Inc., US).

The derived parameters from the compression-separation test include the initial gradient (mN/mm), force of the adhesive peak (mN) and distance to the adhesive peak (mm), the total area under the negative curve ($mN.mm^2$), and the pre-area ($mN.mm^2$). The latter was calculated from the area under the negative curve by drawing a line from the peak force to the zero value on the distance axis and calculating the area from the start of the curve to the said line. Prior to obtaining the above parameters, the horizontal time-axis (s) was transposed into the distance (mm). The initial gradient of the negative curve is calculated by selecting two points on the early parts of the negative curve before it starts to deviate from a straight line.

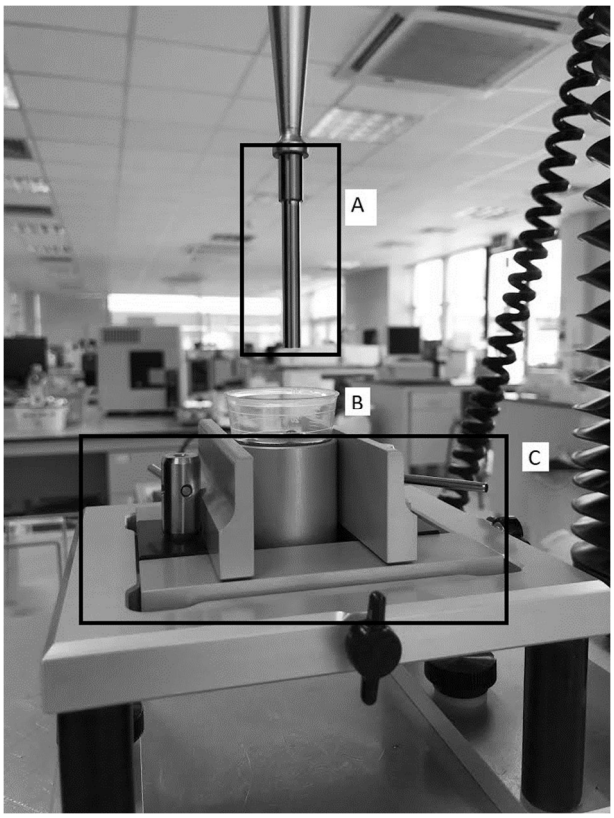

Figure 2-6. Texture analyser experimental set-up

A: flat ended 6mm diameter circular probe, B: 30 mL pot container housing the model food sample, C: 3-point bending rig that holds a hollow metal cylinder that accommodates the plastic container.

2.4.2 Rheological measurement

The linear viscoelastic range (LVE) of the model food samples was determined by amplitude sweep tests using oscillatory shear technique. For each model food, 25 data points were collected using an angular frequency of ω=10 (1/s) and a stepwise (logarithmic ramp) oscillatory shear strain (γ), starting at 0.01% and ending at 100%. All tests were performed 3 times at 25 °C. The LVE range was determined from a plot of shear strain on the x-axis and G' and G" on the y-axis, considering the range where G' and G" are constant and do not depend on the oscillating shear amplitude (Figure 3-8 and Figure 3-9). By determining the LVE range for each of the model food samples, it was possible to determine the optimum oscillatory strain for further experiments that could measure rheological values without structurally damaging the samples. The limit of the LVE range was 0.1% and

was calculated using RheoCompass™ software (V1.21.825-Release, Anton Paar, Austria).

The stress relaxation measurements were carried out using a stress-controlled rheometer Physica MCR 301 (Anton Paar, Austria), using the concentric cylinder geometries for running the experiments. The measuring cell (cup) was a (C-PTD200) with the measuring system (concentric cylinder) CC27. The cup was filled with 25 g of a model food sample which was almost equal to the mark inside the cup indicating the filling level. The measuring system was lowered into the cup and then it was left for 30 min to allow the sample to rest and its internal structure to reform. At the start of the resting time, the top of the cup was covered with filter paper, which was kept moist throughout the experiment to prevent evaporation. RheoCompass software was used to collect the data. The values of the storage modulus (G´), the loss modulus (G˝), the complex viscosity (η*) and the angular frequency (ω) were obtained using the RheoCompass software.

The relaxation modulus was measured using a step-strain test. The step strain test consisted of recording 100 points at four second intervals at 0.01% strain and then at an increased strain of 0.1% strain by recording 400 points at four second intervals. As mentioned above, the shear strain values were selected from the Linear Viscoelastic Region (LVE-range).

Following that, the relaxation data were generated in three steps (using a routine embedded in the RheoCompass software):

1. The relaxation modulus G(t) was measured in the step strain experiment,
2. The relaxation-time spectrum H (lambda) measurement was calculated,
3. The relaxation-time spectrum was converted to G´ (omega) and G˝ (omega).

2.4.3 Bulk modulus measurement

The experimental set-up to do the bulk modulus measurements is described here. The items used for bulk modulus apparatus are listed in Table 2-6.

Table 2-6. Bulk modulus apparatus items.

Item	Description/comment
DURAN® pressure plus+ laboratory bottle GL 45	Screw cap GL 45, PP, 2 port GL 14
DURAN® pressure plus+ laboratory bottle GL 45 fittings	1129816 Insert for screw cap GL 14, ID 3.0 mm (~1/8 inch)
Pressure air gauge	Thamesair
Tube	PVC flexible 1/8-inch tube (diameter: 1.27 mm)
Syringe	20 mL (SOFT-JECT)
VWR chemicals - Silicone oil 20 cSt (84543.290)	Polydimethylsiloxane, viscosity (25°C): 18-22 cSt, density: 0.95 kg/l,

A pressurised Duran bottle contained the model food samples. The bottle had two ports (Figure 2-7 c, d). A 1/8-inch flexible tube for pressurised air input was attached to one port (Figure 2-7d), whilst another tube with the same diameter was attached to the second port (Figure 2-7c) and was used to fill and then seal the bottle.

Prior to each experiment, the bottle was inspected visually for any signs of damage, minor cracks, or major scratches.

Figure 2-7. Duran bottle used for bulk modulus measurement- a: Duran bottle, b: sealing cap, c, d: ports of the bottle, e: cap used for input air pressure.

The first step of setting up the apparatus was to put the model food sample with a known volume into the Duran bottle. Before the bottle was tightly capped, it was filled with silicone oil to the top (approximately 100 mL).

After that, two 20 mL syringes, nominally named syringe-A and syringe-B, containing approximately 15 mL silicon oil were attached to the ports of the bottle. The plunger of syringe-A was compressed to fill the remaining empty head space of the bottle. This resulted in almost all the air being replaced by the oil; the air was forced out of the bottle into syringe-B. Syringe-B was then detached from the bottle, and a pressurised air tube input was attached which had been filled to a specific level by silicone oil. In order to eliminate any remaining air, the bottle was inverted and tapped gently until all smaller bubbles was contained in one large bubble. The bottle was then slowly returned to its vertical position, so the single air bubble could be removed by pulling back the plunger of syringe-A. Finally, this syringe was removed from the bottle, and it was sealed using a screw cap. If any air bubbles still remained in the bottle, they could be removed through the pressurised air tube input by tapping the tube a few times to allow the bubble to rise through

the oil in the tube. The pressurised air tube was held by a stand clamp to keep the level of silicone oil in an upright position. It was crucial to position the bottle and the air tube precisely so that the level of oil could be observed before and after applying pressure.

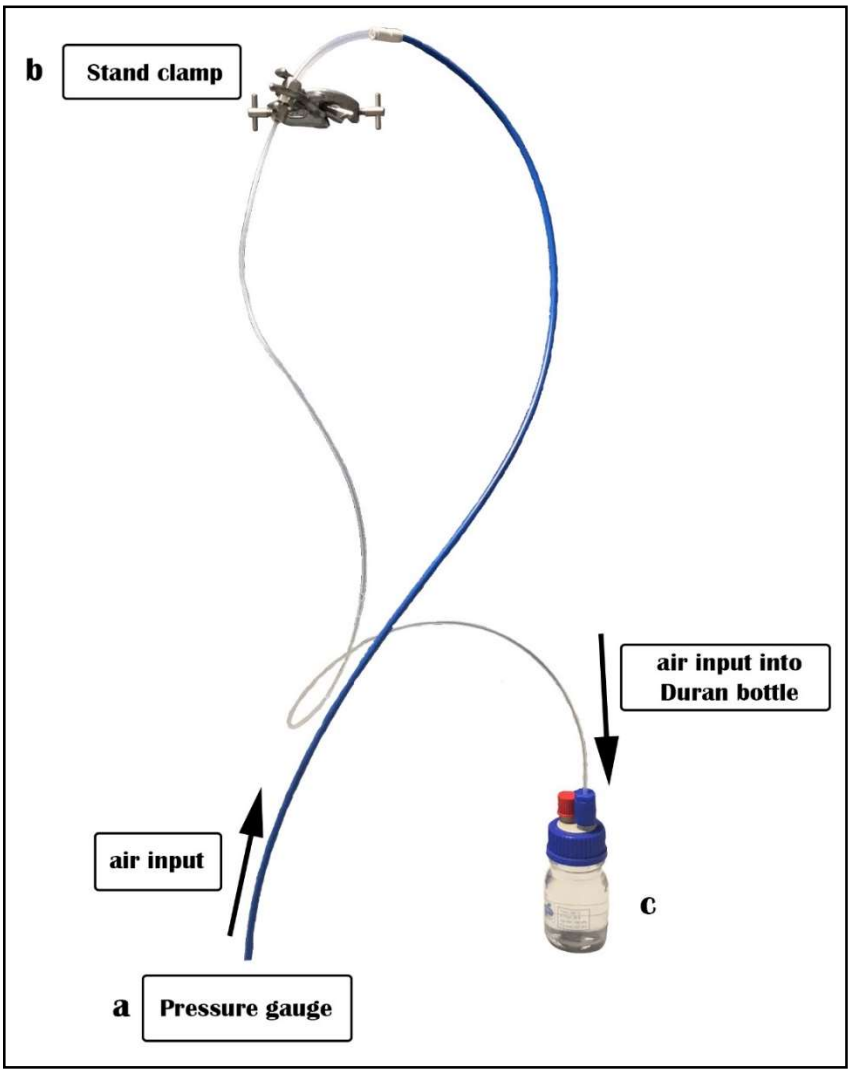

Figure 2-8. Bulk modulus apparatus – a: pressure gauge for a constant pressure input, b: stand clamp for keeping the tube in an upright position, c: Duran bottle which is the sample holder.

As the set-up was now a hydraulic system, applied pressure deforms the sample, and the amount of deformation was dependent on the sample. The air pressure was set to the constant level of 100 kPa, which was below the highest-pressure resistance of the bottle (150 kPa). When the sample is deformed by the pressure applied, the silicone oil indicates this displacement

and can be measured by a ruler. As the diameter of the tube was known, the volume of the oil displaced could be calculated using the tube area formula.

Bulk modulus was measured by using:

$$K = \frac{P}{\Delta V / V} \quad (2\text{-}19)$$

K is the bulk modulus, P is pressure, $\Delta V / V$ is volume change (Steffe, 1996).

Therefore, bulk modulus values can be compared to check how they change for varied sticky model foods.

It should be noted that the experimental set-up described above for the measurement of bulk modulus was not commercially available and was developed by the research team of the current thesis. The learnings from previous research studies were used to improve the design of the experimental set-up. Three replications were performed.

2.5 Statistical analysis

Different statistical analysis techniques were used in this thesis because of the type of data that was gathered.

Both one-way/one-factor analyses and multi-factor analyses of ANOVA were carried out, depending on the goals of the data studied. The Tukey HSD (Highest Significant Difference) post hoc comparison was also used to show significant differences between the data from TA, rheology, sensory evaluation and EMG. A p-value of 0.05 was used.

The categorization of assessors based on the percentage difference between their initial and final measurement points was conducted as follows: To determine the category, the criterion to select the last data point was set that there must be at least 2 data points within the time interval. The average values of the initial and final data points for each assessor were chosen, and the percentage difference was calculated using Microsoft Excel 2016. Assessors were then grouped into three categories: 0-30%, 31-60%, and 61-100%. Subsequently, a graph was generated using these categories.

In order to have a more accurate comparison among the assessors, the sensory time has been normalised as below:

$$Standardisation\ of\ each\ value = \frac{x - mean}{maximum - minimum} \quad (2\text{-}20)$$

x: value

The mean, maximum and minimum values belong to each individual.

Some of the obtained data was analysed using Pearson and Spearman correlations. Specifically, Spearman correlation was utilized to assess correlations within EMG data, owing to its non-linearity and non-parametric nature. Unlike Pearson correlation, Spearman correlation is based on the rank order of data and is not influenced by extreme values or outliers. However, Pearson correlation is more sensitive to small changes in data, particularly when the relationship between the variables is linear. Hence, Pearson correlation was employed for all other data correlations in the thesis.

The study utilized Cohen's d method to evaluate the capacity of EMG features to differentiate between model foods. This approach is crucial as the EMG methods adopt different units, rendering them challenging to compare using traditional statistical techniques. Measuring the effect size with Cohen's d method is particularly valuable in this regard. Cohen's d method was done using SPSS. Which is based on the mean and standard deviation of EMG data and calculation of their differences. After calculating the difference in means and obtaining the pooled standard deviation, the Cohen's d value was derived by dividing the former by the latter. This process aimed to quantify the effect size. Subsequently, the Cohen's d values were interpreted to determine the significance of the differences observed among the model foods, categorizing them into small, medium, large, very large, and huge effect sizes. This interpretation allowed for a more comprehensive understanding of the variations between the different model foods.

To facilitate a comprehensive analysis and interpretation of the data, a Principal Component Analysis (PCA) with a Direct Oblimin rotation was

employed on the complete dataset. This statistical technique allowed for the comparison of all the available data points, aiding in the extraction of meaningful patterns and relationships among variables. By applying the PCA with a Direct Oblimin rotation, the data were transformed into a set of new orthogonal variables, known as principal components, which captured the maximum amount of variance in the dataset. This approach enabled a more robust understanding of the underlying structure and interdependencies within the data.

The statistical analyses were conducted using IBM SPSS software version 28 (IBM Corp, USA) to perform various data analysis procedures. Linear regression and polynomial regression analyses, when relevant, were specifically conducted using Microsoft Excel 2016. These software tools provided the necessary functionality for implementing the regression models and generating the corresponding results.

3 Results and discussion

3.1 Sensory evaluation

This section focuses on the analysis and interpretation of data acquired through a discrete-time intensity method in the sensory evaluation of model foods. The utilization of this particular method was motivated by the fact that stickiness, being a dynamic texture attribute, undergoes changes throughout the oral processing phase, which cannot be fully captured through static sensory measurements alone. Therefore, the primary objective of this section is to evaluate and quantify the stickiness of the model foods at 5-second intervals during the mastication process, aiming to provide a comprehensive assessment of stickiness as a whole.

The selection of 5-second intervals stems from their identification as the minimum time required for subjects to effectively evaluate the stickiness of the model foods based on preliminary sensory testing. Additionally, this section delves into the initial research hypothesis, which explores the potential decrease in stickiness experienced by the model foods prior to swallowing. The examination of this hypothesis provides further insight into the behaviour of stickiness throughout the mastication process, contributing to a more thorough understanding of its dynamics.

Ensuring the accuracy, reliability, and consistency of collected data in sensory studies is paramount. Hence, conducting quality assessments on sensory data plays a vital role in detecting and rectifying any errors and inconsistencies that could compromise the validity of the findings. Neglecting to perform these quality checks can lead to misleading or erroneous conclusions. Næs et al. (2010) summarised the main quality checks for sensory data as mentioned in Table 3-1.

Table 3-1. Main quality checks for sensory data (Næs et al., 2010)

Parameter	**explanation**
Discrimination	Discriminate samples
Consensus among assessors	Ranking of the samples
Use of scale	How the samples were scored
Reliability of assessors	When performing replications

In the current section, these quality parameters will be checked for different assessors. The goal is to ensure meticulous scrutiny and evaluation of these parameters. There were significant effects of assessor and replicates as well as samples in the 3-factor ANOVA ($p<0.05$). The significant effect of assessor will be explored in the following sections to understand the source of the variation.

Discrimination of samples and consensus among assessors:

Table 3-2 exhibits the Tukey HSD post-hoc comparison, providing insights into the individual assessors' ability to distinguish model foods (The information in this table is organized vertically). It also displays the level of consensus intra- assessors in rating the samples.

The results obtained from the assessment indicate that all assessors unanimously rated model food 10-90 as the least sticky, while model food 65-120 received consistent ratings as the stickiest, with statistically significant differences observed. However, there were some variations. For instance, assessor a5 displayed variations in rating model foods 65-75 and 50-120, while assessor a10 exhibited differences in evaluating model foods 50-75 and 65-75. These instances highlight the presence of some discrepancies among assessors in their assessments.

While the majority of assessors tended to assign higher values to indicate stickiness for the model foods, it should be noted that a few assessors deviated from this trend. Specifically, assessors a5 and a10 rated the stickier model foods with lower values, such as 50-120. This discrepancy among assessors indicates variations in their perception and evaluation of

stickiness, highlighting the need for further analysis and understanding of individual differences within the assessment process. Furthermore, additional training on scale use or attribute understanding might also be needed.

Table 3-2. Tukey HSD post-hoc comparison of the ability of individual assessors to discriminate model foods (The information in this table is organized vertically). Assigned letters show the significant differences for each model food ($p < 0.05$). Values are given as mean (±SD) - each value was calculated based on the four replicates of each assessor at all time intervals.

Model foods	**Assessors**									
	a1	**a2**	**a3**	**a4**	**a5**	**a6**	**a7**	**a8**	**a9**	**a10**
10-90	0.4^{a} (0.3)	0.5^{a} (0.2)	0.5^{a} (0.3)	0.1^{a} (0.1)	0.4^{a} (0.2)	0.4^{a} (0.1)	0.3^{a} (0.2)	0.2^{a} (0.1)	0.2^{a} (0.1)	0.5^{a} (0.2)
35-90	2.1^{b} (0.4)	2.8^{b} (0.5)	$1.4^{a,b}$ (0.5)	2.7^{b} (1.1)	1.7^{b} (0.7)	1.5^{b} (0.7)	1.3^{a} (0.3)	1.4^{b} (0.4)	1.5^{a} (0.7)	3.0^{b} (1.1)
50-75	4.8^{c} (1.2)	3.7^{c} (1.2)	3.4^{c} (1.2)	7.1^{c} (1.1)	2.9^{b} (1.2)	4.2^{c} (1.3)	3.4^{b} (1.1)	3.6^{c} (0.7)	3.7^{b} (1.1)	$5.1^{c,d}$ (1.3)
50-120	7.6^{d} (1.3)	7.0^{d} (1.5)	7.8^{d} (1.4)	9.1^{d} (1.7)	4.9^{c} (1.7)	8.1^{d} (0.9)	7.7^{c} (1.4)	8.1^{e} (0.7)	8.6^{d} (2.2)	5.9^{d} (1.1)
65-75	5.1^{c} (1.5)	4.7^{c} (1.5)	7.6^{d} (1.1)	8.4^{c} (1.5)	5.3^{c} (1.3)	7.3^{d} (2.1)	4.9^{b} (2.3)	7.1^{d} (0.6)	6.0^{c} (2.1)	4.8^{c} (2.1)
65-120	8.5^{d} (0.4)	8.0^{e} (0.9)	8.1^{d} (1.2)	9.9^{e} (0.1)	8.2^{d} (0.7)	8.4^{d} (0.9)	8.6^{d} (1.1)	9.3^{f} (0.2)	9.4^{e} (0.2)	7.0^{e} (1.3)

The analysis of data presented in Table 3-2 revealed noteworthy observations regarding the assessors' ability to discern significant differences among the model foods. The majority of assessors successfully identified significant distinctions between model foods 10-90/35-90 and 50-120/65-120. However, comparatively fewer significant differences were observed for model foods 50-75 and 65-75. Furthermore, all assessors unanimously agreed that model foods 10-90 and 35-90 were significantly different from both 50-120 and 65-120. It is worth noting that the most significant differences of 10-90 and 35-90 were observed with model foods 65-75 and 50-75, respectively, indicating the variation in stickiness within these specific range. However, it should be noted that the current study encountered certain limitations in the sensory evaluation process. One significant constraint was the availability of trained assessors, which prevented the utilization of a fully trained panel and necessitating the involvement of naive assessors instead. Additionally, the available budget was limited. To ensure higher data quality and minimize potential errors in future studies, it is recommended to either employ trained panels or increase the number of naive assessors, ideally ranging from 50 to 150 participants. Previous research conducted by Ares et al. (2011) and Varela and Ares (2012) support the notion that similar results to those obtained with trained assessors can be achieved when a larger number of consumers are employed. In the current study, although the assessors did not receive extensive training, they did attend two training sessions. These sessions were designed to familiarize the assessors with scoring the reference samples. Therefore, it is important to consider the duration of the training, as a longer training period may lead to more accurate results in future studies.

In addition to the influence of assessor training, it is important to recognize that the observed variations among assessors could also stem from their unique chewing habits (Zimoch and Gullett, 1997). Individual differences in chewing behaviour, such as chewing force, duration, and technique, have been shown to impact sensory perception and evaluation. Therefore, it becomes crucial for future studies to address these limitations and account for these individual differences in chewing behaviour.

Use of scale:

Table 3-3 presents the Tukey HSD post-hoc comparison of assessors for each model food, highlighting their utilization of the scale. There was the least variation across the assessors for the less sticky model foods (10-90, 35-90, 50-75 and 65-75), and greater variation for the stickier model foods (50-120, 65-120).

Studies on sensory stickiness have shown that rating stickiness using a scale can result in higher variability for stickier model foods. This is attributed to the complex textures and rheological properties of sticky foods, which can affect how assessors perceive texture attributes. The intricate nature of these foods makes it challenging for assessors to provide consistent and precise ratings on a sensory scale, leading to increased variability in the results. Additionally, stickier model foods can possess stronger adhesive forces, influencing the perception of stickiness and contributing to greater variability in the ratings provided by assessors (Rodrigues et al., 2014, Wang and Hartel, 2021b).

Labbe et al. (2004) have discussed that consumers tend to employ a wider range of scales after receiving training and exposure to reference samples. In contrast, untrained assessors often utilize only a limited portion of the scale. Ares et al. (2011) reported that compared to trained assessors, consumers exhibit higher variability in scale utilization and tend to use a narrower range of the scale. This could be attributed to the assessors' confidence in utilizing the full spectrum of the scale or their ability to detect larger textural differences among samples. Both of these issues can be improved through extensive training (Romano et al., 2008). Therefore, it is recommended to use trained assessors when measuring complex textural attributes such as stickiness.

Table 3-3. Tukey HSD post-hoc comparison of assessors for each model food to enhance the understanding of the use of scale (post-hoc groupings should be read vertically). Assigned letters show the significant differences for each model food ($p < 0.05$) comparing all the assessors. Values are given as mean (±SD) - each value was calculated based on the four replicates of each assessor.

Assessors	Model foods					
	10-90	**35-90**	**50-75**	**50-120**	**65-75**	**65-120**
a1	$0.4^{a,b}$ (0.3)	$2.1^{a,b}$ (0.4)	$4.8^{a,b}$ (1.2)	$7.6^{b,c}$ (1.3)	5.1^{a} (1.5)	$8.5^{a,b,c}$ (0.4)
a2	$0.5^{a,b}$ (0.2)	$2.8^{a,b}$ (0.5)	3.7^{a} (1.2)	$7.0^{a,b}$ (1.5)	4.7^{a} (1.5)	$8.0^{a,b}$ (0.9)
a3	$0.5^{a,b}$ (0.3)	1.4^{a} (0.5)	3.4^{a} (1.2)	$7.8^{b,c}$ (1.4)	$7.6^{a,b}$ (1.1)	$8.1^{a,b}$ (1.2)
a4	0.1^{a} (0.1)	$2.7^{a,b}$ (1.1)	7.1^{b} (1.1)	9.1^{c} (1.7)	8.4^{b} (1.5)	9.9^{c} (0.1)
a5	$0.4^{a,b}$ (0.2)	$1.7^{a,b}$ (0.7)	2.9^{a} (1.2)	4.9^{a} (1.7)	5.3^{a} (1.3)	$8.2^{a,b}$ (0.6)
a6	$0.4^{a,b}$ (0.1)	$1.6^{a,b}$ (0.7)	4.2^{a} (1.3)	$8.1^{b,c}$ (0.9)	$7.3^{a,b}$ (2.1)	$8.4^{a,b,c}$ (0.9)
a7	$0.3^{a,b}$ (0.2)	1.3^{a} (0.3)	3.4^{a} (1.1)	$7.7^{b,c}$ (1.4)	4.9^{a} (2.3)	$8.6^{a,b,c}$ (1.1)
a8	$0.2^{a,b}$ (0.1)	1.4^{a} (0.4)	3.6^{a} (0.7)	$8.1^{b,c}$ (0.7)	$7.1^{a,b}$ (0.7)	$9.3^{b,c}$ (0.2)
a9	0.2^{a} (0.1)	$1.5^{a,b}$ (0.7)	3.7^{a} (1.1)	8.6^{c} (2.2)	$6.1^{a,b}$ (2.1)	$9.4^{b,c}$ (0.2)
a10	$0.5^{a,b}$ (0.2)	3.0^{b} (1.1)	$5.1^{a,b}$ (1.3)	$5.9^{a,b}$ (1.1)	4.8^{a} (2.1)	6.9^{a} (1.3)

Reliability of assessors:

Table 3-4 presents the Tukey HSD post-hoc comparison of different sensory replications for individual assessors, providing insights into the consistency, reliability, and differences in ratings across replications. The table allows to examine how individual assessors' ratings vary within their own assessments. Upon analysing the table, it is evident that the majority of assessors do not exhibit significant differences between their replications. This suggests a consistency and reliability in their ratings. However, two assessors, namely a1 and a10, show significant variations across their replications, indicating some degree of inconsistency in their assessments.

There are several possible explanations for the significant differences in assessors' performance between the different replications. As the assessors in the current study were inexperienced, it is likely that they would benefit from additional training sessions to improve the consistency of their stickiness evaluations. The disparities in the sensory acuity or sensitivity of the assessors may also be a factor in the variances in their evaluations. These individual variations can influence how assessors perceive and assess the sensory attributes of the food products, leading to discrepancies in their ratings or responses across the replications (Lawless and Heymann, 2010). Using multiple assessors in sensory evaluations is crucial to ensure reliable and valid results.

Table 3-4. Tukey HSD post-hoc comparison of different sensory replications for individual assessors. Assigned letters show the significant differences for each model food ($p < 0.05$). Values are given as mean (±SD) - each value was calculated based on the four replicates of each assessor.

Replications	**Assessors**									
	a1	**a2**	**a3**	**a4**	**a5**	**a6**	**a7**	**a8**	**a9**	**a10**
Replication1	3.8^{a} (3.0)	5.0^{a} (2.5)	4.6^{a} (3.7)	6.4^{a} (3.1)	4.2^{a} (3.2)	5.8^{a} (3.6)	3.1^{a} (2.8)	5.4^{a} (3.8)	6.5^{a} (3.7)	$5.2^{a,b}$ (2.5)
Replication2	5.7^{b} (3.5)	4.1^{a} (2.5)	4.9^{a} (3.8)	7.0^{a} (3.4)	3.9^{a} (2.7)	4.7^{a} (3.6)	4.1^{a} (3.8)	5.0^{a} (3.5)	5.2^{a} (3.2)	5.6^{b} (3.5)
Replication3	$4.8^{a,b}$ (3.1)	4.3^{a} (2.6)	4.0^{a} (3.0)	6.0^{a} (3.7)	3.1^{a} (3.2)	4.3^{a} (3.0)	5.1^{a} (3.4)	5.0^{a} (3.2)	5.4^{a} (3.4)	$3.7^{a,b}$ (2.7)
Replication4	$4.6^{a,b}$ (3.3)	4.5^{a} (3.8)	5.7^{a} (3.3)	6.6^{a} (3.2)	4.4^{a} (2.7)	5.4^{a} (3.6)	5.1^{a} (3.9)	4.5^{a} (3.7)	5.5^{a} (3.6)	2.9^{a} (2.3)

While most of the assessors do not show significant differences between replications, they show significant differences in the use of scale as well as to discriminate the model foods. Despite the significant differences in utilization of scale by assessors, the fact that they arrive at similar evaluations indicates a level of reliability in their judgments. This consistency suggests that the assessors have a shared understanding of the stickiness definition and the evaluation methodology.

Table 3-5 shows the Tukey HSD post-hoc comparison of the ratings of individual assessors at different time intervals (significant differences are shown by letters). It can be seen that there are no significant differences for individual assessors at the first 3 time intervals of 5 s, 10 s and 15 s ($p < 0.05$). However, at the time intervals of 20 s and 25 s, some of the assessors differ significantly from the others.

The table also compares the measurement of all the assessors at different time intervals (significant differences are shown by numbers). The intensity of significant differences between assessors can be seen to be somewhat weaker at the 5 s, 10 s, and 15 s time intervals, but to grow more evident at the last two intervals.

Table 3-5. Tukey HSD post-hoc comparison of assessors' ratings at different time intervals (each column of data) are indicated by letters ($p < 0.05$). On the other hand, stickiness ratings of individual assessors (each row of data) at different time intervals are indicated by different numbers ($p < 0.05$).

Assessors	Time intervals				
	5 s	10 s	15 s	20 s	25 s
a1	$5.3^{a,1}$	$5.3^{a,1}$	$5.7^{a,b,c,1}$	$5.4^{a,b,c,d,1}$	$5.2^{b,c,1}$
a2	$4.5^{a,1}$	$4.7^{a,1}$	$5^{a,b,1}$	$5.1^{a,b,c,d,1}$	$5.2^{b,c,1}$
a3	$7.2^{b,c,d,1}$	$7.9^{c,1}$	$7.9^{d,1}$	$7.5^{e,f,1}$	$7.3^{d,e,1}$
a4	$8.2^{d,1}$	$8.0^{c,1}$	$8.1^{d,1}$	$8.0^{f,1}$	$8.0^{e,1}$
a5	$6.3^{a,b,c,1}$	$5.7^{a,b,1}$	$4.9^{a,b,1}$	$3.6^{a,2}$	$2.5^{a,2}$
a6	$8^{c,d,1}$	$7.4^{b,c,1}$	$7.2^{c,d,1}$	$6.6^{d,e,f,2}$	$5.9^{c,d,2}$
a7	$4.6^{a,1}$	$4.9^{a,1}$	$4.7^{a,1}$	$4.6^{a,b,c,1}$	$4.7^{b,c,1}$
a8	$5.6^{a,b,1}$	$5.8^{a,b,1}$	$5.8^{a,b,c,1}$	$5.8^{b,c,d,1}$	$5.7^{b,c,d,1}$
a9	$7.5^{c,d,1}$	$7.2^{b,c,1}$	$6.7^{b,c,d,1}$	$6.4^{c,d,e,f,1}$	$5.9^{c,d,2}$
a10	$4.7^{a,1}$	$4.6^{a,1}$	$4.4^{a,1}$	$4.2^{a,b,1}$	$4.0^{a,b,1}$

The continuous trends of individual assessors as well as the variances among other assessors are revealed by the patterns in the assessors' stickiness ratings. The progressive rise in ratings over the course of the time interval suggests that stickiness has been considered to increase. Additionally, the discrepancies seen across assessors and within a single assessor over time show that each person perceives stickiness differently. Varied people may have varied oral impressions of stickiness, according to the changes in evaluations between assessors at particular times. Some assessors might be more sensitive to stickiness, which would cause them to give it a higher rating even at shorter intervals. However, even if the sample is thought to be sticky by others, assessors who are less sensitive to stickiness may give the sample a lower value. The variances in the assessors' ratings may be caused by these individual variations in oral perception. Additionally, the variations in the same assessor's ratings made at different times allude to a changing oral perception of stickiness. As the exposure time increases, the sample's interactions with the oral environment and saliva may alter, altering how stickiness is perceived. As a result, ratings for shorter and longer time periods may differ. As they continue to evaluate the sample, assessors may believe the stickiness to increase or decrease, leading to noticeably different evaluations.

To investigate the variations among assessors at each time interval, a profile table comparing individual model foods and assessors was employed (appendix 6.4). It revealed an interesting inverse relationship between the total oral processing time and overall stickiness, particularly for model food 65-120, among certain assessors. (See appendix 6.4). The overall stickiness generally tends to increase with higher values of total chewing time for some assessors and model foods. However, the relationship between total chewing time and overall stickiness varies across different assessors and model foods, as indicated by the post hoc groupings. It is important to consider the specific assessor and model food combination when analysing the data and drawing conclusions. The findings of this study align with the research conducted by Wagoner et al. (2016), where it was observed that increased levels of stickiness led to longer oral processing times. Wagoner et

al. (2016) noted higher stickiness in caramels that contained corn syrup, compared to those where agar and gelatin served as replacements. Similarly, Çakir et al. (2012) reported similar results for caramel samples with higher proportions of sugar and corn syrup, indicating a positive correlation between stickiness and oral processing time in these studies.

Table 3-6 illustrates the significant levels of overall stickiness for all the assessors. In this table, the letters assigned to each value indicate the significant differences between the model foods ($p < 0.05$). The overall stickiness is a useful value to compare the perception of stickiness in different model foods.

Table 3-6. Perception of overall stickiness by assessors. Each data point is the average of all data points collected for each model food. It can be seen that all model foods are significantly different ($p < 0.05$). assigned superscript letters indicate significant differences among model foods.

Sample	Significant levels of overall stickiness
10-90	0.2^{a} (±0.3)
35-90	2.3^{b} (±1.3)
50-75	4.8^{c} (±1.7)
65-75	6.5^{d} (±2.2)
50-120	7.9^{e} (±1.6)
65-120	9.0^{f} (±1.2)

Table 3-6 shows that the overall stickiness of all the model foods differs significantly from each other. Model foods with the lowest sugar and highest water content (e.g., 10-90) have the lowest stickiness, while model foods with high sugar and low moisture content have the highest stickiness (e.g., 65-120). It should be also mentioned that by increasing the heating time, the stickiness is perceived more strongly by the assessors. The tendency of high sugar confectionery (especially disaccharides) to stickiness levels has been highlighted in the literature (Adhikari et al., 2001, Wang and Hartel, 2021b). Disaccharides can affect stickiness via the glass transition temperature and dextrose equivalent, with a higher amount of small-molecule sugars leading to higher DE and a decrease in Tg leading to increased stickiness (Fan and Roos, 2017, Burke and Hartel, 2021). The amount of sugar and water in

model foods can lead to different rheological behaviours. A product with a higher sugar content could have a more deformable texture. An interesting observation on the deformability of caramel samples was highlighted by Mayhew et al. (2018) where the more deformable samples were positively correlated with stickiness measurement of stickiness, while it was the opposite for the fragile samples. The same observation was also made by Wang and Hartel (2021b) that the more deformable foods had a higher degree of stickiness. While the current study did not directly measure the relationship between deformability and stickiness, it was observed that the model foods with higher stickiness exhibited visual cohesiveness and deformability. On the other hand, the model foods with lower stickiness, particularly 10-90, displayed a visually fragile gel-like structure. Assessors also reported that the stickier samples underwent more deformation compared to the samples with lower stickiness during sensory evaluation sessions.

Since stickiness is a textural feature that changes significantly during oral processing, it is important to study the evolution of stickiness during chewing. To this end, a discrete time intensity study was conducted, and stickiness ratings were collected at 5 second time intervals. The results are shown in Table 3-7 and are reported as mean (±SD). The results are categorised by time interval for each model food. In order to enhance comprehension regarding the quantity of active assessors (those still chewing), a percentage of data points for individual values is also provided. The percentage is based on a maximum of ten assessors with four replicates. For example, if 50% is given, it means that 20 points (out of possible 40) were collected for that model food in that time interval. The values with the same letters in each time interval are not significantly different from each other. Each value is the average of a maximum of ten assessors with four replications.

Table 3-7 shows that 65-120 was rated as the stickiest sample in all time intervals, while sample 10-90 was perceived as the least sticky. The second stickiest model food is 50-120, followed by 65-75. In addition, comparison of the stickiness of the model foods shows a gradual decrease from 65-120 to

50-75, followed by a large gap in sensory rating between samples 50-75 with 35-90 and 10-90.

Table 3-7. Time intervals and significant differences ($p < 0.05$) between model foods. Values are given as mean (±SD) - each value was calculated based on the number of assessors (maximum 10) with four replicates. In each time interval (each column), the same letters mean no significant differences. On the other hand, the significant differences for each model food over time (each row) are indicated by different numbers. The percentage indicator in each cell is the number of subjects still chewing at that point in time.

	Model foods	Time intervals (s)				
		5	10	15	20	25
Low stickiness	**10-90**	$0.4^{a,1}$ (±0.4) 93%	$0.3^{a,1}$ (±0.3) 85%	$0.2^{a,1}$ (±0.3) 68%	$0.2^{a,1}$ (±0.3) 35%	$0.1^{a,2}$ (±0.2) 18%
	35-90	$2.1^{b,1}$ (±1.3) 100%	$2.0^{b,1}$ (±1.1) 100%	$2.0^{b,1}$ (±1.1) 98%	$1.7^{b,1}$ (±1.2) 90%	$1.0^{a,b,1}$ (±1.3) 55%
	50-75	$4.7^{c,1}$ (±1.9) 100%	$4.4^{c,1}$ (±1.8) 100%	$4.1^{c,1}$ (±1.9) 100%	$3.7^{c,1}$ (±2.1) 93%	$2.5^{b,c,1}$ (±2.4) 68%
	65-75	$6.5^{d,1}$ (±2.1) 100%	$6.5^{d,1}$ (±2.3) 100%	$6.2^{d,1}$ (±2.3) 100%	$5.2^{d,1}$ (±2.8) 90%	$3.8^{c,d,2}$ (±3.4) 68%
	50-120	$7.8^{e,1}$ (±1.6) 100%	$7.7^{e,1}$ (±1.7) 100%	$7.6^{e,1}$ (±1.8) 100%	$7.0^{e,1}$ (±2.4) 98%	$5.2^{d,e,2}$ (±3.6) 78%
High stickiness	**65-120**	$8.8^{e,1}$ (±1.4) 100%	$8.8^{f,1}$ (±1.3) 100%	$8.6^{e,1}$ (±1.3) 100%	$8.1^{e,1}$ (±2.2) 95%	$7.2^{e,1}$ (±3) 88%

Table 3-7 shows that at time intervals of 5, 15 and 20 seconds, all model foods are significantly different, except for 65-120 and 50-120, which are rated as the stickiest samples. The only time point at which all model foods are significantly different is at 10 seconds. A possible explanation for this is the effect of saliva hydrating and diluting the model foods and the mechanical breakdown of the foods during chewing (Janssen et al., 2007, Foegeding et al., 2015, Young et al., 2016). It was also emphasized by Chen (2020) that the material properties of a food product alone cannot fully account for its characteristics, and that oral processing and sensory perception parameters represent complex mechanisms that transform food materials into a swallowable bolus. Therefore, it is important to consider both material and sensory properties when characterizing food products.

It is also conceivable that this structural manipulation of food samples may be associated with a greater number of oral mechanoreceptors, resulting in a more complex sensory signal (Foegeding et al., 2015). As mentioned by Young et al. (2016), continuously increasing the moisture content of the bolus results in a low degree of stickiness, but the optimal amount of moisture may have a reinforcing effect on stickiness. Although the saliva content of the model food was not analysed in the current study, it can be assumed that increasing the moisture content of the model foods due to mixing with saliva together with mechanical manipulation in the time interval of 10 s leads to an increase in stickiness. This increase in stickiness can make it easier for assessors to discriminate between different samples. With further chewing, the moisture content of the bolus increases, and the stickiness values decrease with increasing oral processing time. Although the moisture content of the boli was not measured in the current study, this is consistent with Hawthornthwaite et al. (2015) statement that, following Hutchings and Lillford's oral breakdown pathway, the sample reaches the point of swallowing where salivary secretion reduces the perception of cohesive sticky sensation. Although the time of chewing is short, saliva has a further effect on starch structure by breaking it through the action of salivary amylase (Mosca and Chen, 2017). It has been suggested that starch breakdown leads to more necking or long behaviour, increasing the value of stickiness (Dunnewind et al., 2004, Janssen et al., 2007). It should be noted that there are other parameters (e.g., the enzymatic activity of bolus during mastication) that influence the properties of the bolus and its texture perception, which are outside the scope of this thesis.

Table 3-7 also shows that the percentage of assessors who are still chewing the stickier samples is higher than for the low sticky model foods. This suggests that the stickier model foods require a longer oral processing time among assessors compared to the low stickiness model foods. In line with the present results, Kilcast and Roberts (1998) have shown that the sugar content of 44.7% serves as a threshold for oral perception of stickiness. This could be the reason why perceived stickiness decreased dramatically for 35-90 and 10-90, which are below the 44.7% threshold. Kilcast and Roberts

(1998) also reported that low water content and high soluble solids content are important parameters for higher stickiness. Table 2-2 shows that for most model foods there is a relationship between water content and overall stickiness, with the lower the water content, the higher the stickiness of the model foods. Furthermore, by considering the water content after cooking mentioned in Table 2-2 the order of the model foods from low to high stickiness is 10-90, 35-90, 50-75, 50-120, 65-75 and 65-120, respectively. It can be seen that the overall stickiness of all model foods follows their water content, with the exception of 50-120 and 65-75, the former being perceived as stickier.

It was suggested by Hollowood (2018) that the use of generic terms (in the total impression method) would reduce the impact of individual differences in the perception of similar terms. The total impression method involves assessors providing a holistic evaluation of a sample based on their overall impression, rather than analysing individual attributes separately which might provide a more realistic evaluation of the samples which is closer to real life situations. Considering this, it was aimed in the current study to use a combination of terms to make a comprehensive definition of stickiness in order to cover its complexity and the total oral processing time of sticky model foods. Although the idea of using multiple terms was inspired from the total impression method, the chosen terms aimed to provide a more precise and accurate definition of stickiness, distinct from the holistic approach adopted by the total impression method.

In the current study 5 terms have been used to define stickiness (Table 2-3). These terms were developed by Mayhew et al. (2018) in a study on perceptions of stickiness. These 5 terms were selected from a number of other terms and were found to have a strong correlation with sensory perception of stickiness. As each of the terms was able to describe a part of the stickiness during oral processing and none of them had the ability to describe the stickiness throughout the mastication, it was decided in the current research to make a comprehensive definition of stickiness using these terms. Two terms of toothpacking and enveloping describe the degree of cohesive failure and the amount of sample remaining on the oral surface

(Wang and Hartel, 2021a), and are closely related to cohesiveness. Although the term cohesiveness is closely related to stickiness, it is unable to draw a thorough understanding of stickiness. As stickiness is a time-dependent attribute (Wang and Hartel, 2021a), it is also crucial to consider the terms that may define stickiness during oral processing. For example, stringiness has been reported to be a parameter that defines stickiness at early stages of oral processing (Mayhew et al., 2017) which impose some limitations to applicability of this parameter when considering the total mastication time. It should also be considered that the term stringiness as well as tacky can greatly differ depending on the components of the foods in question. For example, a food containing higher amounts of long chain molecules (such as amylose in starch) will demonstrate increased values for stringy as a viscoelastic property while tacky would be seen predominately as a surface characteristic of the food material (such as food powders). The model foods of the current study possessed both stringy and tacky properties depending on their recipes. These two terms are also affected by the amount of shear during mastication, meaning the higher shear can lead to more tacky perception while the opposite applies to stringy and the long texture perception (Noren et al., 2019). During the training sessions, assessors were provided with reference samples that demonstrated each of the aforementioned terms. They engaged in discussions about their understanding of the definitions in relation to the reference samples. Whenever confusion arose, additional time was allocated to specific assessors for further clarification. The training process commenced by evaluating each specific term and gradually progressed to the assessment of their combinations.

It can be seen in Table 3-2 that while the variances in the ratings assigned to the model foods were not consistently statistically significant, the remarkable aspect lay in the uniformity observed among the assessors in their perception of stickiness. A similar trend was also observed for the replications in which the consistency of assessors (Table 3-4) in evaluating model foods across multiple replications remains non-significant (for most of

the assessors). These results might suggest that the assessors had a strong understanding of the stickiness definition.

Despite above discussions and the benefits, using five terms may introduced variability to the data. It is also possible that the assessors had some difficulty in using the terms equally. This scenario appears to be particularly true for certain assessors who exhibited variations in their measurement. In order to reduce possible confusions in the measurement of stickiness during the sensory evaluations, the terms have been reviewed with the assessors just before the sensory session to ensure that they have a fresh memory of stickiness definition. Moreover, significant differences were observed among assessors in their utilization of the scale. While the factors contributing to these variations may vary, it is important to acknowledge that a deficient understanding of the terms may contribute to the divergence in scale usage. Given the inherent complexity of assessing stickiness as a textural attribute, the utilization of less simplified definitions has the potential to further compound the challenges associated with its evaluation by assessors.

The discriminatory capacity of some assessors may be diminished by the use of multiple terms, particularly for samples with few variations in their recipes or levels of stickiness. The degree of stickiness of a few of the model foods in the current investigation is similar. For example, model foods 10-90/35-90, 50-75/65-75 and 50-120/65-120 showed to have less significant differences. It is evident in Table 3-3 that the minimum significant differences are among the above pairs of model foods. Although these differences can be related to other parameters such as the level of the experience of the assessors, the use of multiple terms can also be responsible.

Several methods can be put into place to reduce variability and improve consistency among assessors when employing a thorough definition for stickiness. Utilizing trained assessors is one such measure, since research indicates that trained panels show higher attentiveness and concentrate on the prescribed phrases than less qualified assessors. This strategy ensures more accurate and consistent assessments of stickiness (Ares et al., 2011). Moreover, it has been proposed that providing enhanced training to

assessors can contribute to a more solid comprehension of more intricate definitions. Additionally, employing standardized measurement protocols can help mitigate any potential challenges associated with using comprehensive definitions (Tomic et al., 2013). Additionally, it is advised that when using more complicated definitions, the integration of sensory and instrumental measures might improve the results of sensory research. Instrumental and sensory measures can collaborate effectively if precise, quantitative definitions are established. Accordingly, the use of EMG has been suggested as an extra technique to supplement sensory measurement.

The total impression approach can help with product development, quality assurance, and marketing decisions if used appropriately since it offers a meaningful evaluation of how the product is viewed as a whole by customers or experts. While distinct oral processing steps that affect stickiness are taken into account in this situation, stickiness can be assessed more precisely.

The normalised stickiness ratings for the model foods are presented in Table 3-8, with each assessor's ratings normalised for their respective oral processing times using the normalisation method described in section 2.5. Notably, the normalised ratings maintain the same order as the non-normalised data, with 10-90 and 65-120 being the least and most sticky samples, respectively.

Table 3-8. Normalised-time and significant differences between model foods ($p < 0.05$). Each value is the average normalised data of all assessors. In each time interval (column), the same letters mean no significant differences. On the other hand, the significant differences for each model food over time (each row) are indicated by different numbers. Time point 0 is the start of chewing and time point 1 is the last rating time of stickiness before swallowing.

	Model foods	**Normalised-time**											
		0.17	**0.20**	**0.25**	**0.33**	**0.40**	**0.50**	**0.60**	**0.67**	**0.75**	**0.80**	**0.83**	**1.00**
Low stickiness	**10-90**	$0.7^{a,1}$	$0.4^{a,2}$	$0.5^{a,2}$	$0.5^{a,2}$	$0.4^{a,2}$	$0.4^{a,2}$	$0.3^{a,2}$	$0.4^{a,2}$	$0.3^{a,2}$	$0.4^{a,2}$	$0.3^{a,2}$	$0.3^{a,2}$
	35-90	$2.3^{b,1}$	$2.2^{b,1}$	$2.0^{b,1}$	$2.1^{b,1}$	$1.9^{b,1}$	$2.0^{b,1}$	$2.0^{b,1}$	$2.2^{b,1}$	$1.9^{b,1}$	$1.8^{b,1}$	$2.1^{b,1}$	$1.6^{b,1}$
	50-75	$5.0^{c,1}$	$4.8^{c,1}$	$4.3^{c,1}$	$4.5^{c,1}$	$4.4^{c,1}$	$4.1^{c,1}$	$4.1^{c,1}$	$3.9^{b,1}$	$3.8^{b,1}$	$4.0^{c,1}$	$3.5^{b,1}$	$3.5^{c,1}$
	65-75	$7.1^{d,1}$	$6.7^{d,1}$	$5.7^{d,1}$	$6.9^{d,1}$	$6.5^{d,1}$	$6.1^{d,1}$	$6.6^{d,1}$	$6.3^{c,1}$	$5.1^{c,2}$	$6.0^{d,1}$	$5.6^{c,1}$	$5.1^{d,2}$
	50-120	$8.5^{e,1}$	$6.9^{e,2}$	$8.2^{e,1}$	$8.2^{e,1}$	$7.0^{d,1}$	$8.1^{e,1}$	$6.9^{d,2}$	$7.5^{d,1}$	$8.0^{d,1}$	$6.4^{d,2}$	$7.1^{d,1}$	$6.7^{e,2}$
High stickiness	**65-120**	$9.1^{e,1}$	$8.6^{f,1}$	$8.4^{e,1}$	$8.9^{e,1}$	$8.4^{e,1}$	$8.9^{f,1}$	$8.3^{e,1}$	$8.6^{e,1}$	$8.9^{e,1}$	$8.3^{e,1}$	$8.3^{e,1}$	$8.0^{f,1}$

Table 3-8 indicates that the normalised stickiness levels of the samples display some fluctuations in their scores, which did not alter the order of the model foods. These fluctuations could be attributed to variations in the assessors' interpretation and utilization of the scale, as shown in Table 3-3, where some assessors displayed significant differences in their use of the scale. Additionally, the observed fluctuations may also be influenced by the effect of samples in different replications. Despite significant differences being observed only for a1 and a10, there were some non-significant differences within replications that could contribute to the fluctuations in the normalised data. Table 3-8 reveals that there are significant differences in the stickiness of some of the model foods, particularly towards the end of the chewing process. At time point 1, model foods 65-75 and 50-120 exhibit significant differences, while model food 10-90 displays significant differences from the initial stages of chewing. The remaining model foods do not display any significant changes throughout the chewing process. It can also be seen that normalisation reduced the magnitude of differences between 65-120, 50-120 and 65-75 compared to non-normalised values. This could be due to the fact that normalisation transforms the data into its realistic form and reduces the variance between the different time scales (Stone et al., 2012). The process of normalisation might have some influence on sensory and consumer data but given that variables are typically measured on a similar scale, the effect of normalisation is usually moderate in terms of the conclusions derived from the study. While the visual representations may display slight variations, the overall findings are often comparable (Næs et al., 2010).

When normalising sensory data, the possible disadvantages and limitations of normalisation should also be considered. Normalisation might lead to the loss of information, increasing the complexity of data analysis and decreasing the sensitivity of sensory data by removing the variability among assessors (Kemp et al., 2009, Lawless and Heymann, 2010). In order to acquire a more thorough understanding of the potential discrepancies between normalized and non-normalized data, these restrictions have been taken into account in the current study by assessing the normalised data

using the quality checks. A comparison between the non-normalised results shown in Table 3-7 and the normalised results displayed in Table 3-8 reveals that the significant differences between time intervals for each model food are almost the same for non-normalised and normalised data. The small differences can be attributed to the elimination of the confounding effect of assessors who have already completed their chewing at the latest stages in the non-normalised data which is in fact the ability of time normalisation in reducing the variation within the data set. Both non-normalised and normalised data consistently show a lack of significant differences over time for each type of food model, with a few exceptions. This suggests that the perception of stickiness does not undergo noticeable changes for most model foods, and this lack of change may not be attributed to other factors.

Further analysis of the data reveals the extent of the reduction in stickiness for each assessor compared their first and last measurement points. Comparing the data from Table 3-7 to the normalised data in Table 3-8 shows that both non-normalised and normalised data have similar trends at different time intervals. Comparing the first and last time intervals, it can be seen that there are significant differences for model foods 10-90, 65-75 and 50-120. While there may not be significant differences among other model foods, a progression can be observed from low-sticky model foods to high-sticky model foods. These results contradict one of the initial hypotheses of the study that the stickiness of the model foods undergoes a certain and substantial decrease before swallowing. This decrease in stickiness was suggested to be the trigger for swallowing of different types of cereal products. It was discussed that the oral manipulation of the bolus increases the stickiness before swallowing, which named the stickiness as a trigger for swallowing (Adhikari et al., 2001, Loret et al., 2011, Peyron et al., 2011).

To understand the degree of agreement between the assessors, they are divided into subgroups with significant differences (Figure 3-1). Each subgroup shows the number of significant groups ($p < 0.05$) in each time interval. It can be seen that there were some differences between the assessors at 5 s (first rating point), which divided them into 5 different subgroups. When chewing continued (at 10 and 15 second intervals),

significant differences between assessors reduced to 3 subgroups and only 2 assessors rated the samples significantly different from the rest of the panel. In such cases, it is suggested that these assessors should be trained more to ensure accuracy of rating (Tomic et al., 2013).

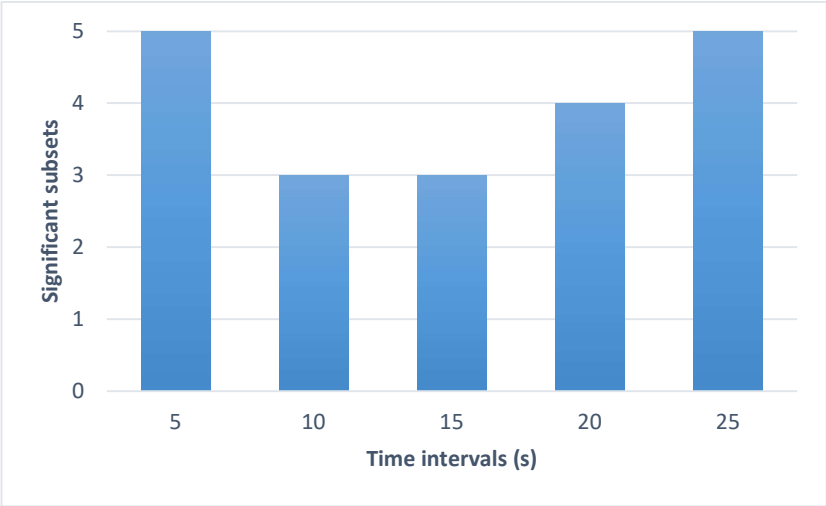

Figure 3-1. The significant differences between each assessor and the other assessors on stickiness perception ratings over time ($p < 0.05$). Assessors are grouped into subgroups (each bar) with significant differences. The figure shows that the number of subgroups is lowest at 10 and 15 seconds, while it is highest at the 5 and 25 seconds time intervals.

Stickiness is a complex and dynamic sensory property that can develop during chewing as a result of changes in the texture and viscosity of the food (Yang et al., 2018). Although the texture of model foods in the current study was not instrumentally measured at different stages of chewing, it has been highlighted in the literature that as the food is broken down at the beginning of chewing, the food undergoes a higher level of mechanical stress compared to later stages of the chewing which might lead to more distinguishable textural perceptions by the assessors at the beginning of the chewing (Pu et al., 2021). Existing literature has also highlighted the influence of individual differences as a factor contributing to the higher significant differences at the initial stages of sensory measurement (Parente et al., 2010). As chewing progresses towards the middle stages of oral processing, the food material tends to become more uniform, resulting in texture perception becoming dominant over the initial characteristics of the

food, as perceived by assessors (Devezeaux de Lavergne et al., 2017). Another potential explanation is that assessors may require an initial period of adjustment at the beginning of chewing to acclimate themselves to the sensory stimuli and develop a greater familiarity with the specific product under evaluation at middle stages of mastication process (Poveromo and Hopfer, 2019). As assessors gain a deeper familiarity with the sensory characteristics of the evaluated product, their evaluations tend to exhibit increased consistency, resulting in a reduction in the occurrence of significant differences. It should also be mentioned that the significant differences among assessors in the use of scale is another important parameter that increases the number of subsets at different time intervals.

In the final time intervals (20s and 25s), there were higher variations among the assessors. This could be attributed to the fact that some of the assessors had already swallowed the model foods. At 25s and particularly for 10-90 and 35-90 which resulted to a reduction of 80% and 45% in the number of assessors who were still chewing (Table 3-7). These reductions in the number of active assessors can also be seen for the other model foods with the maximum level of 32% for both 50-75 and 65-75.

While the limited number of assessors could contribute to increased significant differences, it is not the sole determining factor. The significance of saliva in oral processing is paramount. Although the current study did not investigate saliva's role, its influence on stickiness perception should not be overlooked. Saliva serves various functions that can impact stickiness during oral processing. Its lubricating properties potentially reduce stickiness as mastication progresses (Hawthornthwaite et al., 2015). In addition to lubrication, saliva contains enzymes like amylase, which can break down sugar and starches, the primary constituents of the model foods in this study. This enzymatic action may diminish stickiness at the later stages of mastication (Janssen et al., 2007). Saliva secretion varies among individuals, which may introduce additional variability to sensory data (Nishinari et al., 2019).

Table 3-9 provides insightful data regarding the chewing time of individual assessors. Within the panel, there exists a range of chewing speeds, with some assessors being categorized as slow chewers while others as fast chewers. The table highlights the significant differences in oral processing times among assessors, which directly impact the duration of chewing required before swallowing.

Table 3-9. Chew Time of assessors based on EMG data. Using this data, assessors can be categorized into slow and fast chewers ($p < 0.05$).

Assessors	ChT
a7	8.28^{a}
a3	8.43^{a}
a1	$10.69^{a,b}$
a4	11.30^{b}
a10	$11.34^{b,c}$
a2	$11.73^{b,c}$
a9	$12.59^{b,c}$
a8	$12.65^{b,c}$
a5	14.11^{c}
a6	16.66^{c}

In a study conducted by Devezeaux de Lavergne et al. (2015a), the impact of mastication time on the perception of stickiness in soft and hard texture sausages was investigated. The findings revealed that assessors with varying eating durations had distinct perceptions of stickiness. Initially, at the onset of chewing, the assessors perceived soft and hard sausages similarly. However, as the chewing process progressed, their perceptions diverged. Short-term assessors perceived soft sausages as stickier compared to long-term assessors, who described a grainy sensation. Additionally, long-duration assessors reported a more pronounced after-feeling of residue compared to their short-duration counterparts. Notably, the bolus particle size was larger among short-term assessors than long-term assessors. The study concluded that the differing chewing behaviours of the two assessor groups might lead to different triggers for swallowing. These findings shed

light on the complex relationship between mastication time, sensory perception, and swallowing behaviour in the context of stickiness evaluation.

The profile table in appendix 6.4 shows that, with the exception of a few instances, such as 10-90 and 65-120, there are no significant differences seen for any of the model foods at either the 5s or 10s time intervals. These results imply that the substantial variations in assessments for each model food (appendix 6.4) cannot solely account for the significant differences observed at 5s and 10s in Table 3-5. There are more significant variations at the 15s time period than at the 5s and 10s intervals, according to the comparison of individual assessors for each model food (appendix 6.4). Particularly, variations between 10-90, 50-75, and 50-120 are found to be significant. The assessors show significant changes for all of the model foods in the last two time intervals, with the exception of the 35-90 and 65-75. These significant discrepancies between the model foods could be caused by a number of factors. The use of the scale may be one of the causes. When assigning scores on the scale, the assessors may have different interpretations and biases, which causes variations in their assessments. The variations between the model foods that have been observed may be caused by this variation in scale usage. The amount of time that the assessors spend processing oral input is another element that could affect the disparities (Table 3-9). Each assessor may take different amounts of time to chew their food and evaluate it before giving a rating. Their perception of stickiness may be impacted by the difference in oral processing times, which may also be a factor in the observed variances in ratings among the model foods (Wang and Hartel, 2021b). Another explanation for the significant differences could be that some of the assessors have already swallowed and the data points are reduced (Table 3-7). As a result, there are noticeable changes as a consequence of the reduction in data points. To fully comprehend the underlying mechanisms causing the observed significant differences between the assessors and the model foods, more investigation and analysis would be helpful.

For further analysis, the assessors were divided into groups based on the percentage difference between their last score and their first score (Figure

3-2). These results are presented in three categories: 0-30, 31-60 and 61-100 percent reduction in stickiness, with each category corresponding to a low, medium and high reduction in perceived stickiness, respectively.

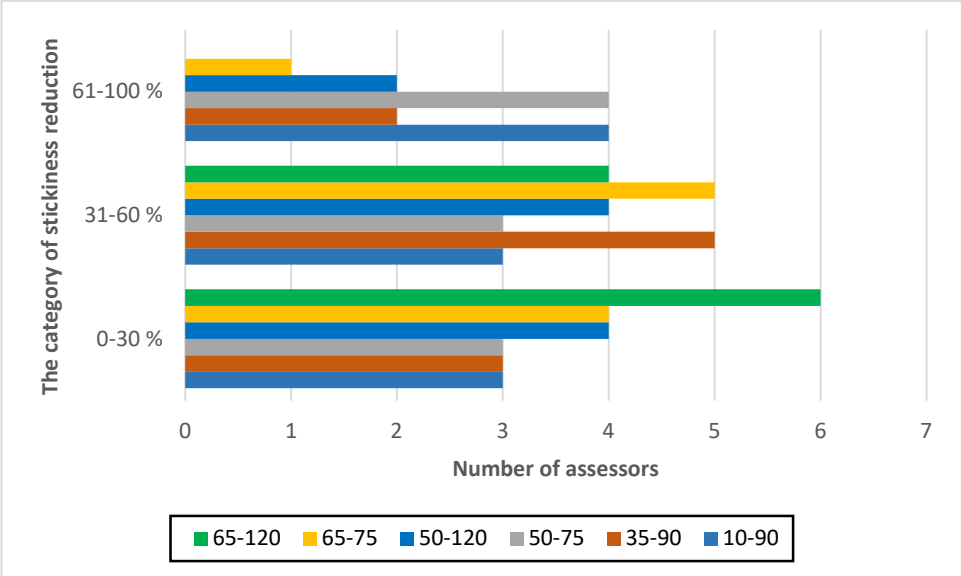

Figure 3-2. Categorising assessors on the basis of the percentage difference between their last and first measurement point. The criterion for selecting the last data point to determine the category was that there were at least 2 data points in the time interval. Each bar represents the number of assessors. For the 0-30% category, it can be seen that the highest number of assessors belonged to the sample with the highest stickiness (65-120), which means that the decrease in stickiness was lower for the model foods with higher stickiness than for the model foods with low stickiness (10-90, 35-90). On the other hand, the decrease in stickiness was greatest over time for the foods in the 61-100% low stickiness category (10-90, 35-90, 50-75) compared to the high stickiness foods (65-120).

The most notable aspect of the data presented in Figure 3-2 is that by increasing the sugar content, the high values (61-100%) of stickiness of the model foods decreases to a minimum (one and two assessors for 65-75 and 50-120 respectively) while do not even occur for 65-120 as the stickiest model food. Conversely, for the low reduction levels (0-30%), the number of assessors is higher for the least sticky samples (10-90, 35-90 and 50-75). For the low reduction levels, the highest number of assessors is found for the 65-120 model food, followed by 65-75 and 50-120. Categorisation of the data has also been suggested by Young et al. (2016) to group assessors based on various parameters, such as their bolus characteristics. They recommended that categorisation of assessors could lead to more realistic and applicable studies in which the results can be used to formulate tailored

food products. What is remarkable about the categorisation of the model foods is the ability of the different assessors to cope with different levels of stickiness just before safe swallowing, suggesting that they swallowed boli with even very high levels of stickiness. These may be related to parameters such as physiological differences between assessors (e.g., salivary secretion and chewing behaviour) and the physical properties of the food (Chen, 2009). As the number of assessors in the present study was limited, the categorisation of them cannot be generalised to a wider population, but it can be used as a reference for future grouping of foods with similar texture properties.

The literature emphasizes the existence of stickiness thresholds, which represent the minimum level of stickiness that individuals can perceive. These thresholds vary within individuals and are influenced by factors such as sensory sensitivity, familiarity with the food being evaluated, gender, and intraindividual variability (Dunnewind et al., 2004, Akissoe et al., 2006, Sarkar and Krop, 2019, Wang and Hartel, 2021b). Therefore, to obtain a comprehensive understanding of stickiness during oral processing, it is essential to consider multiple parameters in research and analysis. By considering these various factors, a more complete picture of the perception of stickiness can be obtained.

In the initial stages of the current thesis, it was hypothesised that the stickiest model foods with a highly cohesive-sticky texture would undergo the greatest changes during chewing. One of these changes should be the evolution of perceived stickiness during chewing. Figure 3-2, shows that the sensory data do not support this hypothesis of the present work that stickiness decreases to a certain point before swallowing. This hypothesis is based on the literature that products with an initial low moisture content tend to form a sticky bolus when swallowed (Adhikari et al., 2001, Loret et al., 2011, Peyron et al., 2011) and the current work investigated whether this was also true for products with a higher initial moisture content. One of the reasons for this difference could be the breakdown pathway for the model foods in the current study. The traditional breakdown of foods (according to Hutchings and Lillford (1988) suggests that foods are hydrated during the chewing

process before swallowing. However, the model foods in the current study are already hydrated and may not go through the traditional breakdown. This hypothesis is discussed in terms of muscle activity in the EMG section (see section 3.3).

3.2 Instrumental measurement

3.2.1 Texture analyser

The Texture Analyser (TA) is one of the most common and easy-to-use measuring devices used in both industry and research. There are several methods for measuring stickiness using TA, the accuracy of which may depend on various parameters such as the speed of the test and the rheological properties of the samples. In this section, three of the most commonly used parameters from TA are compared with two new parameters introduced for the first time by the research team of this thesis to develop methods which might give a more complete picture of stickiness. It should be noted that these new parameters have not been discussed in the literature previously.

By performing the compression-separation test, five parameters are derived from the obtained graph. Figure 3-3 provides a typical example of the graph with an explanation (see the legend) of how each parameter is calculated. The three parameters force of the adhesive peak (mN), distance to the adhesive peak (mm) and total-area (mN/mm^2) are the most commonly used parameters by researchers to measure stickiness (Fiszman and Damásio, 2000b, Chen et al., 2008, Yang et al., 2018, Wang and Hartel, 2021a). One of the new parameters introduced in this study is the pre-area (mN/mm^2) which is the corrected form of the total-area parameter. The aim of this correction is to reduce or eliminate the effect of necking (which is discussed in section 1.1.7) during the probe detachment. The other new parameter is the initial gradient which is calculated by writing a macro software based on the method of Kazemeini and Rosenthal (2021b). The initial gradient is calculated by selecting two points within the linear early stages of the probe detachment prior to the creation of the meniscus within the sample (Kazemeini and Rosenthal, 2021b, Kazemeini and Rosenthal, 2022). During the compression-separation experiment, when the model food is attached to the probe and before the curve reaches the negative peak value, the linear

force-distance behaviour starts to deviate, leading to a change in geometry. This change leads to the onset of necking and subsequently to a deviation in which measurements other than stickiness (e.g., rheological parameters) are considered. As a result, the initial gradient is calculated from the linear force-distance section of the negative area under the curve. The use of the initial gradient has been proposed in liquid foods (Kazemeini and Rosenthal, 2021b, Kazemeini and Rosenthal, 2022). The authors pointed out that the initial gradient, which has units of tension, mN/mm, has various advantages such as complete contact between the probe and the surface of the sample during the linear part of the force-distance detachment curve. As this method was suggested primarily for liquid samples, it should be used and considered with caution. Therefore, the interpretation of the findings could be limited or related to the physical state of the samples.

Figure 3-3 shows a typical force-distance curve from a compression-separation test. It can be seen in the figure that once the curve deviates from linearity (highlighted in blue), there is a corresponding thinning of the liquid strand which is the same phenomenon as necking (see section 1.1.7).

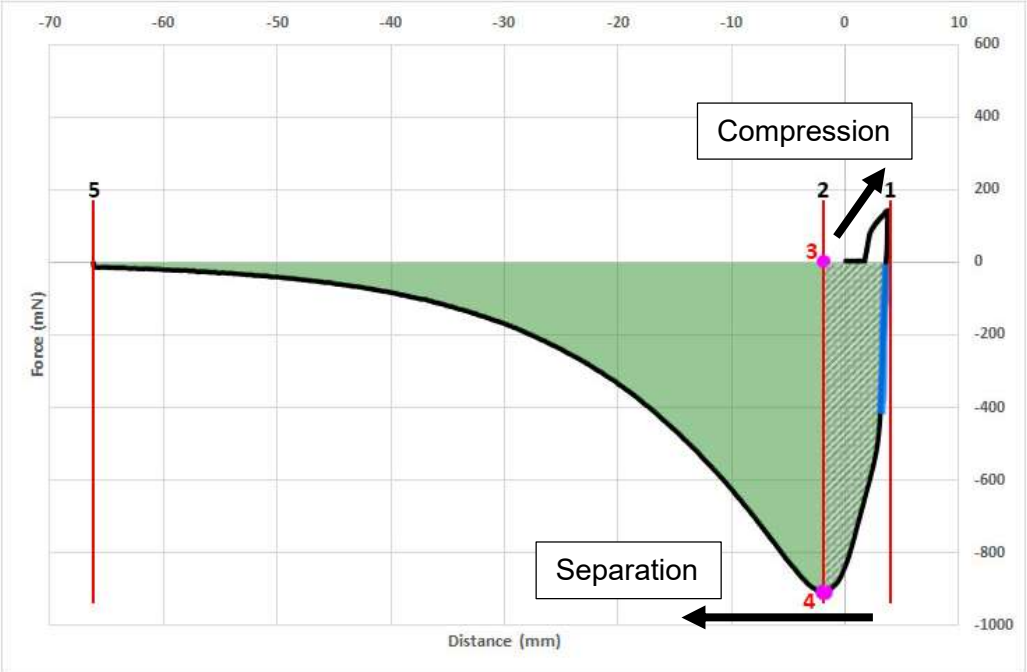

Figure 3-3. A typical force-distance curve from a compression-separation test (an example graph of model food 65-120). The texture analyser protocol started by moving the probe to the surface of the sample. Then, it was instructed to apply some force to ensure a complete contact between probe and sample. The main compression was to apply 10 g force at 1 mm/sec. Finally, separation occurred by moving the probe to 70 mm. The following parameters were extracted from the recorded data: Initial gradient: calculated by selecting two points from the highlighted blue part, force of the adhesive peak (mN): is marked as point 4 in pink, distance to the adhesive peak (mm): the distance between point 3 and point 4, Total-area (mN/mm^2): the area under the negative curve (above the curve) between line 1 and line 5 (highlighted in green), Pre-area (mN/mm^2): the area under the negative curve (above the curve) between line 1 and line 2 (dashed area).

The phenomenon of necking is more pronounced than surface tackiness in viscoelastic materials, which is the main reason for modifying the total-area under the negative curve (highlighted in green, in Figure 3-3) and obtaining the pre-area parameter. Another reason is the extent of contact between the probe and the sample, which continuously decreases during detachment. Referring to the work of Kazemeini and Rosenthal (2021b), it can be suggested that at maximum force (point no.4, Figure 3-3), both the necking and the reduction of the contact area are in the early stages, which can lead to more reliable data.

Figure 3-4 and Figure 3-5 represent the plots that resulted from the compression-separation test (each plot is randomly chosen). A typical test in the current research began with a compression step that resulted in an increase in force up to the maximum positive force. Then the probe was

pulled away from the surface of the sample (separation step - negative area) and the force returned to zero, with the speed of return to zero (distance of zero force at the end of separation) depending on the individual model foods.

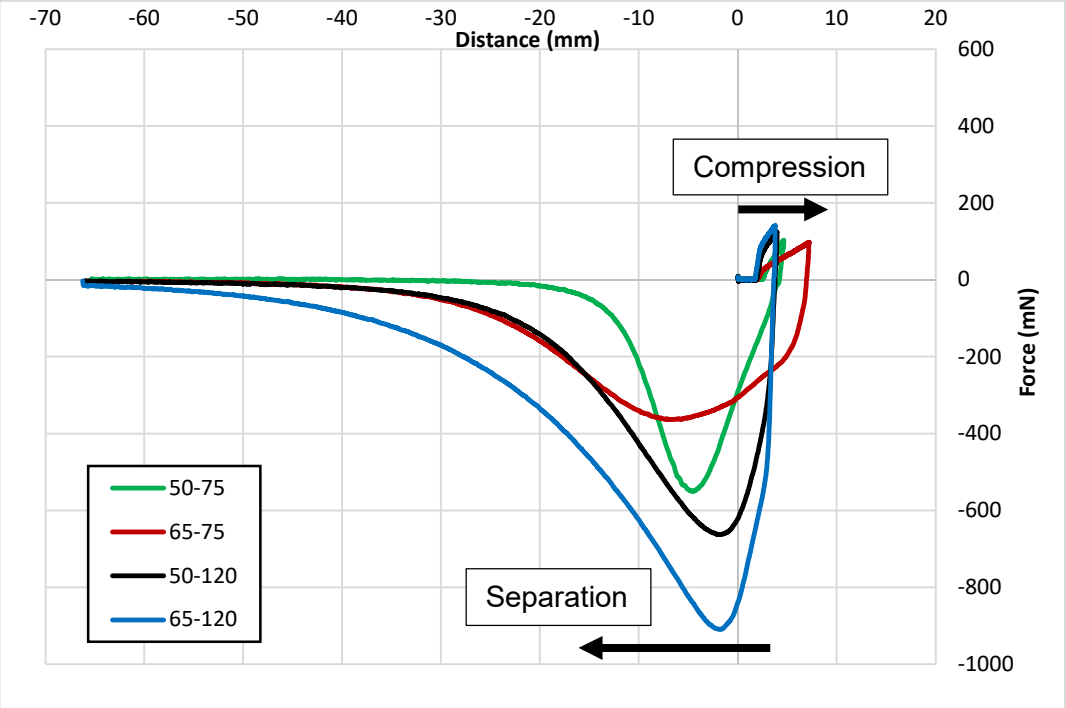

Figure 3-4. Examples of the shape of the compression and withdrawal of model foods 50-75, 65-75, 50-120 and 65-120 performed by texture analyser.

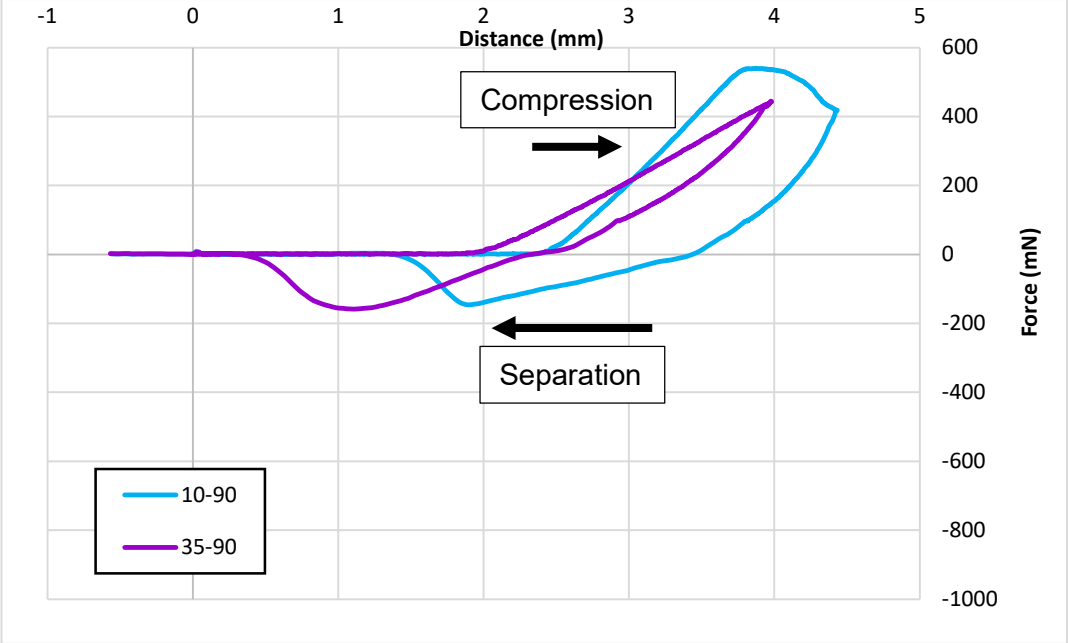

Figure 3-5. Examples of the shape of the compression and withdrawal of model foods 10-90 and 35-90 performed by texture analyser. It can be seen that both model foods return to the zero force very fast (in the negative area- distance the zero force at the end of separation).

Comparing the shape of the plots for the model foods represents fast returners (in the negative area) to zero force for the samples 10-90, 35-90 and slow returners for samples 50-75, 50-120, 65-75 and 65-120. This behaviour can be related to the speed of the test and thus the type of the failure. A fast return is indicative of adhesive failure, which contrasts with cohesive failure, which usually shows a slow return (Kilcast and Roberts, 1998). The observations of the present research experiments suggest that the shape of the plots for model foods is a combination of mainly the necking (long behaviour) as well as the mode of failure. At the same time, cohesive and adhesive failure also depends on the speed of the test. This means that an adhesive failure can be converted into a cohesive failure if the speed of the test is increased (Kazemeini and Rosenthal, 2022). It is also important to note that the protocol for the compression-separation tests was designed to minimise the necking by performing various tests, most notably by modifying stages 4, 5 and 6 of the compression-separation test (see Table 2-5). Another objective was to achieve complete detachment of the samples from the probe. This shows that the model foods with the highest surface stickiness tend to stick to the probe. This also accords with the comment of

Fiszman and Damásio (2000b), who mentioned that if the sample is still in contact when the probe reaches the trigger position (mainly for the TPA test and before the second compression step), the values for adhesiveness must be discarded. In addition, the necking can range from low to high extension depending on the consistency (Dunnewind et al., 2004). This can serve as a basis for the claim that although the mode of failure in the model foods is mainly adhesive, the degree of necking increases to a level that prevents cohesive failure (rupture). This means that the 10-90 and 35-90 samples have a higher resistance to stretching, resulting in a faster return of force to zero. It may not be "stickiness" in the sense that no strand forms over the sample when the probe is pulled away. The reason for achieving a complete detachment (sometimes not a clean detachment as some residue may remain on the probe) is to be sure that the curve reaches zero force and then the total-area parameter can be calculated, otherwise the validity of the recorded values is questionable.

It should be stressed that the aim of the compression-separation test is to measure surface stickiness. If cohesive failure occurs, this may affect the measurement of surface stickiness and the results will differ from the research objectives. Presumably, it is possible to tell if cohesive failure is occurring with a particular material and control the rate so that adhesive failure occurs. In the study by Schmidt et al. (2018), adhesiveness was defined as the maximum peel work, while stickiness was the ratio of separation work to penetration work, both referring to the area under the negative force/distance curves. The authors pointed out that adhesiveness (as a surface property) cannot fully explain stickiness. In another study, it was suggested that cohesive forces could contribute to a stronger prediction of stickiness (Dobraszczy, 1997).

Table 3-10 shows the values of the measured parameters for all model foods. Statistical analysis of the TA parameters was performed using the ANOVA multivariate test followed by Tukey HSD post-hoc comparison to test for significant differences in model foods based on the 5 instrumental parameters ($p < 0.05$). For a deeper understanding of the significant categories, the overall sensory stickiness scores are also included and can

be used to compare between the mean values of the model foods. It should be noted that the inclusion of the sensory data here is only to identify possible trends in the data that will be explored later in the section of correlating TA and sensory (see section 3.2.2).

As presented in Table 3-10, the values of most parameters increase with increasing sugar content, from model food 10-90 to 65-120. Model food 10-90 has the lowest values for all five parameters. In contrast, model food 65-120 has the highest values. The main exception is model food 50-120, which has higher values compared to model food 65-75 despite its lower sugar content. It is important to note that parameters initial gradient, pre-area and total-area, although negative, are greater in terms of stickiness as they receive more negative values (in the direction of stretching). These findings are consistent with those of Fiszman and Damásio (2000b) who also found that higher sugar content increased instrumentally measured adhesiveness for total area and the force of the adhesive peak (mN). They reported significant differences in adhesiveness for samples without added sugar and with 40 percent sugar content.

Table 3-10. Average data (based on 3 replications) of texture analyser experiments (values are presented as mean (standard deviation) resulting from Tukey HSD post-hoc comparison). At the separation speed of 10 mm/sec. The assigned letters indicate the significant differences, where the same letters are not significantly different ($p < 0.05$).

Model foods	Sensory stickiness rating	Initial gradient (mN/mm)	Force of the adhesive peak (mN)	Distance to the adhesive peak (mm)	Pre-area (mN/mm^2)	Total-area (mN/mm^2)
10-90	0.2^{a}	-103.0^{a} (30.0)	-127.0^{a} (30.7)	4.5^{a} (2.0)	-86.0^{a} (37.6)	-138.0^{a} (66.1)
35-90	2.3^{b}	-172.0^{a} (49.4)	-184.0^{b} (112.9)	7.1^{b} (1.8)	-163.0^{a} (153.4)	-281.0^{a} (242.3)
50-75	4.8^{c}	-76.0^{b} (12.1)	-557.0^{c} (26.1)	14.0^{c} (3.7)	-3514.0^{c} (1451.7)	-7881.0^{b} (3816.8)
50-120	8.0^{d}	-723.0^{c} (462.0)	-931.0^{d} (536.1)	9.0^{d} (2.1)	-4053.0^{d} (2460.2)	-17052.0^{c} (10887.3)
65-75	6.5^{e}	-297.0^{d} (265.8)	-502.0^{e} (202.4)	12.8^{e} (1.9)	-3925.0^{d} (430.5)	-13126.0^{d} (5506.4)
65-120	9.0^{f}	-2284.0^{e} (2122.4)	-1819.0^{f} (1541.2)	7.8^{b} (2.3)	-5733.0^{e} (4471.7)	-16971.0^{c} (7198.0)

It can be seen from initial gradient data that there are significant differences between model foods 65-120, 50-120 and 65-75. A closer look shows that the only exception that does not represent the rating order (in terms of sugar content) concerns the model food 50-75 with the lowest value. The interesting aspect of the initial gradient parameter is that although Kazemeini and Rosenthal (2022) used this parameter for liquid food samples, most of the model foods in the current study are not liquid and their TA values are consistent with their sugar content and heating time, except for the model food 50-75. Further research and analysis on different solid/semi-solid foods would be necessary to deeper understand the true physical meaning of the initial gradient.

Among the parameters presented in Table 3-10, the distance to the adhesive peak (mm) shows the greatest inconsistency in the data, as 50-75 is classified as the stickiest sample. This is in contrast to all other parameters where either the model foods 65-120 or 50-120 are rated as the stickiest samples. A possible explanation for this inconsistency may be the necking of the samples as the ability to stretch. It has been reported that necking can alter the recorded data from the measurement of surface stickiness to the rheological properties of the model foods (Hoseney and Smewing, 1999). Furthermore, when the effect of the increased amount of sugar in the model foods as well as the extended heating time on the increase of stickiness is considered, the force of the adhesive peak (mN) provides a more reliable assessment of stickiness compared to the distance to adhesive peak (mm). Another possible explanation for this may be the glass transition temperature (Tg). This temperature is about 36°C for sucrose, while it is 7.8°C and 8.6°C for fructose and glucose, respectively (Jeong-Ah et al., 2012). When sucrose is converted into its constituents fructose and glucose during heating, the Tg decreases. Mayhew et al. (2017) further concluded that the tactile stickiness of samples with a Tg below 15°C is high compared to samples with a Tg above 35°C. Mayhew et al. (2017) also stated that the samples with higher Tg had a brittle and crumbly texture, which could correspond to the low sticky samples in the current study (10-90 and 35-90). On the other hand, samples with low Tg have a deformable texture that has high tactile stickiness.

Although the Tg value was not measured in the current study, this could be a possible explanation for why 50-120 with a lower sugar content has a higher degree of stickiness compared to 65-75, which could be related to the higher proportion of glucose and fructose with a lower Tg value.

Looking at the pre-area data, samples 50-120 and 65-75 are not significantly different from each other, which is also true for samples 10-90 and 35-90. It should be noted that the original idea of proposing the pre-area parameter in the current research was to reduce the effects of necking. As mentioned in section 1.1.7, necking is an indication of rheology and not stickiness (Hoseney and Smewing, 1999). This is because the necking occurs before cohesive failure and not before adhesive failure (Kilcast and Roberts, 1998). Pre-area is the only parameter that provides a comparable rating of the model foods in terms of sensory stickiness. The main difference between the pre-area and the total-area is the higher value of 50-120 than 65-120 for total area.

Post-hoc comparisons of total-area results show that 10-90 has the lowest stickiness value and is not significantly different from sample 35-90. On the other hand, sample 50-120 has the highest stickiness value with no significant difference from sample 65-120. In agreement with these results, Dhaliwal et al. (1990) have shown that the total-area parameter provides significant values for stickiness of different wheat doughs.

The parameters that appear to separate samples based on sugar content are initial gradient (mN/mm), force of the adhesive peak (mN) and pre-area (mN/mm^2). For all these three parameters, either 65-120 or 50-120 samples had the highest stickiness (instrumental stickiness). The higher stickiness values for 50-120 could be explained by the effects of heating time and water evaporation. The longer heating time for sample 50-120 resulted in more evaporation, leading to a higher concentration of Soluble Solid Content (SSC) concentration compared to sample 65-75. Table 2-2 shows that evaporation resulted in 13.17% water loss for sample 65-75 compared to 26.12% for sample 50-120. In agreement with the present results, Downton et al. (1982) have shown that the hydrolysis of sucrose correlates positively

with heating time. This means that with increasing heating time, a greater amount of low molecular weight sugar is produced, which in turn leads to higher stickiness (Bhandari et al., 1997). These explanations could also be some possible reasons why sample 65-120 is found to be stickier than sample 50-120 for most parameters.

On the other hand, and with regard to the less sticky samples, it should be mentioned that the small diameter probe was not large enough to register a force on the less sticky samples (10-90 and 35-90). It could also be that the method developed was not sensitive enough for these samples. There were certainly sensory differences between the samples, although it may be that the TA protocol could not measure them.

Looking at the data presented in Table 3-10, the texture analyser data shows significant variation for some of the samples and parameters. This variability is more pronounced in stickier model foods and two specific parameters, pre-area and total-area. The higher variation of stickier model foods can be attributed to their complex texture, as noted by Fiszman and Damásio (2000a), who emphasized the impact of stickiness on TPA results. They recommended using various compression forces for different samples to reduce variability, but this approach may not be practical when comparing multiple samples. Nevertheless, it may help explain the higher variability in stickier model foods. Wee et al. (2018) also discussed the greater adhesive forces between stickier samples and the probe, which can contribute to the observed variability.

The higher variability of pre-area and total-area parameters can be attributed to their unit of force over area, which takes into account the surface area of the sample being compressed. In contrast, the force of the adhesive peak parameter, which only considers the force required to compress the sample, does not take the surface area into account. Thus, any variation in the surface area between the probe and the sample can affect the measurement of force per unit area, leading to higher variability.

The role of ingredients on surface stickiness

The role of low molecular weight sugars and starch on surface stickiness has been highlighted in the literature and for this reason their effect in relation to the model foods of the current study is discussed in more detail below.

Low molecular weight sugars are important ingredients affecting the stickiness of food products (Maidannyk and Roos, 2017, Wang and Hartel, 2020). Sucrose and its related monosaccharides glucose and fructose are considered low molecular weight sugars (Adhikari et al., 2001). The main constituent of the model foods in the present study is sugar, which undergoes catastrophic changes due to long heating times. Heating sugar solutions and converting them into glucose and fructose (invert sugar) is a complex chemical reaction (Montgomery and Wiggins, 1947, Richards and Shafizadeh, 1978, Msagati, 2012). The addition of citric acid to the mixture acts as a catalyst and further accelerates the hydrolysis process (Bhattacharya, 2014). In relation to the model foods, it can be seen that a higher sugar content leads to a higher content of low molecular weight sugars. Based on the above information, it can be concluded that longer heating times could explain the higher stickiness of sample 50-120 compared to sample 65-75. Although the amount of low molecular weight sugars was not measured in the current study, it can be assumed that the longer heating time leads to the formation of higher amounts of low molecular weight sugars. It should be mentioned that the amount of these sugars can be measured using Differential Scanning Calorimetry (DSC). This method measures the glass transition of the samples. At a lower Tg, the average proportion of low molecular weight sugars is more dominant, resulting in a lower degree of stickiness.

Starch plays an important role in stickiness. One of the first effects of sugar/starch solutions is to increase the gelatinisation temperature of starch, as well as to compete strongly in water absorption (Eliasson, 2004). The increased heating time prolongs the hydrolysis effect of the added citric acid on the starch granules. The glucose equivalent (DE) is an indicator of the extent of hydrolysis (Surendra Babu et al., 2015). Although the DE values

were not measured in the study, it is known that an increase in acid hydrolysis (either due to a longer time or a higher acid concentration) increases the DE value of the solutions (Surendra Babu et al., 2015). The effect of DE on peeling force and peeling work was studied on caramel samples. With the increase of DE, the peeling force and the work of adhesion also increase (Wang and Hartel, 2021a). Increased DE values mean higher amounts of low molecular weight sugars, which in turn is another explanation for the higher stickiness of sample 50-120. Some studies have highlighted the effects of leached amylopectin as well as the ratio of amylose/amylopectin on the instrumental measurement of stickiness (Cameron and Wang, 2005, Patindol et al., 2010, Li et al., 2016, Li et al., 2017). These authors reported that higher molecular sizes of leached amylopectin were positively correlated with stickiness. Furthermore, the presence of amylopectin with a higher proportion of low molecular weight components increases the possibility of molecular interaction, leading to enhanced binding between the samples and the probe (Li et al., 2017). This enhanced binding is apparently related to the higher degree of stickiness. Although the amount of amylopectin was not measured for the model foods in the current study, it can be speculated that the samples with higher stickiness may have higher levels of low molecular weight amylopectin due to the longer heating time.

Visual analysis of the probe surfaces of the model foods during contact showed that 65-120, 65-75, 50-120 and 50-75 were able to make more extensive contact compared to samples 10-90 and 35-90 (this behaviour could be due to their ability to deform and flow). The latter model foods had a firmer and more textured surface, which could limit their ability to make complete contact regardless of the given contact time. The softness of the samples allows them to make full contact through deformation. If the material is less deformable, it could make poor contact with the probe even if sufficient time is given for contact before conducting the experiments (Crevoisier et al., 1999, Chuang and Yeh, 2006). In addition to deformability, wetting of the surface by the probe is another important factor that influences contact time and contact force to promote the formation of an acceptable

contact. Contact formation has a great influence on the strength of an adhesive bond (Zosel, 1997).

In summary, the results from TA provide interesting insights into model foods. While the usual TA parameters used by researchers provide useful information, two new parameters of pre-area and initial gradient, proved to be very effective in analysing surface stickiness. These parameters reduce the effects of necking on the stickiness of materials and measure stickiness in a more controlled manner and are less influenced by other rheological parameters. Among the parameters of TA, the pre-area provided ratings in line with the sensory evaluation of stickiness.

3.2.2 Texture analyser measurement in comparison to the sensory analysis

This section presents the correlation analysis of the TA measurement and sensory stickiness. The correlation was run for both individual time intervals and overall sensory stickiness values to see how the results differ for these two measurements.

Table 3-11 presents the Pearson correlation coefficients relating texture analyser parameters to sensory evaluation data.

The distance to the adhesive peak (mm) provides the lowest correlation coefficients compared to all other TA parameters. One explanation for the weak correlations of distance to the adhesive peak (mm) with the sensory data may be due to the different curve shapes of the different model foods, which lead to unreliable data compared to other parameters.

Initial gradient provides moderate correlations with sensory evaluation results. It should be noted that the initial gradient compared to the distance to the adhesive peak gives a more consistent rating of the samples compared to the sensory ratings. The reason for this could be that the initial gradient is obtained from the linear part of the separation phase of the compression-separation test, where the influence of rheological properties (such as necking) is minimised. As Kazemeini and Rosenthal (2021a) point out, the contact between the probe and the model foods is complete within the initial linear gradient, but as soon as linearity ends, the contact area starts to decrease and necking occurs. The same reason (necking) could be responsible for the unreliable data of the distance to the adhesive peak. The necking affects the results by including other physical parameters in the measurement than the surface adhesiveness.

Table 3-11. Pearson correlation coefficients relating texture analyser parameters to sensory evaluation data. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01 (both two-sided), respectively.

EMG Features	Time intervals					Overall Stickiness
	5 s	10 s	15 s	20 s	25 s	
Initial gradient (mN/mm)	-0.70	-0.71	-0.72	-0.74	-0.79	-0.71
Force of the adhesive peak (mN)	-0.84*	-0.85*	-0.85*	-0.87*	-0.92*	-0.85*
Distance to the adhesive peak (mm)	-0.44	-0.43	-0.42	-0.41	-0.34	-0.43
Pre-area (mN/mm^2)	-0.96**	-0.96**	-0.96**	-0.97**	-0.96**	-0.96**
Total-area (mN/mm^2)	-0.98**	-0.98**	-0.98**	-0.98**	-0.96**	-0.98**

It can be seen in Table 3-11 that force of the adhesive peak provides fairly strong and significant correlations with sensory data. These results are consistent with the findings of Chen et al. (2008) who suggested a positive correlation with sensory perception of stickiness for the same parameter. They studied different liquid foods by performing a separation test with a texture analyser to obtain the adhesion peak and a tactile test in which the testers measured the stickiness of the liquid samples with their thumb and index finger.

The comparison of TA data with sensory evaluation should be carried out taking limitations into account. Matsuyama et al. (2021) discussed that the stainless steel probe is not able to act similarly to oral surfaces. They argued that the aluminium surface is a hydrophobic material, whereas the oral surface is hydrophilic. Therefore, there would be some deviations from reality and a reduction in the quality of correlations when conducting TA experiments compared to sensory evaluations. Another point of attention is the effect of saliva during oral processing. The lack of saliva when performing TA experiments can cause deviations compared to the oral manipulation of foods. Although the use of saliva in stickiness related studies

is limited and sometimes their results are contrary to expectations, the importance of saliva should not be underestimated (Wang and Hartel, 2021a).

In relation to the above results, it can be mentioned that the TA parameters showing the work of adhesion (total-area and pre-area) have strong and significant correlation with the sensory data. As explained in the TA section, the total-area parameter captures not only the surface stickiness of the model foods, but also their rheological properties. By correcting this parameter and introducing the pre-area parameter, only the surface stickiness of the model foods should be considered. One point to consider, which is more of a speculation, is that if the pre-area and total-area had been compared with some other surface properties of the model foods or the tactile sensory perception of stickiness by the fingers, this may have resulted in stronger correlations for the pre-area, but the comparisons were made with sensory data that included both surface and rheological properties of the model foods. Therefore, further research is needed to apply the initial gradient to model foods with different textural properties in order to draw a more robust conclusion.

3.2.3 Rheology

Rheology, the study of the flow and deformation of substances, was used to characterise the model foods. As detailed in the materials and methods section (see section 2.1.2), six model foods, based on a Turkish Delight recipe, were made with varying amounts of sugar and different heating times. The samples were coded with the first part as the percentage of sugar and the second part as the length of cooking time, for example, sample 10-90 refers to a sample which is 10% sugar and was cooked for 90 min. It was clear from observations that the more sugar and the longer cooking time created a sticker food, so rheology was used to probe this complex rheological properties and link them to the textural attributes. This should improve the overall understanding of stickiness measurements which are currently lacking in the literature. To assess the complex viscoelastic properties of model foods, the stress relaxation method was chosen. The advantage of stress relaxation measurements is ability to probe slow dynamics and viscoelastic response at low frequencies which typically cannot be accessed using small amplitude oscillatory shear tests. The impact of slow relaxation dynamics and low frequency viscoelastic response was highlight in the literature Kazemeini and Rosenthal (2022) as important factors in slow relaxing systems, such as Turkish Delight.

3.2.3.1 Relaxation behaviour of model foods

To perform stress relaxation test a two-step strain test was performed (see section 2.4.2). The first step was performed at 0.01% strain (within LVE range) for almost 7 minutes and was to attempt to limit the impact of the sample history caused by the preparation steps as well as increase reproducibility. For some of the model foods, mainly high sugar content ones (e.g., 50-120 and 65-120), the reproducibility of the experiments was poor. Therefore, instead of using the absolute values, the magnitude of the changes was analysed to give the overall picture of the results. This was sufficient for the needs of this research.

Once the pre-strain step had occurred, a strain of 0.1% was imposed on the samples for almost 27 minutes (data recorded every 4 sec) and the relationship between shear stress and relaxation time during the initial part of that period are presented truncated at the point that the stress reaches equilibrium to show the detail at the start of the experiment (Figure 3-6, Figure 3-7). Figure 3-6 shows that the stresses within the model foods with 50% and 65% sugar reduce quickly, showing liquid-like behaviour. They release the stress easily and are all able to reach almost complete relaxation within 180 seconds. Sample 50-75 showed a higher elastic modulus in the amplitude sweep, however the difference between the elastic and viscous moduli was not very much which could explain this result. The equilibrium stress for this sample were also slightly higher than that for samples 50-120 and 65-75. Sample 65-120 took longer to reach complete relaxation than the other samples and showed a fluctuating decrease rather than a smooth one. The two remaining samples (10-90 and 35-90) were able to retain the stress, which correlates with the information from the amplitude sweep results showing a higher elastic modulus for these samples.

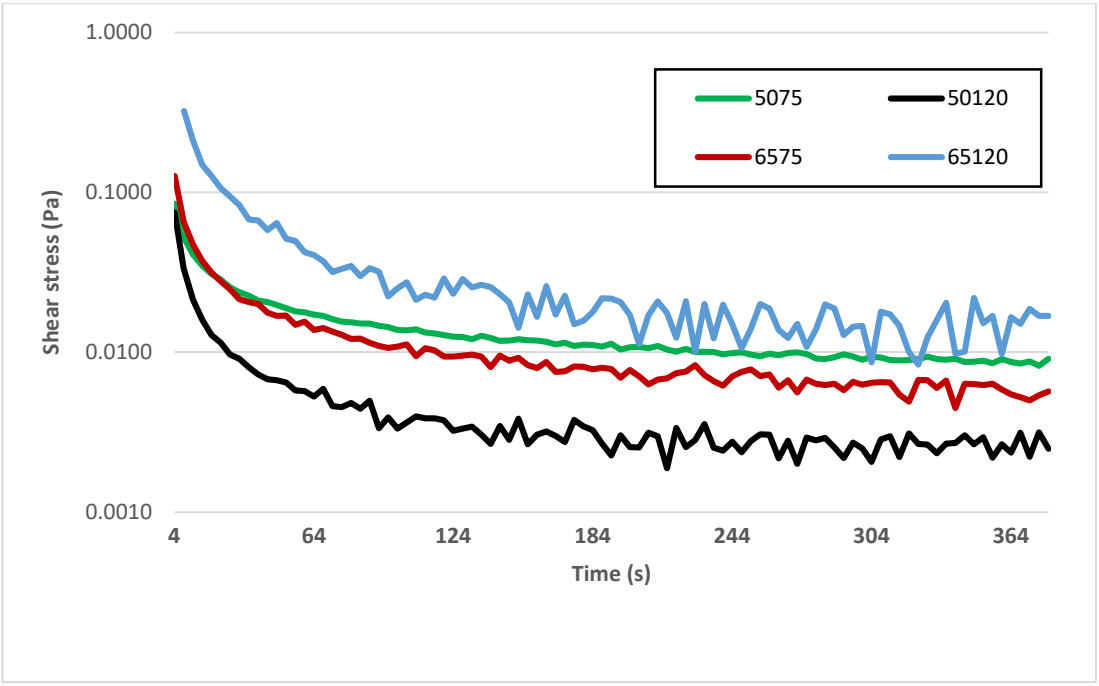

Figure 3-6. Relaxation curves for 50-75, 50-120, 65-75 and 65-120. The tests were performed at constant 0.1% strain (second step strain interval), while the shear stress of the samples was recorded every 4 seconds at 25℃. These model foods have a different relaxation behaviour comparing to 10-90 and 35-90 (figure 3-16). Although 1600 s of data were recorded, the graph was truncated after equilibrium was achieved.

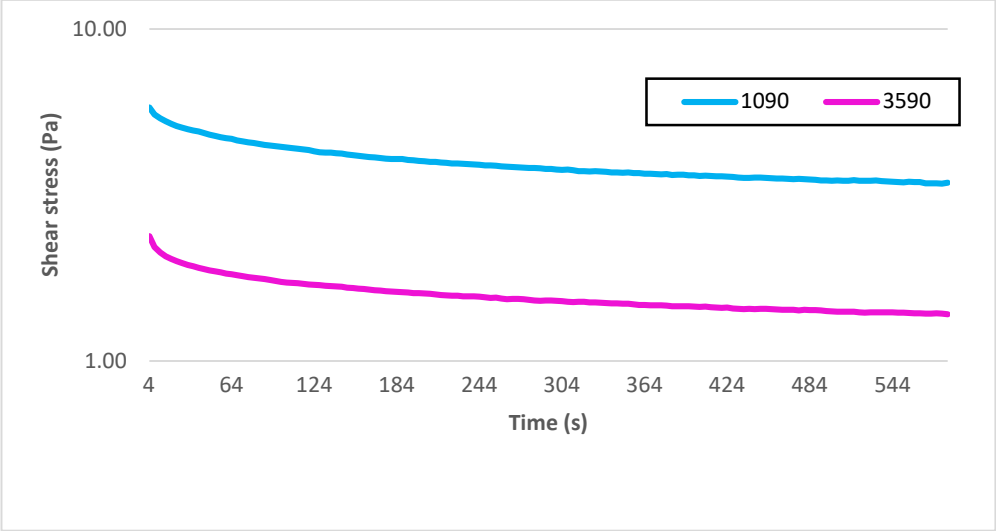

Figure 3-7. Relaxation curves for 10-90 and 35-90. The tests were performed at constant 0.1% strain (second step strain interval), while the shear stress of the samples was recorded every 4 seconds at 25℃. Although 1600 s of data were recorded, the graph was truncated after equilibrium was achieved.

3.2.3.2 Determining the linear viscoelastic range

The Linear Viscoelastic range (LVE) is a critical value within which the stress relaxation test should be performed. The results of amplitude sweep tests to determine the LVE of model foods are presented in Figure 3-8 (lower sugar samples) and Figure 3-9 (higher sugar samples). The LVE is the region where the oscillatory strain varies linearly with the complex stress (reflected by the storage modulus in the figures).

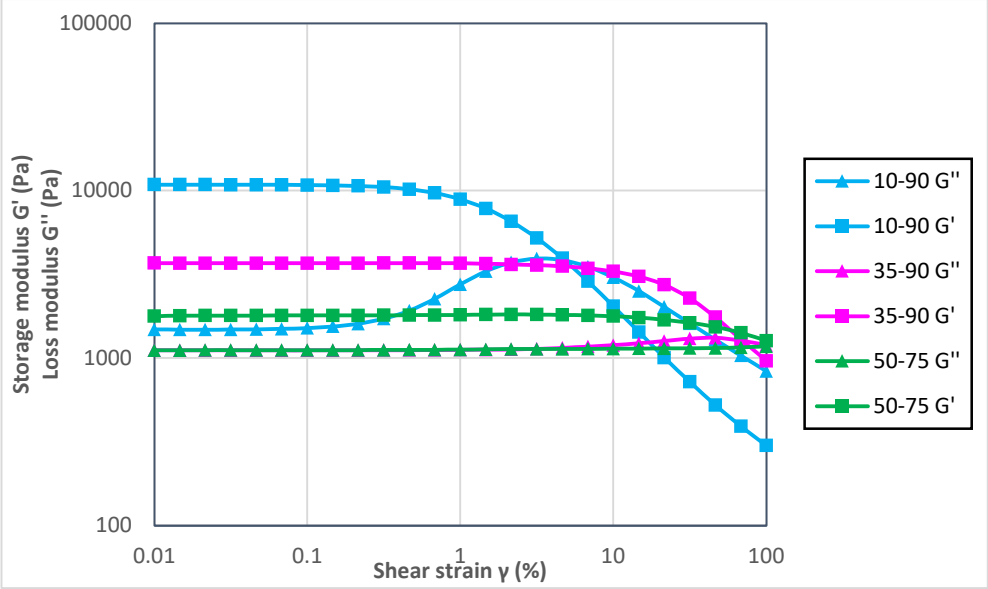

Figure 3-8. Amplitude sweep test to measure the LVE for model foods 10-90, 35-90, and 50-75. Shear strain of 0.01% to 100% is applied at 25 ℃ and angular frequency of 10 1/s. It can be seen that the 10-90 samples have a shorter LVE compared to 35-90 and 50-75. All three samples show a crossover point within the shear strain range measured, which occurs sooner for 10-90 and later for 50-75. A crossover point at a lower shear strain indicates a weaker structure than a crossover at a higher shear strain. Note that the G˝ data for the 35-95 sample is mostly overlaid by the G˝ data for the 50-75 sample.

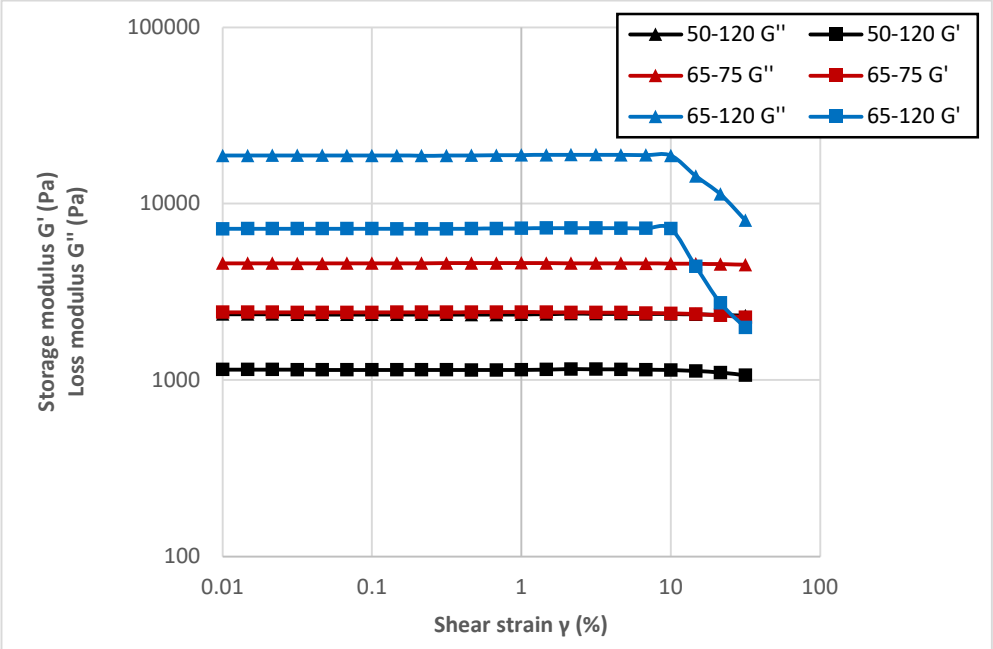

Figure 3-9. Amplitude sweep test to measure the LVE for model foods 50-120, 65-75 and 65-120. Shear strain of 0.01% to 100% is applied at 25 ℃ and angular frequency of 10 1/s. It can be seen that these sticker model foods have a long LVE, and none show a crossover within the applied shear strain which indicates a strong internal structure. Note that the G˝ measurement for 50-120 sample is overlaid by the G´ measurement for the 65-75 sample.

It can be seen from above figures that model foods containing higher sugar possess longer LVE compared to low sugar model foods.

Based on the data from Figure 3-8 and Figure 3-9 and considering the 3% LVE (more than 3% percent deviation from linearity), the LVE limit for each model food is proposed in Table 3-12.

Table 3-12. Linear viscoelastic region proposal for each model food. These have been selected using the 3% LVE based on the amplitude sweep test at shear strain of 0.01% to 100% at 25 ℃.

Model food	**10-90**	**35-90**	**50-75**	**50-120**	**65-75**	**65-120**
Proposed LVE (%)	0.1	1.0	10.0	10.0	10.0	5.0

The linear viscoelastic region of the model foods range between 0.1 and 10% (Table 3-12). Therefore, the stress relaxation experiments were performed at a constant strain of 0.1% for all samples. This was selected so that the applied strain would not destroy the structure of any of the model foods and avoid physical changes that cannot be recovered during the time of the test (Wagoner et al., 2016). In this case,10-90 was the most sensitive model food with the lowest LVE limit. Determining the LVE is a crucial step prior to testing the model foods. Steffe (1992) stated that performing experiments in LVE, the material function does not depend on the amount of applied stress or strain, so it provides a proportional response.

3.2.3.3 Relaxation modulus of model foods

The relaxation modulus ($G_{(t)}$) of a sample is defined by stress variations under constant unit strain, and calculated using the equation below:

$$G_{(t)} = \frac{\tau_{(t)}}{\gamma_0} \qquad (3\text{-}1)$$

where $\tau_{(t)}$ is the stress relaxation function and γ_0 is the preset shear strain.

The relaxation modulus of the samples in this research is shown in Figure 3-10. This shows a similar curve profile to the stress time data, which is to be

expected as the shear strain was set at a constant 0.1% for all experiments. The two sample with the lowest sugar concentrations reach a constant level of relaxation quickly, but these are high compared to the other samples (sample 10-90 has a $G_{(t)}$ of 4,080 Pa and sample 35-90 has a $G_{(t)}$ of 1,543 Pa). The sample 50-120 has the lowest $G_{(t)}$ of around 8.6 Pa. Samples 50-75 and 60-75 have $G_{(t)}$ values of around 10 Pa and sample 65-120 is slightly higher at around 15 Pa. A similar behaviour was also observed by Del Nobile et al. (2007) who reported a quick and recoverable deformation for elastic samples (such as pan bread) comparing to samples with bulky texture (such as agar gel). A faster dissipation energy was linked to a less degree of polysaccharide entanglement as an indication of weak gel structure (Kajuna et al., 1998). It can be hypothesized here that the faster dissipation of energy within the model foods might be due to less entanglement of starch structure. Faster energy dissipation is an indication of viscous modulus that results in higher levels of adhesion (Grillet et al., 2012). This behaviour was also discussed by Jones et al. (1997) using spring and dashpot concepts. They mentioned that at high frequencies there is enough time for the spring element to elongate, while the dashpot does not have enough time to move. Consequently, a gel-like behaviour would be observed. On the other hand, at lower frequencies, as there is enough time for movement of both spring and dashpot, the material would behave more like a liquid. This is in line with previous findings of the current research and their level of stickiness. The stickiest samples (50-120, 65-75 and 65-120) have a fluctuating $G_{(t)}$ through time which gets more pronounced after 100 s. This may be caused by the stickiness of the sample interfering with the measurement.

In order to compare the speed of the reduction of $G_{(t)}$ between samples the “apparent relaxation time” can be used. This involved collecting the first $G_{(t)}$ value, calculating percentage reductions and finding the $G_{(t)}$ at which this reduction was seen. Goh and Sherman (1987), devised this parameter to find the point at which their cheese samples reached a $G_{(t)}$ of 36.77% their starting level. In this study, $G_{(t)}$ values at 74%, 37% and 20% were calculated and they are shown, with the maximum reduction (%), in Table 3-13 for all samples. As expected, model foods 10-90 and 35-90 had different

behaviours compared to other model foods indicated by the highest stress reduction for both is to about 50% of their initial stress levels so only the 74% value can be obtained. The maximum reduction for other samples is considerably lower with samples 65-120 and 50-120 lowest at 0.23 and 0.24 Pa, respectively. Comparing the data at 74% as a higher level of stress revealed that model foods with 50 and 65% sugar relax very quickly; at the second collection point (8 seconds), they have all decayed by at least 26% of their initial stress in which 50-120 decay the maximum level of stress at the last collection point which is followed by 65-75.

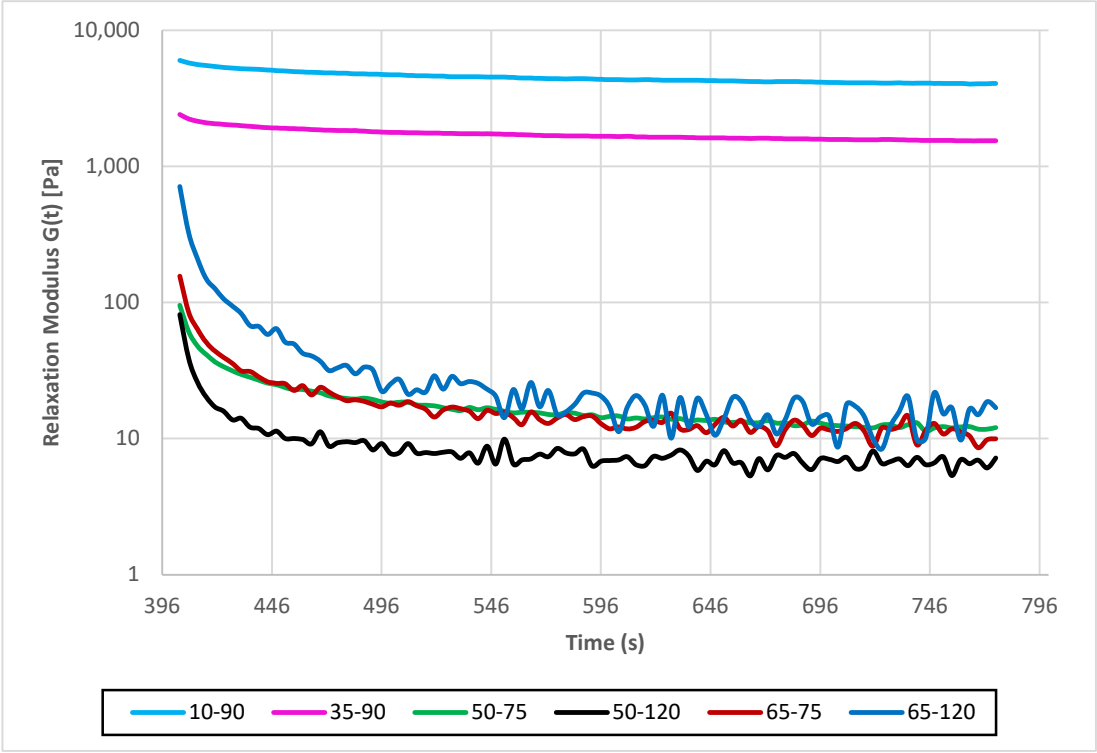

Figure 3-10. Relaxation modulus over time for all the model foods. Model foods 10-90 and 35-90 reach a constant level of relaxation modulus much quicker than all the other model foods. It can be seen that 10-90 has a reduction of about 53% in relaxation modulus comparing to 65-120 with over 99% reduction. This behaviour is related to the level of their elastic and viscous modulus. The time value starts from 396 s as the relaxation time of the second step strain starts.

Table 3-13. Apparent relaxation time of model foods at 74, 37 and 20% reduction of initial stress level and maximum reduction in the level of stress at 1600s as the last data collection point. Note that points were taken every 4 seconds during the experiment.

Model food	Time (s) to reduced levels of shear stress (% of maximum)			Maximum shear stress reduction (%) (at 1600 s)
	74%	37%	20%	
10-90	124	N/A	N/A	53.62
35-90	92	N/A	N/A	51.86
50-75	8	24	68	9.07
65-75	8	12	28	3.16
50-120	8	12	20	0.24
65-120	4	8	16	0.23

3.2.3.4 Effect of frequency on loss and storage modulus

Due to poor reproducibility of some of the model foods, a frequency sweep experiment was performed to examine the behaviour of the samples at low frequencies to explore long term behaviour and moving to high frequencies to explore short term behaviour. Here, the angular frequency was increased from 5 x 10^{-6} to 2 x 10^{3} and the shear strain was held constant at 0.1%. One replicate of each model food was randomly selected to evaluate the behaviour of G´ and G˝ over the frequency domain.

3.2.3.5 Storage (elastic) modulus

Looking at Figure 3-11, it is apparent that G´ of 10-90 and 35-90 shows a narrow range of frequency dependency which is at the lower frequency domain (less than 2.99×10^{-4} rad/s). Above this frequency range, at any value tested, G´ does not change significantly. The other model foods show increasing G´ values from low to high frequency domain, with some fluctuations. A weak gel-like structure was associated higher values of G′ than G″ (Seo and Yoo, 2013). This weak gel-like behaviour is apparent for model foods 10-90 and 35-90 which demonstrate both the mentioned parameters over most of the frequency range.

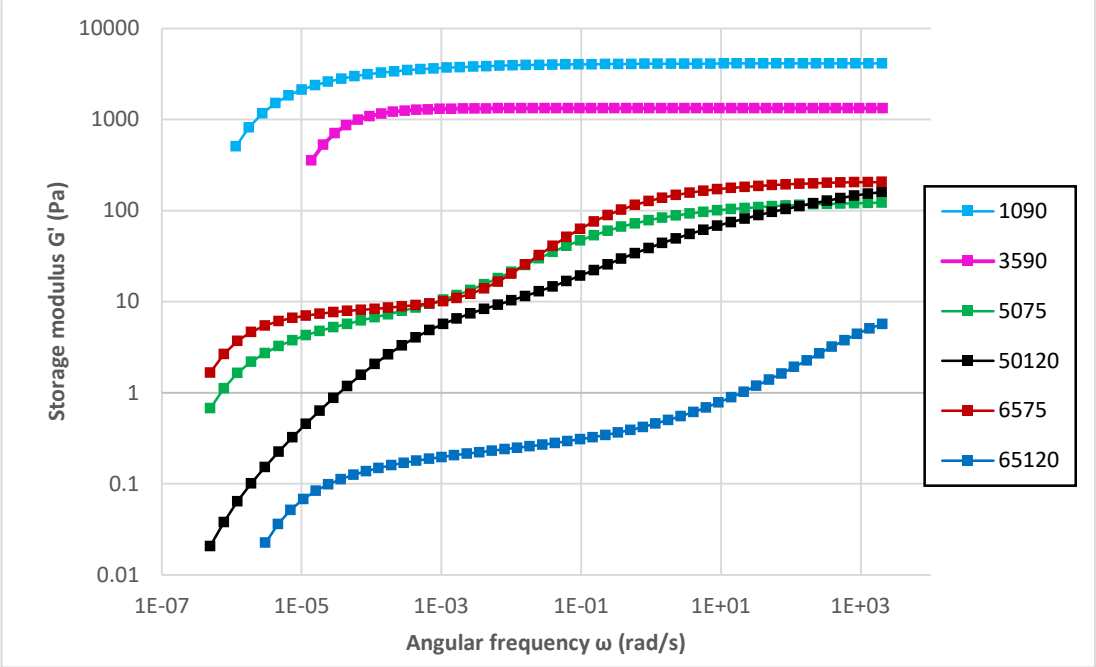

Figure 3-11. Magnitudes of G´ over the frequency domain for all the model foods. Model foods show a frequency dependency over the frequency range.

Figure 3-11 reveals that over most of the angular frequency domain examined, the rating of the samples appears in the same order, roughly following the sugar content and heating time parameters of the model foods. However, the model foods cooked for 75 min (50-75 and 65-75) swap positions in the rating depending on the frequency of the oscillation.

In general, at all frequencies, there is a negative relation between G´ and sugar content/heating time of the model foods, with heating time having more impact than sugar content in the higher sugar samples. It means that by increasing these parameters, G´ decreases regardless of the frequency. Model food 10-90 with the lowest sugar content shows the highest G´, while 65-120 with the maximum sugar content and the extreme side of the heating time, has the lowest G´ values over the frequency range.

3.2.3.6 Loss (viscous) modulus

Figure 3-12 provides the G″ values for all the model foods over the frequency domain. Once again, 10-90 and 35-90 show a different behaviour compared to the other model foods. From the frequency value of 4.38×10^{-5} rad/s to the

highest presented frequencies (over 10^3 rad/s), G″ constantly decreases from 1000 and 500 Pa, to 1.72 and 1.6×10^{-4} Pa for 10-90 and 35-90, respectively. Due to this, G″ reduction for 10-90 and 35-90, the frequency of 4.38×10^{-3} rad/s becomes an important point in which by crossing that the ranking of the samples will not follow the sugar content and heating time parameters mainly for 10-90 and 35-90. Prior to this frequency, most of the model foods show G″ values according to their formulations and processing time.

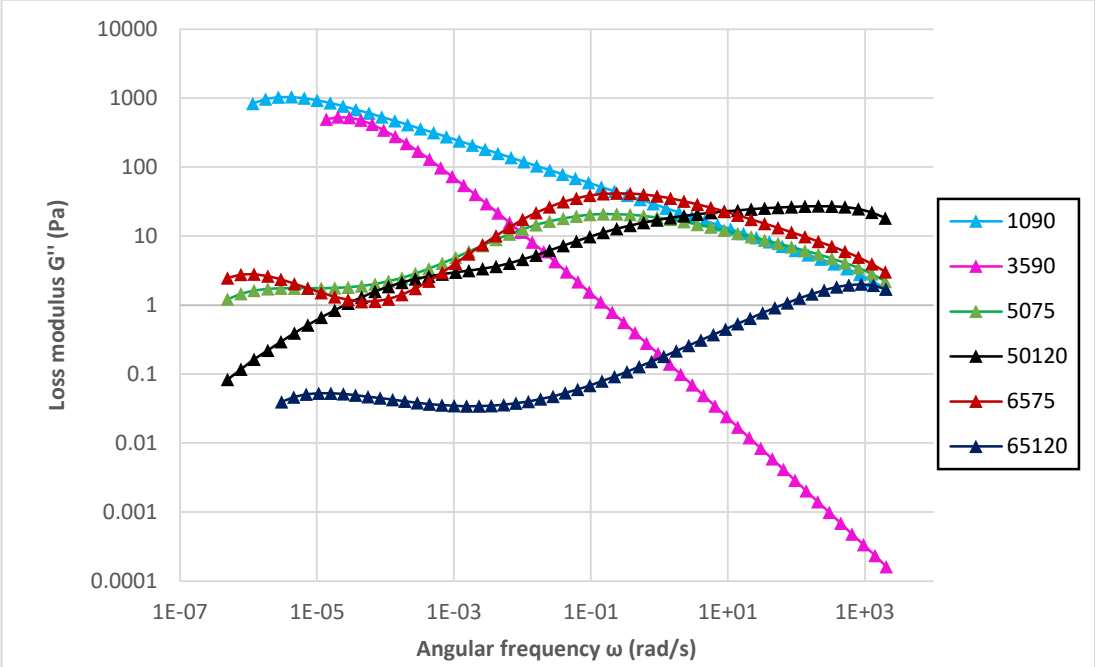

Figure 3-12. Magnitudes of G" over the frequency domain for all the model foods. The frequency dependency of model foods over the frequency range is apparent.

Comparing the Figure 3-11 and Figure 3-12, reveals that the model foods 50-75, 50-120, 65-75 and 65-120 show a viscous dominancy at the frequency range of 10^{-4} – 10^{-6} rad/s. It should be noted that the frequency dependency of sugar content food products was also reported by others (Steiner et al., 2003, Ahmed et al., 2006, Schmidt et al., 2018).

Considering Figure 3-11 and Figure 3-12, the effect of frequency change can be discussed in more detail. It can be seen that at low frequencies, storage modulus of the high sticky model foods is significantly lower comparing to 10-

90 and 35-90. By increasing the frequency, both 50-120 and 65-120 demonstrate the maximum increase in their storage modulus. On the other hand, 10-90 and 35-90 do not show any frequency dependency from medium to high frequency values. This frequency dependency of storage modulus was also discussed by Ahmed et al. (2006) for caramel samples. They reported that the increase of loss modulus lead to higher rigidity values in the samples. The relationship between rigidity and frequency might be related to the need of carbohydrates for a degree of relaxation which is not available at higher frequencies. The opposite is valid as the samples at lower frequencies have enough time to relax and subsequently behaves as a liquid. The same behaviour was observed in the high sticky model foods of current study with more pronounced behaviour in 65-120 and 50-120. These samples were difficult to handle when the pace of handling was fast but easier to handle at lower paces. This can also be noted from the low G´ at the low angular frequencies for the higher sugar samples. This observation is in accordance with Schmidt et al. (2018) who noted the same flowability behaviour for toffee and ungrained candies. They also added that this is the reason why samples showed a tendency to creep.

The importance of both elastic and viscous characteristics has been highlighted by Moll et al. (2022) in which both modulus are required for modifying the levels of stickiness. They suggested that a minimum level of both is essential for increasing the stickiness. It was also suggested by Roos et al. (2002) that beside G′ and G″, applying stress is also essential to make an optimal adhesion. The solid part usually associated with high molecular polymers make a backbone for adhesion and liquid part mainly resulted from low molecular materials provides the flow and deformation. Considering the model foods of current study, the high sticky samples have a lower storage modulus comparing to low sticky model foods at low frequencies, while it is mainly the opposite at higher frequencies. Then it can also be suggested based on the findings of the current research that in order to modify or control the levels of stickiness in sugar/ starch-based samples, the amount of storage and loss modulus can be altered by either formulation or processing.

3.2.3.7 Tan delta (tanδ)

The loss or damping factor, tangent of the phase or simply tan delta, (tanδ), can also be obtained from stress relaxation test. It is a function of frequency and it describes the viscoelasticity of the samples as a ratio of viscous modulus over elastic modulus, see equation (3-2) (Steffe, 1996).

$$\tan(\delta) = \frac{G''}{G'} \qquad (3\text{-}2)$$

Ideally elastic behaviour would give a tanδ of 0 since G´ would completely dominate G″, and ideally viscous behaviour would give a tanδ of ∞ since G″ dominates G´. If the elastic and viscous behaviour balance, i.e., G´ = G″, then tanδ would = 1.

The tanδ values for the model food samples are shown in Figure 3-13. It can be seen that tanδ for 10-90 and 35-90 constantly reduces by increasing the angular frequency. This pattern happens for all model foods (except for 65-120) after the angular frequency of 9.69 x 10^{-2} (rad/s). The tanδ value for 65-120 continues to increase up to 73.6 (rad/s) after which it starts to diminish.

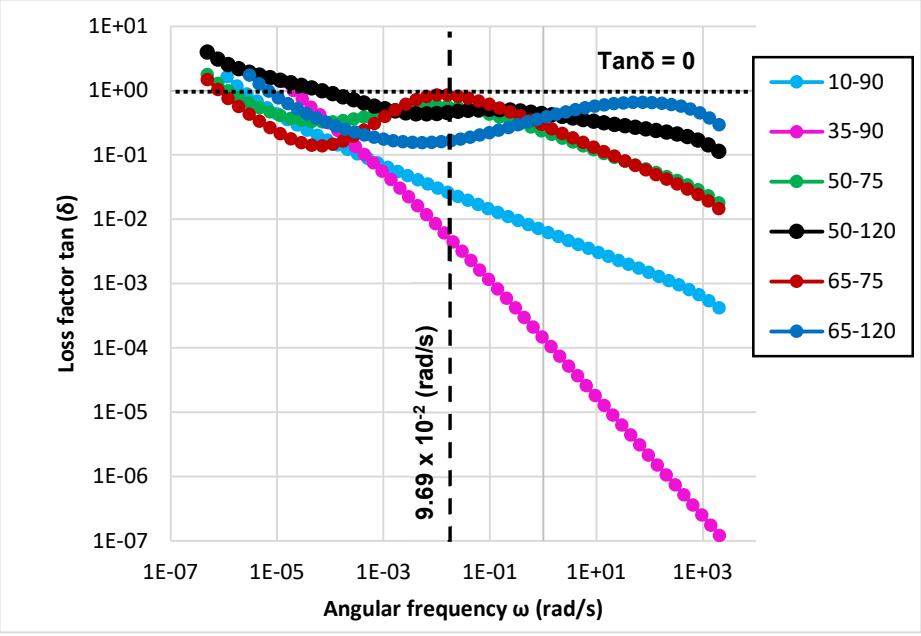

Figure 3-13. Tanδ as a function of angular frequency for model foods. The dashed line is at 9.69 x 10^{-2} (rad/s), the point at which all but 65-120 samples decrease in tanδ. The dotted line is at Tanδ = 0, the point at which G´ = G″.

The effect of frequency domain change shows a different behaviour for 50-120 and 65-75. Both model foods show multiple points at varied frequency domains where magnitudes of G´ and G˝ become equal (tanδ = 1). This point is called crossover frequency (Rao, 2013) and it is an indication of starting the elastic behaviour or approaching gel properties (Norziah et al., 2001). The crossover frequency is a useful parameter defining the viscoelastic behaviours of the material. If the crossover happens at low frequencies, it means the more contribution of G´.

By comparing the model foods rating by the assessors (high to low stickiness: 65-120, 50-120, 65-75, 50-75, 35-10 and 10-90) with tanδ values, it can be noted that above the frequencies of 2.67 (rad/s), the higher value of tanδ corresponds to higher stickiness levels as the 65-120 exhibited the highest value and follows by 50-120, 65-75 and 50-75, respectively. Surprisingly, at the frequency range of 1.53×10^{-3} (rad/s) - 6.17×10^{-2} (rad/s), the relation between the tanδ and the stickiness becomes opposite which means that the lower tanδ values represent higher sticky model foods. The correspondence frequency of stickiness and tanδ for 10-90 and 35-90 is at frequency values of lower than 2.99×10^{-4} (rad/s) in which the lower frequency represents the lower stickiness levels among model foods.

3.2.3.8 Complex viscosity (η*)

The complex viscosity (η*) is another parameter which can be obtained from a frequency sweep, and it is an indication of flow resistance of materials. It is the ratio of complex modulus (indicating the deformability) to the angular frequency (Tunick, 2011).

Figure 3-14 illustrates the evolution of the complex viscosity of model foods versus angular frequency. The data for all models shows a decrease in complex viscosity as the angular frequency increases which is a display of shear thinning behaviour. The same pattern occurs as is seen for the other rheological measurements with 10-90 and 35-90 having the highest complex viscosity, and 65-120 having the lowest viscosity. The other model food samples are located in the middle of these two extremes with slight variation

in order throughout the experiment: there is a very clear distinction between the samples comparing to their sensory stickiness levels.

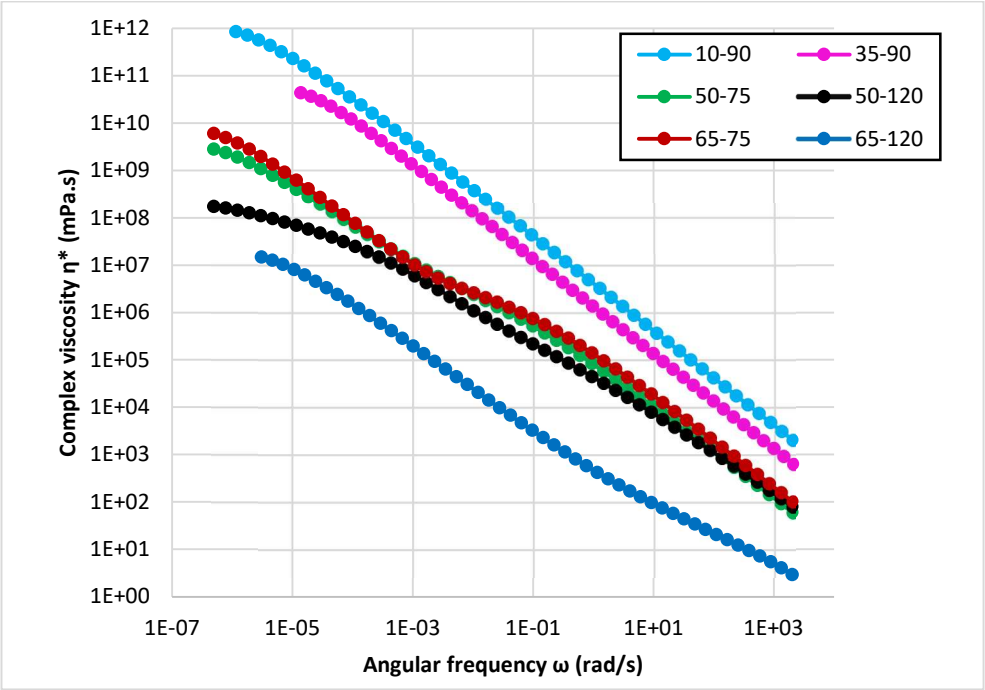

Figure 3-14. Complex viscosity as a function of angular frequency for model foods. The frequency dependency of all the model foods is apparent.

3.2.3.9 Discussion

As the data from these rheological experiments point to a similar pattern, the information has been discussed together so that overall conclusions can be drawn.

3.2.3.9.1 Relationship between the structure and the rheological properties

The data in Figure 3-12 and Figure 3-13 were used to determine the LVE. However, these figures also provide information about the strength of the samples by noting their crossover points. The crossover point indicates the limit of the strength of the internal structure of a material. As the sugar content increases to 50% (with the shorter cooking time), so the amount of shear strain required to break the internal bonds within the sample increases. As the stickiness increases with increasing sugar and cooking time, the

samples are more viscous, indicating that their structure does not form bonds which would give them a solid structure and therefore, cannot break during the oscillatory rheology measurement. The sugar content is related to stickiness as reported in section 3.2.1. Moll et al. (2022) experimented with biopolymer mixtures of pea protein and apple pectin powders. They found that samples with a liquid-like behaviour had a crossover point at higher shear strains, and had a higher stickiness, than samples that demonstrated more solid-like behaviour. This relation between the stickiness and the strength of the internal structure in food samples is the same as found with the model foods in this current research.

The data in Figure 3-12 and Figure 3-13 was used to determine the LVE. However, these figures also provide information about the samples' viscoelastic properties. Firstly, it can be seen from the data, that the elastic modulus decreases with increasing sugar and cooking time. This shows that the solid portion of the samples decreases. Secondly, the samples with lower sugar contents (or lower cooking time for the 50% sugar sample) all have an elastic modulus (G´) higher than the viscous modulus (G˝) in the LVE. This indicates that they will form internal bonds form to give the sample a solid-like character. As the sugar content increases, so the difference between the G´ and G˝ decreases indicating a less firm sample at rest. The higher sugar content samples (or higher cooking time for the 50% sugar sample) are all liquid-like at rest as inferred by their higher G˝. Thirdly, the crossover point, seen in the data for the lower sugar content samples indicates the strain required to start to break the internal bonds within the sample so it will start to flow as if it is a liquid like sample. The lowest sugar sample appears to be the most solid at rest, however, out of the three samples shown in Figure 3-12, the bonds are weakest as they require a lower strain to break them.

Furthermore, based on the LVE limit, model foods can be categorised as weak or strong gels. It has been suggested that weak gels have a narrow strain range comparing to those of strong gels (Ross-Murphy, 1995). In addition, the weak gel does not flow (Nishinari, 2009). It is also mentioned by Ahmad and Williams (1999) that the lower amount of sugar increases the storage modulus significantly. Therefore, 10-90 and 35-90 can be

categorised as weak gels and the other model foods fit into the category of strong gels as there is a considerable difference between the LVE limits of these categories. It was suggested by Grillet et al. (2012) that the less sticky materials are less deformable and demonstrate a brittle failure with making minimum amount of long behaviour in a tack test. This finding might be related to the model foods of the current thesis as the low sticky model foods make the minimum of long behaviour (necking) which is the opposite of high sticky materials with high necking and deformability.

Considering Figure 3-11 and Figure 3-13, higher tanδ with low G´ values are an indication of viscous behaviour (Chuang and Yeh, 2006). Tanδ reduction and approaching zero is an indication of storage modulus dominancy over viscous modulus (Grillet et al., 2012). It should be noted that lower tanδ might be an indication of a more structured sample or in other words a sample with higher levels of storage modulus (Seo and Yoo, 2013). What stands out about model foods 50-75, 50-120, 65-75 and 65-120 is their highly frequency dependency behaviour. They all show a minimum and maximum tanδ value from the start point frequency up to 9.69×10^{-2} (rad/s). In particular, for 50-120 and 65-75 minimum and maximum peaks correspond to multiple dominancy changes of G´ and G˝ in their responses to frequency shift. It can be related to the findings of a study by Ishihara et al. (2011a) which indicated diphasic behaviour of tanδ was linked to heterogeneous structure of polysaccharide gels and it was mentioned that differences in mechanical interaction of the gels lead to different relaxation times.

At these areas, short plateau regions are also observed which means the ratio of viscous modulus and elastic modulus remains constant and their behaviour is independent of applied frequency (Grillet et al., 2012). In a study by Renkema et al. (2002) about rheological behaviours of soy gels, reformation of protein bonds were linked to high values of tanδ. Considering the high sticky model foods (e.g., 50-120), it can be suggested that higher values of tanδ might be associated with lighter arrangement of microstructure of model foods and vice versa. It can also be discussed that beyond angular frequency of 0.04 (rad/s), this rearrangement of molecules becomes less and

less as the loss modulus dominancy increases. These results are in agreement with Grillet et al. (2012) who mentioned polymer gels with stiffer structures have higher elastic modulus and lower tanδ levels.

He et al. (2016) also linked the steep frequency dependency of complex viscosity of thickened liquids to weak gel behaviour. It was also mentioned by Moll et al. (2022) that the slope of reduction or increase of loss modulus might be an indication of the level of frequency dependency of materials. From Figure 3-12 it can be observed that both 10-90 and 35-90 have a steeper slope comparing to all other model foods which makes both very sensitive to the frequency increase. It can also be discussed that the low sticky model foods (10-90 and 35-90) represents a faster decrease in complex viscosity as well as larger frequency dependency compared to other model foods which again is an indication of their weak gel structure.

3.2.3.9.2 Relationship between the ingredients and the rheological properties

These differences in relaxation are probably due to the role of starch in the gelation of the model foods. Ozturk and Takhar (2017) reported that increased moisture content in oat flake with high levels of starch, samples lead to higher relaxation modulus or quicker relaxation behaviours. The effect of water is also related to its plasticizing effect on the cereal biopolymers and starch (Ismail et al., 2016). When starch is mixed with a plasticiser (e.g., water) and then goes under shear force (mixing) while heating (mainly higher temperatures for longer times), beside the huge effects of these processes on the physical state of the model foods, starch would enter a plastic state (Altayan et al., 2017, Fan and Roos, 2017). Gao and Zhou (2021) demonstrated that the bread bolus displayed a dominancy of storage modulus over loss modulus which is an indication of a solid-like structure. Apparent viscosity decreased dramatically for the bread boli during chewing. It was linked to the salivation effect on the plasticization of starch. The decrease of viscosity resulted softer boli as a consequence of higher water content (salivation) during mastication (Gao et al., 2017). Ross et al. (2019) indicated that viscosity at higher shear rates (such as 100 s^{-1}) might

provide a sufficient relationship with stickiness and mouthcoating attributes. The authors expressed that to have a clear insight about the stickiness, it should be considered with other measurements (e.g., tribological experiments).

The production method of model foods in the present research provides all the required elements of converting starch from its natural (crystalline) state into its plastic (gelatinised) phase. This is apparent mainly in model foods 65-120, 65-75 and 50-120, which have the highest amounts of viscous modulus compared to other model foods. As already indicated, temperature is important in the plasticization process in food matrixes. By increasing the temperature, it leads to decreased stiffness (hardness) and increased viscous flow as a consequence of higher molecular mobility (Fan and Roos, 2017) which plays a key role in the development of stickiness by increasing the deformability of food materials. The plasticizing effect of temperature is evident for different model foods. For example, model food 65-120 show higher levels of viscous flow compared to either 10-90 or 35-90.

It was reported by Jin et al. (2020) and Jin et al. (2021) that increasing the water absorption of the noodle samples resulted in higher stickiness values. This was carried out by adding wheat bran of varied sizes, in which finer bran particles led to smoother texture and consequently increased stickiness after cooking. Rheological measurements of the samples demonstrated that the storage modulus was greater than the loss modulus which exhibited a more elastic texture. Higher values of the tanδ were linked to stickier and softer doughs. The presence of extreme water amount in the dough resulted in decreased stickiness as which might be related to the reduced viscosity of the samples. The xylans presented in wheat bran are responsible for viscosity and water adsorption of dough and if their amount exceeds a threshold, they will interfere in the structure development of dough and consequently the resultant stickiness decreases. There are two parts of the above papers that can be discussed in relation to the present study. The first relation is the particle size and fragmentation of food samples in the mouth. Adding finer particles led to more cohesive and stickier samples, while increasing the particle size resulted in reduced stickiness and increased

fragmentation. In the current study, we observe that low sticky model foods (e.g., 10-90) tend to break into fragments by applying force either instrumentally or during oral manipulation. It is true that it is not concerning the particle size of the structure, but the fragmentation of the structure might be another parameter for their low stickiness. For the stickiest model foods such as 65-120, applying force led to the deformation of their structure rather than fragmentation. The second relation is the water content of model foods. As mentioned above, exceeding the amount of water content might be responsible for reduced stickiness among dough samples. Similar behaviour is also evident for the low sticky model foods that contain higher water content. Reddy et al. (1994) explained that in a more concentrated system, starch granules would absorb most of the water while in a more diluted system, starch is not able to absorb all the water content and the remaining amount of water would make a dispersing phase with a dominant viscous behaviour.

As previously discussed in section 1.1.3, low molecular weight sugars are produced during the prolonged heating time, and they are accompanied by high weight molecular polysaccharides (starch) which provides the parameters of a strong adhesion. Therefore, while the low weight molecular sugars as a part of the liquid-like part, flow into the surface (e.g., probe or mouth surface), the solid-like (high weight molecular polysaccharides) section favours the backbone of the adhesion.

3.2.3.9.3 Relationship between rheological parameters and stickiness

The Dahlquist criteria indicates that a sample with a high elastic modulus of above 10^5 Pa is unable to exhibit surface stickiness (Grillet et al., 2012). This is because, as the G´ gets closer to Dahlquist value, the physical structure of the material becomes less deformable and is subsequently unable to make a complete contact to be able to demonstrate stickiness. In Figure 3-8, the G´ for the 10-90 sample is highest among the model foods (10,000 Pa) and its stickiness is the minimum comparing to all model foods (see section 0).

Similarly, the Dahlquist criteria was used by Wang and Hartel (2021a) to categorise the tack level of different caramel samples. In their study, the caramel samples with the lowest tack had a G´ of 30,000 Pa, similar to that found in with the 10-90 sample (compared to other model foods).

It has been shown that samples with ingredients that would foster stickiness and which have viscous properties that deform and have a higher decrease in shear stress over shorter time periods are more likely to be highly sticky foods. Moll et al. (2022) show that their more liquid-like model samples made from pea protein and apple pectin demonstrated higher levels of stickiness comparing to rigid and solid-like samples. The flowability of their samples was a key factor for higher stickiness levels. Guan et al. (2020) experimented with different wheat doughs and found that higher strength and lower stickiness were found in doughs with a slower relaxation time and more elastic structures. This is shown in the results of the model foods of present study that a quicker relaxation behaviour is linked with more sticky samples such as 65-120 and the opposite for samples such as 10-90.

Caramel samples at low frequencies have enough time to rearrange that might be the reason for their deformation, while at higher frequency they do not have time to rearrange and subsequently results in a more solid-like behaviour (Ahmed et al., 2006). Therefore, the deformability of model foods can be related to the viscous dominancy at low frequency range, while having higher values of storage modulus over a big range of frequency range might explain the solid-like structure of model foods.

Regardless of increase or decrease of tanδ over the frequency domain, the above results indicate that higher values of tanδ are associated with higher stickiness levels. Comparing the model foods rating by the assessors (see section 3.1) with tanδ values, it can be noted that above the frequencies of 2.67 (rad/s), the higher value of tanδ corresponds to higher stickiness levels as the 65-120 exhibited the highest value and follows by 50-120, 65-75 and 50-75, respectively. Surprisingly, at the frequency range of 1.53×10^{-3} (rad/s) 6.17×10^{-2} (rad/s), the relation between the tanδ and the stickiness becomes opposite which means that the lower tanδ values represent higher sticky

model foods. The correspondence frequency of stickiness and tanδ for 10-90 and 35-90 is at frequency values of lower than 2.99 $\times 10^{-4}$ (rad/s) in which the lower frequency represents the lower stickiness levels among model foods.

To summarise this chapter, various rheological parameters of model foods have been discussed using the stress relaxation experiments. It was discussed that relaxation behaviour of model foods was related to the stickiness behaviour of model foods and their energy dissipation seems to be an important parameter in defining the levels of stickiness.

3.2.4 Rheological measurement in comparison to the texture analyser data

Texture analyser and rheological parameters as the instrumental measurements of the current study will be correlated in the current section. It is crucial to obtain reliable instrumental results that reflect the actual stickiness values. Therefore, the selection of valid parameters that are less influenced by other texture characteristics is of great importance.

Table 3-14 to Table 3-17 show the Pearson correlation coefficients relating rheology data to the texture analyser parameters. Significant differences are marked by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

The results of the correlation analysis of storage modulus and TA parameters are summarised in Table 3-14. What stands out in this table is the low R-squared for most of the parameters except for pre-area at lower angular frequencies with significant levels of $p < 0.05$. This could mean that there is no consensus among the parameters on a particular frequency at which they all give strong predictions. The other two parameters total-area and distance to adhesive peak also show moderate correlations.

The initial gradient provides the lowest correlation coefficients. What can be indicated about the initial gradient is that this might not be a powerful parameter in order to predict the rheological behaviour of the model foods. Section 3.2.1 mentioned the advantages of using this parameter to measure surface stickiness. Although reducing the effect of necking could be beneficial when measuring surface stickiness, it could be detrimental to the correlation of the initial gradient with rheological behaviour. This is consistent with the findings of Kazemeini and Rosenthal (2022) who originally introduced this parameter to minimise the effects of reduced contact area and strands formed during probe detachment. The importance of contact area was also highlighted by Yang et al. (2018) for various cooked rice samples. The authors suggested that the measured stickiness data should

be calibrated with respect to the contact area between the probe and the samples during detachment to ensure that the same conditions prevail for different physical conditions of the samples.

Another aspect of the data in Table 3-14 is that the maximum R-squared values for most parameters are found at lower frequencies. It can be seen that the only frequency range where significant differences occur are the lower frequency ranges belonging to the pre-area. It can also be seen that the pre-area, as a corrected form of the total-area, gives stronger correlations than the total-area, which means that the idea behind developing the pre-area to measure surface stickiness might be valid. At the same time, it should be kept in mind that the pre-area is a new parameter introduced by the current thesis and that further research needs to be conducted to understand the exact relationships between this parameter and the storage modulus.

Table 3-14. Pearson correlation coefficients relating storage modulus to texture analyser parameters. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

Angular frequency (rad/s)	**Initial Gradient**	**Force of the adhesive peak**	**Distance to adhesive peak**	**Pre-area**	**Total-area**
10^{-5}	0.34	0.51	0.70	0.72	0.68
10^{-4}	0.38	0.58	0.74	0.82*	0.78
10^{-3}	0.38	0.58	0.75	0.82*	0.78
10^{-2}	0.38	0.58	0.74	0.82*	0.78
10^{-1}	0.39	0.58	0.73	0.81	0.77
1	0.39	0.59	0.73	0.81	0.77
10	0.40	0.59	0.72	0.81	0.77
100	0.40	0.59	0.72	0.81	0.77
1000	0.40	0.59	0.72	0.80	0.76

From Table 3-14 it can be seen that all correlations are hardly influenced by the change in frequency. This is mainly due to the behaviour of the storage modulus of the model foods in response to the applied frequency range. At

the same time, most of the parameters of TA increased due to the increase in stickiness.

Table 3-15 shows the results of the correlation analysis of loss modulus and the TA parameters. The results show a similar trend compared to the storage modulus values (Table 3-14), but only in the lower frequency ranges. From Table 3-15, it can be seen that both the pre-area and the total-area have the highest number and significant correlation coefficients ($p < 0.05$) at low frequencies. Significant correlations at low frequencies support the goal of TA experiments where only surface stickiness and very low forces are used to prevent probe penetration into the sample and minimise necking.

Although the correlations are less strong at higher frequencies, the relationship becomes negative especially for the pre-area, the total-area and the distance to the adhesive peak.

Table 3-15. Pearson correlation coefficients relating loss modulus, to texture analyser parameters. significant differences are presented by single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

Angular frequency (rad/s)	Initial Gradient	Force of the adhesive peak	Distance to adhesive peak	Pre-area	Total-area
10^{-5}	0.41	0.63	0.77	0.89*	0.85*
10^{-4}	0.42	0.64	0.77	0.92*	0.87*
10^{-3}	0.39	0.58	0.73	0.80	0.76
10^{-2}	0.40	0.54	0.56	0.65	0.63
10^{-1}	0.53	0.57	0.11	0.41	0.39
1	0.53	0.45	-0.31	0.08	0.02
10	0.39	0.22	-0.43	-0.19	-0.35
100	0.13	-0.05	-0.22	-0.29	-0.52
1000	-0.04	-0.19	-0.06	-0.30	-0.54

The frequency dependence of the loss modulus compared to the TA parameters is more pronounced than that of the storage modulus. The loss modulus of the model food is very frequency dependent (see section 3.2.2),

which means that the negative correlations are poor, especially at higher frequencies.

Table 3-16 shows the results of the correlation analysis of the parameters tanδ and TA. The results are quite different compared to pre-area and the total-area. It can be seen that the initial gradient and the force of the adhesive peak provide significant ($p < 0.05$) and negative correlations at high frequencies. The next strong correlations are between the total-area and pre-area. All correlations at low frequencies are not significant, which means that tanδ at low frequencies has a very low relationship with the TA parameters.

Table 3-16. Pearson correlation coefficients relating tanδ, to texture analyser parameters. significant differences are presented by single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

Angular frequency (rad/s)	Initial Gradient	Force of the adhesive peak	Distance to adhesive peak	Pre-area	Total-area
10^{-5}	-0.16	-0.12	0.38	0.12	-0.06
10^{-4}	-0.11	-0.25	-0.01	-0.26	-0.46
10^{-3}	0.08	-0.20	-0.78	-0.61	-0.66
10^{-2}	0.19	-0.03	-0.89*	-0.53	-0.52
10^{-1}	0.00	-0.24	-0.82*	-0.69	-0.73
1	-0.60	-0.78	-0.46	-0.94**	-0.99**
10	-0.94**	-0.99**	-0.04	-0.86*	-0.85*
100	-0.99**	-0.98**	0.10	-0.77	-0.73
1000	-0.99**	-0.98**	0.14	-0.74	-0.70

Tanδ is the ratio of viscous modulus over elastic modulus with a more pronounced effect of loss modulus affecting the frequency dependency of the correlations. This is more pronounced at lower frequencies, while more constant and significant correlations are observed at higher frequencies. This means that tanδ is strongly associated with surface stickiness measured at higher frequencies. This is due to the physical properties of the model foods being more affected at lower frequencies.

Table 3-17 shows the results of the correlation analysis of the complex viscosity and the TA parameters. It can be seen that both pre-area and total-area are able to provide significant correlations ($p < 0.05$) at lower frequencies. Pre-area provides the highest number of significant correlations compared to all other parameters. This could indicate that tanδ can be strongly associated with pre-area at higher angular frequencies. The distance to adhesive peak gives moderately strong correlations, but the initial gradient and the force of the adhesive peak give the lowest correlations.

Table 3-17. Pearson correlation coefficients relating complex viscosity, to texture analyser parameters. Significant differences are presented by an asterisk (*) and a double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

Angular frequency (rad/s)	**Initial Gradient**	**Force of the adhesive peak**	**Distance to adhesive peak**	**Pre-area**	**Total-area**
10^{-5}	0.35	0.52	0.71	0.73	0.70
10^{-4}	0.38	0.58	0.74	0.81*	0.77
10^{-3}	0.40	0.61	0.76	0.85*	0.82*
10^{-2}	0.39	0.59	0.75	0.83*	0.79
10^{-1}	0.39	0.58	0.73	0.80	0.77
1	0.38	0.57	0.72	0.78	0.74
10	0.41	0.61	0.73	0.83*	0.79
100	0.41	0.60	0.72	0.81	0.77
1000	0.39	0.57	0.72	0.78	0.74

The complex viscosity is an indication of the flow resistance of model foods. Referring back to section 3.2.3.8; The lower the complex viscosity, the higher the stickiness. The complex viscosity is also related to the storage modulus as an indication of elasticity, which was highest for model foods with low stickiness (such as 10-90). Therefore, it can be assumed that the complex viscosity behaves similarly, as the storage modulus of the model foods was less frequency dependent.

It should be noted that the force of the adhesive peak provided the strongest correlation with sensory stickiness (see section 3.2.2), while it provided

moderate correlations with rheological parameters (with the exception of tanδ with strong correlations). One explanation for the moderate correlations could be the nature of the data recorded in these evaluations. While the sensory evaluation is done by dramatically changing the structure of model foods (e.g., by mechanical manipulation and the addition of saliva) and the subsequently recorded data come from the receptors in the mouth, only the stress and strain responses were recorded in the rheological experiments. It can be concluded that the addition of artificial saliva to the model foods during the rheology and TA experiments could have yielded stronger correlations. Future studies should take into account that instrumental measurements, especially rheological and TA methods, are limited to only physical measurements and mimicking the oral processing beyond only physical aspects should also be included in the experimental approaches.

The most surprising aspect of the correlation between TA and the rheological parameters is that the frequency range from 10^{-2} to 10^{-5} rad/s has the strongest values for the correlation of the rheological parameters (except tanδ) with pre-area and the total-area. This is partly consistent with the correlation of rheological and sensory parameters discussed in section 3.2.5. The main difference between them is that the TA parameters in the current section provide the strong correlations at lower frequency ranges, while the rheological parameters provide stronger relationships at wider frequency ranges from low to high values.

The correlations of TA with rheological parameters could explain the difference between the recorded data. While the TA experimental method was designed to record only the surface stickiness of model foods by reducing the effect of rheological parameters such as necking, the stress relaxation test for rheological experiments aimed to measure the behaviour of model foods at small to large deformations. This could also explain why the correlation for most TA parameters is moderate. Therefore, it can be suggested that the TA experimental method has more to do with low frequencies, as shown by the correlations with rheological parameters. In relation to TA and rheological measurements, the importance of stickiness as both a surface and bulk characteristic was highlighted by Godoi et al. (2017),

who pointed out that stickiness is related to higher viscosity values and that both surface properties (e.g., lubrication activities) and bulk properties of the sample should be considered.

To summarise this section, both TA parameters of pre-area and the total-area can be proposed as the most reliable parameters in relation to rheological measurements, as they show significant correlations ($p < 0.05$) at low frequencies. The introduction of the initial gradient and pre-area parameters in the current study was an attempt to measure food stickiness. Although initial gradient could not provide strong correlations with most rheological parameters, pre-area was the most reliable TA parameter as it provided stronger and significant correlations. Although conventional TA parameters have been considered easy to measure by researchers for decades, they have some serious measurement problems. Pre-area parameter showed that some improvements can be made by correcting a widely accepted parameter such as total-area. More detailed studies are needed to investigate the application of pre-area in different model food systems.

3.2.5 Rheological measurement in comparison to the sensory evaluation

The perception of stickiness has been linked to stress and strain responses during oral processing, without identifying specific receptors for the perception of stickiness in the mouth (Foegeding et al., 2015). This means that meaningful and important insights can be gained through correlations between sensory stickiness values and rheological parameters.

The results of the Pearson correlation analysis of the rheological parameters with overall sensory stickiness are summarised in Table 3-18. From these data, G' and complex viscosity are highly correlated with overall sensory stickiness, independent of angular frequency. This means that the overall sensory stickiness of model foods is predominately predicted by storage modulus and complex viscosity. The results are consistent with the findings of He et al. (2016) who found that the complex viscosity of thickened hydrocolloid-based samples was strongly correlated in the angular frequency range of 0.1-100 rad/s. It is worth noting that the limited frequency range applies to the results of the current study, as complex viscosity shows a high correlation with overall stickiness perception at all frequencies applied. Steiner et al. (2003) showed a high correlation between G' and the sensory measurement of stickiness of caramel. They suggested that such correlations could be useful for the development of rapid instrumental measurements.

Correlation analysis for both G″ and tanδ show a frequency dependency where G″ has higher correlations at lower angular frequencies (10^{-5} - 10^{-2} rad/s), while the opposite is true for tanδ (1 – 10^{3} rad/s). The constant strain rate of the mastication was linked to the perception of viscosity during oral manipulation of food products (Shama and Sherman, 1973). The highly viscous materials are only treated under constant strains during mastication. These results may be related to the possible oral manipulation of the model foods, where the higher sticky model foods (e.g., 65-120) could be chewed with higher consistency compared to the low sticky model foods under

constant strain. Therefore, the application of the stress relaxation method used in the present study may be associated with the processes during oral processing.

A closer look at the data in Table 3-18 shows that low frequencies are the range where the overall stickiness and the rheological parameters have the highest correlation.

Table 3-18. Pearson correlation coefficients relating rheological parameters, to overall sensory stickiness. Significant differences are presented by single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01 (both two-sided), respectively.

Angular frequency (rad/s)	G′ (Pa)	G″ (Pa)	Tanδ	Complex viscosity (mPa.s)
10^{-5}	-0.79	-0.89*	0.12	-0.80
10^{-4}	-0.85*	-0.90*	0.42	-0.85*
10^{-3}	-0.86*	-0.85*	0.59	-0.88*
10^{-2}	-0.85*	-0.75	0.47	-0.87*
10^{-1}	-0.85*	-0.55	0.67	-0.85*
1	-0.85*	-0.18	0.96**	-0.84*
10	-0.85*	0.19	0.88*	-0.87*
100	-0.85*	0.39	0.77	-0.85*
1000	-0.85*	0.44	0.74	-0.84*

When conducting sensory studies, it is important to consider other parameters besides stickiness. In a study by Vickers et al. (2015), 15 thickened drinks were prepared using different thickeners such as agar, waxy rice starch and guar gum. Textural properties such as thickness, stickiness, adhesiveness as well as mouth coating properties of the samples and number of swallows were categorised as adhesive properties. Positive correlations were found between these properties, and they were all strongly correlated with the flow index. Among these texture characteristics, two attributes of mouthcoating (Blok et al., 2021) and oral residue (Ross et al., 2019) were highly correlated with perceived stickiness. In addition, it was discussed that the stickiness and mouthcoating attributes were also correlated with food product thickness (Blok et al., 2021). Furthermore, the mouthcoating attribute was found to be closely related to the extensional

viscosity (He et al., 2016). These researchers emphasised the importance of other textural attributes instead of stickiness, which could be perceived as stickiness by the assessors. Therefore, it can be discussed that for a more comprehensive picture of stickiness, not only the measurement of this attribute should be considered, but also the measurement of other textural properties such as extensional viscosity. Although the aim of the current study was to measure only the stickiness and to try to minimise the influence of other textural properties on stickiness (e.g., by introducing pre-are to exclude the influence of rheological properties on the measurement of stickiness), this seems to be both an advantage and a limitation of the current study. Taking into account the limitations of the methods used, it may be suggested to use or develop instrumental measurements that are beneficial in measuring both surface and rheological properties of stickiness (e.g., performing compression and penetration tests with a texture analyser).

In the current study, all rheological measurements were made within the linear viscoelastic range. This may be a limitation of the current study, as Terpstra (2008) discussed that conducting experiments outside the LVE may provide a stronger significant correlation between rheological parameters and sensory evaluations. Therefore, it may be suggested for future studies to consider strain values that go beyond the LVE range, which could lead to more successful correlations between rheological parameters and sensory evaluations. This could be related to the high level of manipulation during oral processing, which aims to drastically change the structure of the food and prepare it for swallowing. In addition, temporal changes as well as viscoelastic behaviour of food during oral processing have been highlighted as important parameters to find more meaningful relationships between sensory and rheological studies (Joyner, 2018).

There are some unexplored aspects to the results of the present study. Steiner et al. (2003) reported that increasing the temperature of the product resulted in lower values of storage modulus and viscosity. The same applies to the loss modulus, which leads to lower consistency (Rincón et al., 2014). It follows that the model foods would differ more from each other at a mouth temperature of about 37 °C than at the temperature 25 °C used for the

rheological experiments in the current study. This could be one reason why the assessors were able to clearly discriminate the model foods compared to the rheological parameters.

In summary, storage modulus and complex viscosity showed the highest correlation with overall sensory stickiness. These results might be used to give an indication of stickiness or maybe for screening purposes but to replace the sensory evaluation measurement, predictive model needs to be developed. However, it should be kept in mind that such correlations are limited to the model foods systems used in the current study. Similar studies should be conducted for other food systems to confirm possible relationships between rheological parameters and sensory stickiness.

3.2.6 Bulk modulus

One of the first experiments to measure bulk modulus was to use potato as a sample. The potatoes were cut into cubes of one centimetre and then their bulk modulus was measured. The results showed non-significant differences ($p < 0.05$) between them (Table 3-19).

Table 3-19. Bulk modulus data by using potato as the sample. Tukey HSD post-hoc comparison of three replications shows that there are not significant differences between bulk modulus values ($p < 0.05$).

Replication	Sample weight (gr)	Oil weight (gr)	Sample volume (m^3)	Δv (m^3)	Oil movement in the tube (mm)	Bulk modulus (kPa)
Potato-1	43.50	38.49	4.05×10^{-5}	4.60×10^{-8}	36.00	88043.02[a]
Potato-2	38.92	34.24	3.60×10^{-5}	4.80×10^{-8}	38.00	75000.09[a]
Potato-3	45.43	39.75	4.18×10^{-5}	3.90×10^{-8}	31.00	107179.07[a]

Further experiments were conducted with cheddar cheese, banana, aubergine, apple and Rowntrees Fruit gums. There was a big variation in bulk modulus across different replicates of the same sample. Air bubbles in the structure seemed to be an important parameter affecting the results. This was evident in the aubergine, which has a spongy texture. As soon as the pressure was applied, the air bubbles started to leave the aubergine and reach the surface of the Duran bottle.

It was investigated how the bulk modulus changed when the model foods were changed at the same pressure. Bulk modulus measurement of model foods showed no significance difference between the samples ($p < 0.05$).

Table 3-20. Bulk modulus data of the model foods. Tukey HSD post-hoc comparison of three replications shows that there are not significant differences between bulk modulus values of model foods ($p < 0.05$).

Model food	Sample weight (gr)	Oil weight (gr)	Sample volume (m^3)	$\Delta\varpi$ ($m^{3)}$	Oil movement in the tube (mm)	Bulk modulus (kPa)
10-90	11.19	9.27	3.25E-07	3.74E-10	21.00	86898.40[a]
35-90	10.93	9.09	3.15E-07	3.87E-10	17.00	81395.35[a]
50-75	10.75	8.90	3.06E-07	3.93E-10	13.00	77862.60[a]
50-120	11.23	9.42	3.30E-07	3.72E-10	17.00	88709.68[a]
65-75	10.71	8.87	3.01E-07	3.96E-10	15.00	76010.10[a]
65-120	11.08	9.19	3.19E-07	3.80E-10	9.00	83947.37[a]

The values of bulk modulus were not following the sensory stickiness of model foods. The main reason for this could be the limited pressure resistance of the Duran bottles, which limited the maximum pressure applied to 100 kPa. This was also investigated in previous studies that the optimum applied pressure was 196 kPa for peach ripeness comparison and 1130 to 1640 kPa for peanut behaviour (Clark and Rao, 1977, Güzel et al., 2007).

Given the above constraints, many contacts were made with various departments at the University of Nottingham (e.g., the Chemistry Department) and some external people or institutes to provide the appropriate equipment for the bulk module experiments. The above contacts were successful in finding the more suitable apparatus for further bulk modulus experiments with pressures up to 30000 kPa. Unfortunately, it was not possible to carry out further experiments due to the limited budget of the project.

3.3 Electromyography

3.3.1 Electromyography data

The EMG technique allows the measurement of a wide range of parameters related to muscle activity that can be associated with the force, time and the rate of oral actions. It is important to remember that EMG is not a direct method of measuring oral processing. For example, various experimental parameters and factors such as electrode placement can influence the measured values, so EMG measurements provide relative data rather than absolute values (Gao and Zhou, 2021). These considerations are important when making correlations between EMG data with sensory attributes and physical measurements.

In this section, EMG results for different model foods are presented. Table 3-21 presents a statistical analysis of EMG characteristics (Fifteen pairs of model foods) using effect size (Cohen's d method). This analysis estimates the ability of the EMG features to discriminate the model foods. The table also include significant differences among model foods ($p < 0.05$ and $p < 0.01$). This table is constructed as follows: The two left columns show which pair of model foods were compared. The effect size is represented by differences in colour intensity. A more intense colour represents the higher values, and the lighter colours indicate the opposite. Significant differences among the pair of model foods are indicated by a single (*) and double asterisk (**) next to the values with a significance level of $p < 0.05$ and $p < 0.01$, respectively. For a clearer understanding of the effect size of the Cohen's d values, the magnitudes of the d values with the size of the reference effects are presented in Table 3-22. It can be seen that the differences in the d-values are indicated from 0.2 to 2 as small to large, respectively (Cohen, 1988, Sawilowsky, 2009).

Table 3-21 shows that there are distinct categories of EMG features based on their abilities to discriminate model foods. Thus, the EMG features of ChW, WR, pW, MV, AV and ChT result in at least eight significant pairs. The marked fact is that all such parameters (except ChT) correspond to the EMG data within the windows. The most distinct EMG feature is ChW which

provides ten significant pairs of model foods. This is followed by ChT with nine pairs. It is notable that for practical purposes ChT can be a feature of choice; this feature is uncomplicated to extract from the measurements and at the same time it is a powerful discriminator of textural attributes. The usefulness of chew time was highlighted by multiple researchers (Van der Bilt and Abbink, 2017, Puerta et al., 2020, Guo, 2021). Other features extracted from the EMG periods and counting (ACh, ChR, $IChT$ and BD) show low number of significant pairs which reflects a low ability to differentiate model foods.

It can be seen that ChW, WR, pW, MV, AV are the only parameters that can discriminate all model foods from 10-90 and 35-90 with an exception for the pair of 10-90/35-90. This pair of model foods can be differentiated by the EMG features of BD, ChT and NCh. Surprisingly, both BD and NCh are the EMG features with low differentiability. 10-90 and 35-90 which are the least sticky samples showed the maximum distinction by EMG features.

None of the EMG parameters were able to significantly discriminate the pairs of 50-120/50-75, 50-120/65-75 and 50-120/65-120. The non-significant effect was also true for the pair of 50-75 and 65-75 which might be an indication of low differences among these pairs of model foods shown by small Cohen's d values. EMG analysis provides useful insights into stickiness of model foods. Arguably, it might not be as discriminative as sensory analysis, but it provides important physiological underpinnings of oral processing and hence can provide valuable input to the development of correlative models for the purpose of predicting sensory perception based on physical methods. For example, Matsuyama et al. (2021) used suprahyoid muscle activity measured by EMG and two other methods (tongue pressure measurement and laryngeal movement) to predict the textural perception prior to swallowing. By using these combination of methods for hardness perception, they gained very high accuracy compared to both TA and sensory evaluation.

Table 3-21. Pairwise comparison of model foods using effect size (Cohen's d). The higher colour intensity represents greater effect size and significant differences are presented by single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01, respectively. The complete name of EMG abbreviations is as follow: ChW: Total Chew Work, WR: Chew Work Rate, pW: proportional Work, ACh: Average duration of Chews, NCh: Number of Chews, ChT: Chew Time, ChR: Chew Rate, IChT: Interchew Time, BD: Burst Duration, MV: Maximum Voltage Peak Amplitude, AV: Average Voltage Peak Amplitude, MFS: Median Frequency Shift.

Model food pairs		EMG features											
		ChW	***WR***	***pW***	***ACh***	***NCh***	***ChT***	***ChR***	***IChT***	***BD***	***MV***	***AV***	***MFS***
10-90	**35-90**	1.06	0.49	0.20	0.03	0.49*	0.53*	0.15	0.14	0.61*	0.62	0.65	0.03
10-90	**50-75**	1.75*	1.09*	0.40*	0.11	0.72*	0.89*	0.15	0.01	0.75*	1.20*	1.35*	0.03
10-90	**50-120**	2.26*	1.12*	0.44*	0.13**	1.13*	1.28*	0.24	0.03	0.75*	1.53*	1.62*	0.05
10-90	**65-75**	2.14*	1.12*	0.46*	0.02	1.09*	1.12*	0.08	0.10	0.83*	1.46*	1.51*	0.06
10-90	**65-120**	2.24*	1.29*	0.47*	0.11*	1.08*	1.32*	0.39*	0.00**	0.74*	1.72*	1.80*	0.13
35-90	**50-75**	0.75*	0.57*	0.24	0.25	0.20	0.35	0.01	0.21	0.19	0.52	0.65*	0.06
35-90	**50-120**	1.22*	0.62**	0.30	0.31	0.54	0.80*	0.11	0.29	0.16	0.79*	0.85*	0.09
35-90	**65-75**	1.06*	0.63*	0.33	0.11	0.50	0.61	0.10	0.09	0.21	0.73*	0.75*	0.03
35-90	**65-120**	1.39*	0.78*	0.34*	0.27	0.57**	0.89*	0.32	0.29	0.17	1.04*	1.03*	0.11
50-75	**50-120**	0.43	0.07	0.23	0.04	0.35	0.52	0.11	0.06	0.04	0.24	0.17	0.03
50-75	**65-75**	0.26	0.09	0.35	0.17	0.30	0.30	0.10	0.14	0.01	0.20	0.07	0.09
50-75	**65-120**	0.71*	0.25	0.39	0.01	0.40	0.64*	0.32	0.02	0.02	0.55**	0.36	0.18
50-120	**65-75**	0.19	0.02	0.15	0.22	0.06	0.22	0.21	0.22	0.04	0.05	0.10	0.13
50-120	**65-120**	0.32	0.17	0.21	0.05	0.10	0.17	0.22	0.05	0.02	0.35	0.20	0.23
65-75	**65-120**	0.50**	0.15	0.11	0.18**	0.15	0.37**	0.42**	0.20	0.02	0.38	0.30	0.07

Table 3-22. The Cohen's d effect size with a reference to the difference they make (Cohen, 1988, Sawilowsky, 2009).

d	0.2	0.5	0.8	1.2	2
Difference	Small	Medium	Large	Very large	Huge

As mentioned above, some of the pairs of model foods are not significantly different and one of the reasons might be the type of the muscles that were used to record the chewing behaviour in this study. Temporalis and masseter compared to digastric muscles. Both temporalis and masseter are responsible for closing the jaw during chewing. The other group of muscles which might provide useful information about the opening phase of chewing is digastric (Ishihara et al., 2011b, Çakir et al., 2012). The activity of digastric muscles was recorded in the current study, but the recorded data was not useable due to the high levels of recorded noise. One of the reasons for that can be that the electrodes were not placed in the right location and, subsequently, the recorded values were marred by high noise. Although multiple pilot trials were performed prior to the main study to make sure of the correct attachment of the electrodes as well as the ability of the principal researcher to perform the EMG study, the low quality of the data obtained from digastric muscles was highlighted after advanced analysis by the EMG expert. For future studies, it can be suggested to use more accurate anatomical landmarks and palpation. Another point of attention about the quality of the data is the difference between sEMG and needle EMG. By employing sEMG as used in the current study, the activity of the total group of muscles is recorded which may reduce the accuracy of the recorded data. In contrary, needle EMG, which is more applicable in neurology studies, can provide accurate data regarding the activity of single muscles but due to invasiveness of the method it will significantly change the habitual chewing of assessors.

Remarkably, there were significant differences observed in 11 EMG parameters between the pair of 10-90 and 65-120, which stood out as the most distinct pair among the model foods. Consistent with sensory evaluation, the pair of 10-90 and 50-120 exhibited significant differentiation across nine EMG features. Additionally, it is worth mentioning that both model foods 50-75 and 65-75 displayed differentiation across eight EMG parameters.

The features extracted from a chewing window (ChW, WR, MV and AV) are closely related to each other, as they are all related to the extracted

information from muscle activity. The variations among EMG features are well-documented in the literature (Kohyama et al., 2010, van Eck et al., 2019, Pu et al., 2021). In order to reduce such variations, it has been suggested to apply controlling measures on the food samples as well as increasing controllable parameters such as highlighting specific texture attributes and develop model foods with varied levels of that attribute (e.g., hardness) (Woda et al., 2006a). Kohyama et al. (2017) emphasized the significance of EMG features in comparison to sensory evaluation, as they serve as a valuable tool for researchers to quantitatively assess the chewing process of foods, particularly by capturing oral muscle activity.

Table 3-23 shows the Spearman correlation coefficients relating EMG features, to stickiness rating at each time interval and overall stickiness. It can be seen that most of the EMG parameters provide significant correlations with sensory data. On the other hand, four EMG parameters of BD, ChR, $IChT$ and ACh do not show any significant correlations with sensory evaluations. With the exception of ChR as a numerical parameter, all other parameters are time-related values. This finding implies that the mentioned parameters may not have a direct impact on the oral perception of stickiness. The absence of significant correlations prompts further investigation to uncover the underlying reasons. It is plausible that these parameters primarily capture aspects of chewing that are indirectly associated with the sensory experience, such as the mechanical efficiency or coordination of jaw movements. Further investigation is necessary to enhance comprehension of their individual roles and potential impact on the overall perception of stickiness.

The effect of stickiness in longer oral processing times was reported by Gao et al. (2015) for steamed breadcrumbs. The prolonged oral processing time of stickier breadcrumbs was associated with two activities, namely the assessors transferring the bolus around the mouth and cleaning the oral surfaces during chewing. A similar profile was also discussed by Cheong et al. (2014) for varied formulations of biscuits. These are consistent with the results of the current study as chew time had a positive relation with stickiness. While the current study did not measure specific oral processing

parameters such as cleaning oral surfaces for the model foods, it is worth considering this aspect as a potential contributing factor to the extended oral processing time observed with stickier model foods. Sticky foods tend to adhere to oral surfaces, including teeth, gums, and the palate, which may require additional efforts to remove and clear the mouth during the chewing process. The time spent on oral cleaning and clearance could potentially contribute to the overall longer oral processing time of sticky foods.

Table 3-23. Spearman correlation coefficients relating EMG features, to stickiness rating at each time interval and overall stickiness. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

EMG Features	Time intervals					Overall Stickiness
	5 s	10 s	15 s	20 s	25 s	
ChW	0.97**	0.96**	0.97**	0.97**	0.98**	0.97**
WR	0.94**	0.93**	0.94**	0.95**	0.95**	0.95**
BD	0.73	0.72	0.72	0.72	0.72	0.75
pW	-0.92**	-0.91*	-0.91*	-0.91*	-0.89*	-0.92**
NCh	0.92*	0.91*	0.91*	0.91*	0.90*	0.92**
ChR	-0.41	-0.42	-0.44	-0.44	-0.52	-0.44
IChT	0.05	0.06	0.07	0.07	0.13	0.07
ACh	0.47	0.47	0.49	0.49	0.55	0.50
ChT	0.91*	0.91*	0.91*	0.92*	0.91*	0.92**
MV	0.96**	0.95**	0.95**	0.96**	0.96**	0.96**
AV	0.96**	0.95**	0.95**	0.96**	0.96**	0.96**

It is important to acknowledge that there are alternative methods for measuring stickiness beyond the utilization of EMG and sensory evaluation. In an interesting article by Devezeaux de Lavergne et al. (2015a), stickiness and cohesiveness were assessed based on the ease or difficulty of swallowing. This unique approach offers a different perspective on evaluating stickiness, focusing on the functional aspect of food texture and its impact on the swallowing process. Incorporating a similar approach in the current study

could have provided valuable insights into the model foods and their oral processing behaviour, complementing the range of instrumental and sensory methods employed to measure stickiness. Assessing the ease or difficulty of swallowing provides a holistic view of the sensory and motor aspects involved in the oral processing of sticky foods. By incorporating such an approach, it might be possible to gain insights into the effects of stickiness on swallowing kinetics, potential discomfort or challenges encountered during the swallowing process, and overall oral processing behaviour. This approach could also shed light on the potential challenges faced by individuals when consuming sticky foods, including any potential alterations in chewing patterns, extended oral transit time, or variations in the effort required for successful swallowing.

Based on the data presented in Table 3-21 and Table 3-23, it can be inferred that Chew Rate does not demonstrate differentiation between the model foods and does not exhibit significant correlations with the sensory ratings. As a result, it is reasonable to suggest that Chew Rate may not serve as a valuable parameter for evaluating model foods in the current study that primarily consist of sugar and starch. Çakir et al. (2012) suggested the use of average Chew Rate for characterising food texture. In an attempt to study Chew Rate, Van der Bilt and Abbink (2017) investigated the oral manipulation of varied type of food products by segmenting the mastication process into different phases. The authors considered the distinguishability of EMG parameters with regard to different phases of mastication. The Chew Rate of different phases was reported to be more relevant to the mastication process than the average Chew Rate of the whole period of chewing. This can offer an explanation why the Chew Rate is not able to discriminate model foods considering the sensory stickiness values. It can also be mentioned that there might be other contributing factors to stickiness that cannot be measured by Chew Rate. Similar approaches were also suggested by Wagoner et al. (2016) to consider discrete stages rather than average values.

Another possible reason for low distinguishability of Chew Rate might be related to the contribution and feedback from the brain stem. It was

speculated that the constant Chew Rate among varied food textures might be due to the nature of mastication as a complex and rhythmic motor behaviour which is regulated by the neurons located in the brain stem, making Chew Rate to be independent of textural attributes of foods (Jean, 2001, Aguayo-Mendoza et al., 2019).

Based on the analysis of Table 3-21, which presented both Cohen's d values and significant differences ($p < 0.05$, $p < 0.01$) among the pairs of model foods, a selective approach was employed to narrow down the focus to a subset of EMG parameters. Given the large number of parameters in the current study, only those that demonstrated either significant differences between the model foods (in at least 6 pairs of model foods) or higher Cohen's d values were deemed relevant for further analysis. The parameters considered for further investigation are ChW, WR, NCh, ChT, MV, and AV. These parameters exhibited substantial differentiation or effect sizes, indicating their potential significance in capturing meaningful distinctions in the chewing patterns and muscle activity among the model foods.

Table 3-24 provides valuable insights into the variations among individual assessors when utilizing EMG parameters during the assessment of different levels of sticky model foods. This table sheds light on the diverse chewing behaviours exhibited by the assessors in their habitual chewing manners. Notably, the results demonstrate significant differences among the assessors across various EMG parameters. Among the observed variations, it is interesting to note that the parameters ChW and ChT exhibit the minimum significant differences between assessors. This implies that, to a certain extent, the assessors demonstrate consistency in these particular aspects of their chewing patterns and muscle activity when confronted with different levels of stickiness in the model foods. Furthermore, a closer examination of the data reveals that assessor a6 stands out that exhibits significantly higher values for ChR, MV, and AV compared to the other assessors. This suggests that assessor a6 tends to have a faster chewing rate and greater muscle activity during the chewing process. On the other hand, the value of ChW is significantly lower than that of the other assessors, indicating a potentially

different chewing behaviour or muscle activity in terms of chewing window duration.

Table 3-24. The values of EMG parameters for individual assessors (each value is the average of 4 replications). Tukey HSD post-hoc comparisons provide information on differences between assessors. Equal assigned letters mean that there are no significant differences between these data ($p < 0.05$).

Assessors	ChW	WR	NCh	ChT	MV	AV
a1	1494.31^{b}	0.09^{c}	26.63^{a,b}	10.69^{a,b}	0.58^{a,b,c}	0.23^{a,b,c}
a2	1539.68^{b}	0.08^{b,c}	28.88^{b,c}	11.73^{b,c}	0.54^{a,b,c}	0.30^{c}
a3	1303.14^{a,b}	0.07^{b,c}	29.78^{b,c,d}	8.43^{a}	0.42^{a}	0.22^{a,b}
a4	1492.27^{b}	0.06^{a,b}	31.3^{b,c,d,e}	11.30^{b}	0.45^{a}	0.24^{a,b,c}
a5	1681.74^{b}	0.07^{a,b,c}	36.00^{d,e,f}	14.11^{c}	0.50^{a,b}	0.27^{b,c,d}
a6	876.72^{a}	0.12^{d}	38.00^{f}	16.66^{c}	0.87^{d}	0.56^{e}
a7	1859.95^{c}	0.05^{a}	21.83^{a}	8.28^{a}	0.53^{a,b,c}	0.19^{a}
a8	1346.60^{b}	0.06^{a,b}	34.08^{c,d,e,f}	12.65^{b,c}	0.68^{c}	0.29^{b,c,d}
a9	1269.78^{a,b}	0.05^{a}	36.81^{e,f}	12.59^{b,c}	0.47^{a,b}	0.19^{a}
a10	1596.22^{b}	0.08^{b,c}	32.56^{b,c,d,e,f}	11.34^{b,c}	0.63^{b,c}	0.32^{d}

It is interesting to mention that although a5 and a6 do not have significant differences in their ChW, they demonstrate significant differences for NCh. This observation highlights the complex relationship between muscle activity and chewing behaviour. Despite exerting similar levels of Chew Work, these assessors may rely on different muscular strategies or chewing patterns to process model foods. It might also emphasize the multidimensional nature of chewing behaviour (Matsuyama et al., 2021). While ChW reflects the overall work performed by the masticatory muscles, NCh provides insights into the frequency and duration of individual chews. The significant differences in muscle activity despite a similar number of chews indicate that assessing chewing performance based solely on the number of chews may not capture the full complexity of muscle engagement and coordination required for processing sticky foods (Van der Bilt and Abbink, 2017).

It was suggested by Maeda et al. (2020) that textural attributes such as cohesiveness are mainly affected by the duration of chewing. Although, cohesiveness of the bolus was not measured in the current study, it can be mentioned that cohesiveness can influence the breakdown and processing of

sticky foods, as it affects the resistance encountered during chewing. Assessors who demonstrate higher muscle activity but similar number of chews (as observed in a5) may encounter greater resistance due to higher cohesiveness of the food. This increased muscle activity can be attributed to the need to exert more force and work to break down the cohesive mass and achieve proper oral processing. On the other hand, assessors who exhibit lower muscle activity but similar number of chews (as observed in a6) may encounter lower cohesiveness in the sticky foods they evaluate. With less cohesive food particles, they may require less muscle effort to achieve adequate breakdown, resulting in lower muscle activity despite a similar number of chews. Further investigations into the relationship between stickiness, cohesiveness, muscle activity, and number of chews can provide valuable insights into the sensory perception and oral processing of sticky foods (Young et al., 2013, Wagoner et al., 2016, Wang and Hartel, 2021a).

The analysis of the data presented in the current section reveals important findings regarding the differentiation among model foods and assessors based on the ChW and ChT parameters. These parameters exhibited notable characteristics that make them worthy of further detailed analysis and investigation. Firstly, in terms of the differentiation among model foods, both ChW and ChT demonstrated the most significant differences. This indicates that these parameters were highly sensitive in capturing distinctions between the different types or levels of model foods. The high Cohen's d effect size associated with these parameters further supports their effectiveness in discriminating between the model foods based on their chewing patterns and duration. Furthermore, in terms of assessing inter-individual differences among the assessors, ChW and ChT showed the minimum significant differences. This suggests that these parameters provide a relatively consistent assessment of chewing behaviour across different individuals.

Further analysis of the Chew Time data, using ANOVA, provides more detailed insights into the model foods. The results presented in Table 3-25 reveal significant differences among the model foods from the 15-second time interval onwards ($p < 0.05$).

Table 3-25. The results of multivariate generalised linear model of Chew Time. It can be seen that at 5 and 10 second time intervals the model foods are not significantly different from each other. The model foods become significantly different from each other from 15 to 25 s ($p < 0.05$).

Time interval (s)	Mean Square	F	Sig.
5	0.380	1.080	0.384
10	0.304	1.191	0.328
15	0.981	4.085	0.004
20	1.590	4.639	0.002
25	2.132	4.633	0.002

To explore the specific model foods that exhibit significant differences from one another, Tukey HSD post hoc test was conducted (Table 3-26). Upon examining the table, it becomes evident that there are fewer significant differences observed between the model foods, particularly during the initial time intervals. This implies that model foods may share similarities in terms of their chewing patterns and temporal characteristics during the early stages of oral processing. However, as the time intervals progress, more distinct variations start to emerge among the model foods, indicating divergent chewing behaviours and durations.

Table 3-26. Chew Time extracted values from chewing sequence (each value is the average of 10 assessors and 4 replications), values are presented as mean (SD). Tukey HSD post-hoc comparisons provide information on the differences between the model foods. Equal assigned letters mean that there are no significant differences between these data ($p < 0.05$).

Model food	Sensory stickiness rating	5 s	10 s	15 s	20 s	25 s
10-90	0.24^{a}	2.53^{a} (0.57)	2.36^{a} (0.56)	1.69^{a} (0.64)	0.99^{a} (0.79)	0.21^{a} (0.32)
35-90	2.29^{b}	2.71^{a} (0.53)	2.52^{a} (0.51)	2.43^{a} (0.43)	1.64^{a} (0.44)	$0.86^{a,b}$ (0.90)
50-75	4.76^{c}	3.04^{a} (0.71)	2.91^{a} (0.48)	$2.52^{a,b}$ (0.44)	$1.86^{a,b}$ (0.59)	$0.71^{a,b}$ (0.49)
50-120	7.97^{e}	2.98^{a} (0.65)	2.69^{a} (0.51)	2.58^{b} (0.55)	1.95^{b} (0.54)	1.30^{b} (0.71)
65-75	6.49^{d}	2.95^{a} (0.64)	2.71^{a} (0.54)	2.38^{b} (0.48)	1.88^{b} (0.61)	1.34^{b} (0.83)
65-120	9.04^{f}	2.65^{a} (0.39)	2.72^{a} (0.39)	2.47^{b} (0.29)	2.20^{b} (0.42)	1.48^{b} (0.65)

Table 3-26 provides interesting insights into the chewing times for the different model foods at various time intervals. A notable observation is that, after the initial 10 seconds, the chewing times for the model foods 50-120 and 65-120 appear to dominate when compared to 50-75. This implies that the chewing process for the former two model foods requires more time and effort in subsequent intervals. Initial texture of the food may have contributed to the longer chewing time seen for the model food 50-75 during the first 10 seconds. It is conceivable that extra processing and mastication are needed in order to effectively process the initial texture of 50–75. As a result, each 5-second period is given more time so that the initial texture of 50–75 can be processed completely. The chewing times for 50-120 and 65-120 exceed those for 50-75 as the chewing process progresses through the first stages, indicating that these model foods may have distinct textural characteristics or require different amounts of mastication to accomplish a similar level of oral processing.

Interesting information about the model foods is discovered through the examination of Chew Time measurements. Chew Time varies at both 5 and 10 seconds, although these variations are not statistically significant. This suggests that, regardless of their levels of stickiness, the model foods exhibit comparable chewing durations during the initial phases of oral processing. But when the chewing process goes on through the early intervals, it becomes clear that the model foods' chew times range significantly from one another. These differences allow for a broad categorization of the model foods into two significant groups: low stickiness and high stickiness. The low stickiness category comprises primarily the model foods 10-90, 35-90, and 50-75, which exhibit relatively shorter chewing durations compared to the high stickiness model foods. On the other hand, the high stickiness category consists of the model foods 65-75, 50-120, and 65-120, which demonstrate longer chewing times. It is significant to note that the present classification of stickiness information is restricted and only offers a broad comprehension of the general pattern shown in the model foods. Although the classification of the samples into low and high stickiness samples presents a general

contrast, it does not give precise information about the various levels of stickiness.

Another interesting aspect of Chew Time parameter is its possible relationship with how the texture of the model foods is perceived. Van der Bilt and Abbink (2017) linked the higher oral processing time with more sophisticated food textures. According to the authors, compared to a food item with a simpler texture, assessors should begin chewing a more complex food structure more slowly and cautiously. This is consistent with this study's findings, which show that the assessors chew for the longest amount of time in the beginning of mastication and then lessen as chewing progresses. There is a difference between the above study with the current study in which Van der Bilt and Abbink (2017) related the hardness (toughness) of the samples to the reduced oral processing time, while here it is hypothesized to be related to stickiness. It is also important to consider that the chewing responses of the assessors may not only be related to stickiness, but also to other textural properties. This point was highlighted in a review study by Tonni et al. (2020). The majority of the papers they looked at made it clear that the chewing reaction was connected to a variety of textural aspects, not only the ones that affected the study's features. It is also significant because factors like the bolus's ability to soften through salivary secretion is a crucial factor in changing texture when chewing, with the degree to which these changes are influenced by the composition of the food samples (Le Reverend et al., 2016).

Furthermore, it is worth noting that model foods 50-120 and 65-120 exhibit similar rheological behaviours (as discussed in section 3.2.2) and are rated similarly by texture analyser parameters (as outlined in section 3.2.1). Additionally, it is interesting to observe that although 65-120 has a longer overall mastication time compared to 50-120, this does not imply that it consistently maintains higher Chew Time throughout the entire mastication process. This observation underscores the dynamic nature of mastication and the rapid changes in stickiness that occur during oral processing.

In an interesting study by van Eck et al. (2019), the stickiness of bread and crackers was defined by sensory evaluation, while their adhesiveness was measured by the TPA method as the area under the negative curve of the first detachment. Higher values of both sensory stickiness and adhesiveness measured by the TPA resulted in longer mastication times. Adding different toppings, such as cheese spread, to the samples shortened the retention time in the mouth. In contrary, Zhu et al. (2013) stated that higher viscosity of semi-solid foods (varied mixtures of chocolate pudding and heavy cream to produce high and low viscosities) could lead to slower chew rate. The longer oral processing might be related to the stickiness which was measured in this study. Notably, Wee et al. (2018) reported that the more adhesive foods were consumed faster by having bigger bites. There were significant correlations between the water content of the food samples and their stickiness. They suggested that a higher water content in the food samples shortened oral processing time by decreasing saliva flow. Therefore, it can be assumed that an acceptable level of stickiness is helpful in controlling food intake by slowing down oral processing. This suggestion was made by Bolhuis and Forde (2020) when it came to developing specific foods to control food intake in vulnerable individuals. This article addresses how stickiness can help in the development of tailor-made foods. In light of the above discussion, the current thesis highlights the value of a comprehensive understanding of stickiness as a complex textural attribute and how a broader and deeper understanding of this attribute can help in the development of food products.

In order to gain deeper insights into the characteristics of the model foods, further analysis was conducted on the Chew Work data. This analysis involved performing an ANOVA to examine the differences in Chew Work among the model foods (Table 3-27). The table reveals that significant differences exist among the model foods at all time intervals, as indicated by the p-values of less than 0.05. This implies that the model foods exhibit distinct Chew Work values throughout the entire duration of the mastication process.

Table 3-27. The results of multivariate generalised linear model of Chew Work. It can be seen that at all time intervals there are significant differences among model foods ($p < 0.05$).

Time interval (s)	Mean Square	F	Sig.
5	8.15	6.12	<0.001
10	6.70	5.76	<0.001
15	6.62	6.66	<0.001
20	4.96	6.10	<0.001
25	3.35	6.05	<0.001

In order to further explore the significant differences in Chew Work among the model foods, a Tukey HSD post hoc comparison was conducted, as presented in Table 3-28. This post hoc analysis provides a more detailed understanding of the specific time intervals at which significant differences in Chew Work exist among the model foods.

The post hoc analysis's findings show that the model foods have significant differences in terms of their Chew Work values. First off, there are significant variations between the model food 10-90, which has the lowest sensory stickiness, and the stickiest model foods, 50-120 and 65-120. The findings imply that, in comparison to the more adhesive and sticky properties of 50-120 and 65-120, the reduced stickiness of 10-90 demands less effort during chewing. Furthermore, it is shown that both 50-120 and 65-120 exhibit significant variations in chew work during the chewing process when compared to the majority of the model foods. These significant variations set these high-stickiness model foods apart from other model foods by indicating that they demand more chew work during mastication. The large variations in chew work provide more evidence that stickiness is a key factor in influencing the mechanical effort required for oral processing.

At the 5 second time interval, model foods are just at the beginning of their mastication process, and it can be observed that Chew Work represents a positive correlation with the stickiness. Similar positive relationships have been suggested for other textural attributes (e.g., hardness of bread) with chewing efforts (Foegeding et al., 2010, Gao et al., 2018), suggesting that the muscle activity might represents the texture of food materials. However, it is noteworthy that as the chewing process progresses and reaches later

stages, such as 20 seconds and 25 seconds, a reduction in the number of significant differences in Chew Work can be observed. This implies that as the mastication process advances, the initial distinctions in Chew Work between certain model foods tend to diminish or become less pronounced.

Table 3-28. EMG Chew Work extracted from chew sequence (each value is the average of 10 assessors and 4 replications), values are presented as mean (SD). Tukey HSD post-hoc comparisons provide information on differences between model foods. Equal assigned letters mean that there are no significant differences between these data ($p < 0.05$).

Model foods	Sensory stickiness rating	5 s	10 s	15 s	20 s	25 s
10-90	0.2^{a}	1.28^{a} (0.84)	1.20^{a} (0.74)	0.79^{a} (0.54)	0.65^{a} (0.54)	0.15^{a} (0.07)
35-90	2.3^{b}	$1.90^{a,b}$ (0.71)	1.88^{a} (0.60)	1.68^{a} (0.69)	$1.13^{a,b}$ (0.72)	$0.58^{a,b}$ (0.76)
50-75	4.8^{c}	$2.72^{a,b,c}$ (0.81)	$2.60^{a,b}$ (0.74)	$2.30^{a,b}$ (0.74)	$1.66^{a,b,c}$ (0.79)	$0.63^{a,b}$ (0.59)
50-120	8.0^{e}	3.25^{c} (1.09)	2.84^{c} (0.96)	2.69^{c} (1.01)	$1.97^{b,c}$ (1.01)	$1.31^{b,c}$ (0.93)
65-75	6.5^{d}	$2.97^{b,c}$ (1.12)	$2.68^{a,b}$ (1.09)	$2.17^{a,b}$ (1.02)	$1.76^{a,b,c}$ (0.94)	$1.22^{b,c}$ (0.95)
65-120	9.0^{f}	3.16^{c} (0.96)	3.12^{c} (0.95)	2.77^{c} (0.91)	2.39^{c} (0.89)	1.56^{c} (0.81)

Table 3-28 shows that Chew Work decreases in the final stages of chewing for all model foods, but the decrease is more pronounced for the less sticky model foods. The table demonstrates that model foods 10-90 and 35-90 have a greater reduction (although not significantly different) in Chew Work compared to their initial values. It can be surmised that these model foods tolerate less mechanical manipulation during oral processing and have a sensitivity to salivary enzymatic activity, particularly starch degradation (Le Reverend et al., 2016, Mosca and Chen, 2017). While starch breakdown during chewing was not measured in the current study, the literature suggests that the longer oral processing times maximise starch breakdown. Therefore, stickiness would decrease (Engelen et al., 2003) which could also be due to the product dependence of oral starch breakdown (Mosca and Chen, 2017). Although this might be the case to some extent for the stickier model foods such as 65-120 and 50-120, it might be limited for the low

stickiness model foods such as 10-90 and 35-90. The low stickiness model foods have a short oral processing time which minimises the effects of starch degradation, while other parameters such as dissolution and physical manipulation of the model foods have a stronger effect compared to oral starch breakdown. This could also be related to the way they are manipulated during chewing. As indicated by Ishihara et al. (2011b), a soft gel is mechanically broken down in the mouth by being squeezed between the tongue and palate rather than being chewed by the teeth. This could explain the low intensity of the recorded EMG values, mainly for 10-90. Another possible explanation could be that the degree of reduction in Chew Work reflects the texture of the bolus in the mouth. Looking at the values of Chew Work for model foods with low stickiness, there is an extreme reduction, which may indicate a very soft bolus, while higher values of Chew Work for stickier model foods indicate a more structured bolus. A similar continuous reduction of Chew Work was also reported for cheese, cake and carrot (Van der Bilt and Abbink, 2017).

In a study conducted by Kohyama et al. (2005), a similar trend regarding the influence of water content on mastication effort was observed. This study emphasized the role of water as a key parameter affecting the mechanical effort required during mastication. The findings indicated that rice samples with higher water content exhibited lower mastication efforts. The results from Kohyama et al. (2005)'s study can help explain the behaviour of the model foods in the current study. Specifically, the model foods 10-90 and 35-90, which have higher water content, were perceived as having low stickiness based on the sensory data (see Table 2-2 for the water content of the model foods). Conversely, the model foods with lower water content, such as 65-120, showed the opposite pattern. These model foods were perceived as having higher stickiness in terms of sensory evaluation, which correlates with the increased Chew Work observed. The lower water content in these model foods may have contributed to their stickier texture and subsequently required greater mastication effort to break them down.

The complexity of texture perception and interpretation of EMG data was discussed by Le Reverend et al. (2016). They questioned the validity of

approaches to EMG data that only consider the first bite and suggested alternatively to consider the whole chewing process. They also discussed that continuous reduction of Chew Work to the point of swallowing happens after the food samples are hydrated and a cohesive-sticky bolus is produced (in their case, cereal products). This is consistent with the values of Chew Work in the present study, where they decreased from the initial to the final phase of chewing. This may also be related to the adaptability of mastication and muscle activity, where continuous oral manipulation of food leads to a higher degree of breakdown and consequently a lower level of muscle activity is required in the final stages of mastication (Woda et al., 2006a, Devezeaux de Lavergne et al., 2016). Another possible explanation could be the dissolution of sugars and the dilution of the model foods, which lead to a softer texture and consequently to lower muscle activity.

It's interesting to think about how age affects chewing behaviour and how stickiness is perceived. According to earlier research, elderly people require more effort from their mouths during chewing than younger people. Kang et al. (2016) reported that older subjects put more effort into mastication, indicating that age may play a role in the chewing process. Additionally, Park et al. (2017) found that relative to older individuals, younger assessors showed greater associations between adhesiveness and general oral processing. Given that the current study's assessors ranged in age from 21 to 27, it is reasonable to hypothesize that older individuals would have shown more pronounced variations in the extracted EMG features associated with mastication. Considering the findings of Kang et al. (2016) and Park et al. (2017), it becomes apparent that age-related factors, such as muscle strength and sensory perception, may affect how sticky foods are perceived and processed. Therefore, as Kang et al. (2016) noted, it may be advantageous to take into account both the hardness level and the stickiness parameter when creating customised foods for older people. Understanding the precise stickiness values that initiate or hasten muscle fatigue may help to improve the composition and texture of foods for this group.

Figure 3-15 demonstrates the mean sensory stickiness compared to Chew Work at different normalised time- epochs. Each colour is allocated to a

model food and individual circles represent each time epoch. The initial stage of mastication starts with circles labelled as number 1 and it increases to higher numbers till the last recorded data point with negative Chew Work values. The figure illustrates that the low sticky model foods (e.g., 10-90 and 35-90) have a lower initial Chew Work at the beginning of the mastication, while the stickier model foods have higher initial Chew Work values. It can also be seen that the stickiness of the model foods decreases slightly from the first epoch to the point of swallowing. Although, the measured stickiness might demonstrate a poor time-dependant behaviour, its changes throughout the mastication are negligible (maximum reduction of 20%). It should be noted that the values of Chew Work diminish considerably (not always significant) for all model foods and their effort to manipulate the bolus decreases continuously, while the assessors perceived the stickiness of model foods similarly throughout the mastication. It might suggest that the consistency of the model foods changed as the mastication proceed to its final stages.

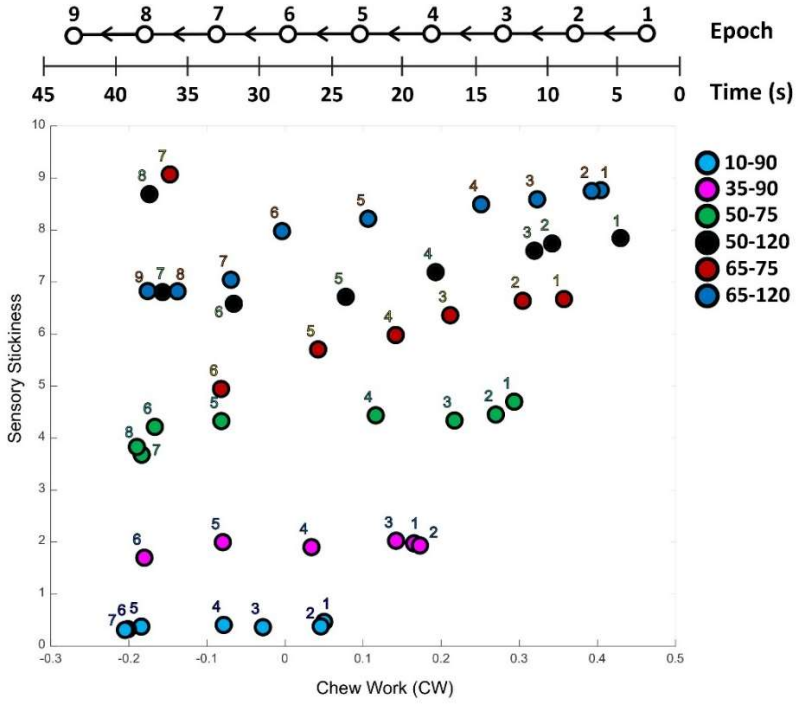

Figure 3-15. (A black-and-white version of the figure is available in Appendix 6.7). Mean sensory stickiness of all the assessors and their normalised chew work at different time epochs. Each model food is assigned with a colour and the numbers above each circle show the epoch or relevant sensory time interval. The first time interval has number 1 and higher values are assigned to subsequent epochs. For example, green circles represent model food 35-90 and its stickiness slightly reduces from the first epoch (right side of the figure) to the last epoch (number 8 on the left side of the figure), while its Chew Work experience a significant reduction. The reason to have a zero on x axis is due to the normalisation of Chew Work.

A similar result was also reported by Iguchi et al. (2015) who found that the activity of the suprahyoid muscle (with a function to open the jaw) decreased from the initial phase to the end of the chewing process, while the adhesiveness of the samples remained constant. In their research, they compared the physical and oral properties of an extremely sticky Japanese rice cake (Mochi) and steamed rice. The adhesiveness was measured as the total-area of the negative area of a two-bite TA test. Both samples exhibited increasing adhesiveness from the early to the later stages of mastication which was more pronounced in the rice cake. This also resulted in a greater EMG activity of the rice cake. Other parameters such as the amount of sample are also responsible for the activity of the chewing muscles, the effect of which can be reduced by the lower amount of sample.

Gao and Zhou (2021) reported that the sticky sensation of breads was a key parameter in categorising similar bread samples. Manipulation of the breads by removing the crust prior to the oral processing led to reduced chewing effort- similar to ChW in the current research (due to the dry texture of crust compared to crumb) and higher values of sticky sensation mainly towards the swallowing point. These findings may indicate that stickier boli might not always result in higher chewing efforts and consequently other textural parameters can dominate over stickiness. What is particularly interesting is that the results of the current study do not agree with the above paper and show that Chew Work is positively associated with stickiness levels. One possible explanation might be due to the differences among the food samples in both studies. The model foods of the current study were homogeneous and semi-solid samples, while Gao and Zhou (2021) used bread samples where the crust makes the texture (compared to the crumb) heterogeneous. Another possible explanation is the difference between the initial texture of model foods and their oral processing path. The model foods in the current study have a totally different oral processing than bread samples. The latter must be chewed until a cohesive-sticky mass is produced, while the former has a cohesive-sticky mass from the beginning. This has been highlighted by Wagoner et al. (2016) who suggested that the cohesive nature of caramel samples means that their oral processing route

differs from the traditional oral route. The traditional oral processing route explains that the food transforms from a fragile texture to a cohesive and sticky texture. They also added that the caramel texture does not go through this route and other parameters such as the dissolution of the sugar are of great importance.

In another study, sensory experiments of hydrocolloid gels with soft and hard textural characteristics were evaluated. The samples had two levels of sweetness by using artificial sweeteners (cyclamate and saccharine). The assessors rated softer and sweeter samples stickier compared to soft-low sweet samples. The same profile was also reported for the hard samples (hardness measured by assessors). In contrary, chewing duration and number of chews (measured by video recording) were higher for lowest sticky model foods (Lasschuijt et al., 2017). It was also reported by Wagoner et al. (2016) that sensory stickiness had a negative relationship with Chew Work. This is in contrary with the finding of the current research. The number of chews and chewing duration for stickier model foods (such as 65-120) were significantly higher than low sticky model foods (e.g., 10-90). Therefore, it can be assumed that for the low sweetness and low stickiness samples, the dominant attributes (either of hardness or softness) had a higher effect on the assessor's perceptions rather than minor textural attributes such as stickiness.

In exploring the relationship between EMG parameters and sensory stickiness, it becomes evident that the correlation between these variables is not consistent across all parameters. While some parameters, such as Chew Work, show a strong correlation with sensory stickiness, others do not. It is crucial to consider other textural attributes like hardness, which can also impact stickiness and should be studied alongside EMG parameters. This could explain why certain parameters fail to associate with sensory stickiness. It has been highlighted that stickier model foods exhibit higher EMG values (e.g., Chew Work and Chew Time). However, caution must be exercised when interpreting correlations, as other factors, such as salivation, can influence the stickiness of model foods during oral processing. It has also been highlighted by Maeda et al. (2020) that the exact relationship

between stickiness and oral processing is unclear but the salivation during mastication of sticky food materials might has an important role. The correlation between EMG and sensory parameters offers valuable insights into the stickiness of model foods. Surprisingly, contrary to the initial hypothesis, the stickiness of model foods does not decrease to a uniform level before swallowing, with a maximum 20% decrease observed during oral processing. This suggests that the oral processing pathway for sticky-cohesive model foods differs from that of foods that become hydrated and sticky prior to swallowing. While some researchers have mentioned this pathway, limited data is available, and further research is necessary to comprehensively understand the oral processing of such samples.

3.3.2 Electromyography in comparison to the rheological measurement

In the current section, the results of EMG data and rheological experiments have been correlated. It should be noted that there is very limited data available in the literature discussing EMG in relation to the rheological characteristics of sticky food materials. Specifically, when it comes to individual EMG parameters, only the most frequently reported features are discussed (such as Chew Work). Therefore, it is sometimes difficult to relate the findings of current research to the literature.

Table 3-29 provides the Spearman correlation coefficients relating EMG features, to storage modulus (G′) at the applied frequency range. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

It can be seen that most EMG parameters (except ChR, $IChT$ and ACh) show significant correlations with storage modulus. A slight increase in the correlation coefficients can be observed by increasing the frequency. Therefore, it can be stated that the correlations of EMG and storage modulus are less affected by the change in frequency. Notably, none of the EMG parameters of ChR, $IChT$ and ACh shows any significant coefficients over the entire applied frequency range.

Table 3-29. Spearman correlation coefficients relating EMG features, to storage modulus (G') at the frequency range. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01 (both two-sided), respectively.

EMG features	Angular frequency (rad/s)								
	10^{-5}	**10^{-4}**	**10^{-3}**	**10^{-2}**	**10^{-1}**	**1**	**10**	**100**	**1000**
ChW	-0.84*	-0.89*	-0.89*	-0.89*	-0.81*	-0.83*	-0.85*	-0.89*	-0.89*
WR	-0.86*	-0.92**	-0.92**	-0.92*	-0.92*	-0.92*	-0.92**	-0.92**	-0.92*
BD	-0.98**	-0.95**	-0.95**	-0.95**	-0.95**	-0.96**	-0.91**	-0.93**	-0.96**
pW	0.92**	0.97**	0.97**	0.97**	0.97**	0.97**	0.97**	0.97**	0.96**
NCh	-0.94**	-0.97**	-0.97**	-0.97**	-0.97**	-0.97**	-0.97**	-0.97**	-0.97**
ChR	0.27	0.21	0.21	0.22	0.23	0.24	0.25	0.25	0.25
IChT	0.001	0.11	0.11	0.10	0.09	0.08	0.07	0.06	0.06
ACh	-0.41	-0.34	-0.34	-0.35	-0.36	-0.37	-0.38	-0.38	-0.38
ChT	-0.92**	-0.94**	-0.94**	-0.94**	-0.94**	-0.95**	-0.95**	-0.95**	-0.95**
MV	-0.87*	-0.92**	-0.92**	-0.92**	-0.92**	-0.92**	-0.92**	-0.92**	-0.92**
AV	-0.84*	-0.91*	-0.91*	-0.91*	-0.91*	-0.91*	-0.91*	-0.91*	-0.91*

Table 3-30 presents the Spearman correlation coefficients relating EMG features to the loss modulus (G") in the given frequency range. It can be seen that there is a significant effect of frequency on the EMG parameters, where most significant correlations are at frequencies from 10^{-5} to 10^{-2} rad/s, while from frequencies above $10^{-}1$, with some exceptions, correlations become poor and insignificant. Similar to the storage modulus.

Similar to the storage modulus, ChR, $IChT$ and ACh are the only parameters with non-significant correlations across the frequency range.

Table 3-30. Spearman correlation coefficients relating EMG features, to loss modulus (G") at the frequency range. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

EMG features	Angular frequency (rad/s)								
	10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}	1	10	100	1000
ChW	-0.91*	-0.91*	-0.89*	-0.82*	-0.67	-0.34	0.02	0.24	0.33
WR	-0.94**	-0.95**	-0.91*	-0.83*	-0.64	-0.29	0.08	0.29	0.36
BD	-0.89*	-0.84*	-0.96**	-0.99**	-0.83*	-0.48	-0.13	0.07	0.15
pW	0.99**	0.99**	0.96**	0.86*	0.58	0.16	-0.20	-0.33	-0.34
NCh	-0.96**	-0.94**	-0.97**	-0.91**	-0.70	-0.31	0.01	0.17	0.22
ChR	0.15	0.11	0.24	0.42	0.74	0.0.67	0.60	0.18	-0.07
IChT	0.21	0.26	0.07	-0.17	-0.54	-0.70	-0.59	-0.25	-0.03
ACh	-0.26	-0.21	-0.37	-0.55	-0.80	-0.73	-0.60	-0.22	0.01
ChT	-0.93**	-0.91*	-0.94**	-0.91*	-0.74	-0.39	-0.07	0.12	0.20
MV	-0.94**	-0.94**	-0.91*	-0.84*	-0.66	-0.31	0.05	0.25	0.32
AV	-0.94**	-0.95**	-0.90*	-0.81	-0.62	-0.26	0.13	0.34	0.40

Table 3-31 shows Spearman's correlation coefficients relating EMG features, to the tan delta in the frequency range. What is interesting about the data is that none of the EMG features show significant correlations at the given frequency range of 10^{-5} to 10^{-1} (rad/s). By increasing the frequency, and specifically at frequencies of 1 and 10 (rad/s), there are significant correlations for $ChW, WR, pW, NCh, MV, ChT$ and AV but a further increase of frequency to more than 10 results in no significant correlations. The frequency dependency of the tan delta as the ratio of loss modulus over storage modulus can be related to the loss modulus response to frequency. G″ showed to be a frequency-dependent parameter for all the model foods (see section 3.2.3). While the low-sticky model foods experienced a constant reduction in loss modulus as frequency was increased, the high-sticky model foods showed an increasing trend, while its loss modulus varied over the frequency range.

The EMG parameters of BD, ChR, $IChT$ and ACh show no significant correlations with tan delta at the whole frequency range. What is interesting about ChR, $IChT$ and ACh is that their correlation coefficients are greatly affected by the frequency. For ChR the maximum correlation is at the frequency of 10^{-2}, while it is the opposite for both $IChT$ and $ACh.$ This is the frequency where tan delta becomes zero as loss and storage modulus are equal.

Table 3-31. Spearman correlation coefficients relating EMG features, to tan delta at the frequency range. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01 (both two-sided), respectively.

EMG features	Angular frequency (rad/s)								
	10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}	1	10	100	1000
ChW	0.15	0.38	0.53	0.40	0.58	0.91*	0.88*	0.80	0.78
WR	0.11	0.42	0.64	0.47	0.64	0.91*	0.83*	0.73	0.70
BD	0.29	0.33	0.49	0.39	0.49	0.63	0.54	0.46	0.44
pW	-0.03	-0.37	-0.74	-0.63	-0.76	-0.89*	-0.70	-0.57	-0.54
NCh	0.12	0.29	0.56	0.50	0.63	0.82*	0.74	0.64	0.62
ChR	-0.64	-0.27	0.35	0.54	0.34	-0.27	-0.67	-0.76	-0.78
IChT	0.70	0.15	-0.60	-0.69	-0.56	-0.10	0.29	0.41	0.44
ACh	0.61	0.22	-0.30	-0.43	-0.25	0.30	0.66	0.73	0.75
ChT	0.15	0.30	0.50	0.42	0.56	0.68	0.79	0.71	0.69
MV	0.12	0.38	0.59	0.45	0.62	0.91*	0.85*	0.75	0.73
AV	0.12	0.44	0.66	0.48	0.67	0.94**	0.84*	0.73	0.70

Table 3-32 provides the Spearman correlation coefficients relating EMG features, to complex viscosity at the frequency domain. As Table 3-32 shows, there are significant correlations for all the EMG parameters (except ChR, $IChT$ and ACh) at the total frequency domain and the significant correlations are minimally affected by the change in frequency.

In contrast, the EMG features of ChR, $IChT$ and ACh do not show significant correlations at given frequencies, as do the other three EMG features of loss modulus, storage modulus, and complex viscosity.

Table 3-32. Spearman correlation coefficients relating EMG features, to complex viscosity at the frequency range. Significant differences are presented by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ (both two-sided), respectively.

EMG features	Angular frequency (rad/s)								
	10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}	1	10	100	1000
ChW	-0.85*	-0.88*	-0.91*	-0.90*	-0.89*	-0.88*	-0.91*	-0.90*	-0.88*
WR	-0.87*	-0.91*	-0.94**	-0.92**	-0.91*	-0.90*	-0.93**	-0.92**	-0.90*
BD	-0.98**	-0.95**	-0.92**	-0.94**	-0.95**	-0.97**	-0.95**	-0.96**	-0.97**
pW	0.93**	0.97**	0.99**	0.98**	0.97**	0.95**	0.98**	0.97**	0.95**
NCh	-0.95**	-0.97**	-0.97**	-0.97**	-0.97**	-0.96**	-0.97**	-0.97**	-0.97**
ChR	0.30	0.20	0.20	0.20	0.20	0.30	0.20	0.30	0.30
IChT	<0.001	0.10	0.20	0.10	0.10	<0.001	0.10	0.10	<0.001
ACh	-0.40	-0.30	-0.30	-0.30	-0.40	-0.40	-0.40	-0.40	-0.40
ChT	-0.92**	-0.94**	-0.94**	-0.94**	-0.94**	-0.94**	-0.95**	-0.95**	-0.94**
MV	-0.88*	-0.92**	-0.94**	-0.93**	-0.92**	-0.91*	-0.93**	-0.92**	-0.91*
AV	-0.85*	-0.91*	-0.93**	-0.92*	-0.91*	-0.89*	-0.92**	-0.91*	-0.89*

EMG parameters of ChR, $IChT$ and ACh are the only features without significant correlations with any of the rheological parameters. They are extracted from chewing frequencies or chewing times which makes them time-dependent parameters. For example, $IChT$ is the time from the previous offset to the next onset (finishing and starting time of each chew), which is equal to the jaw opening time. Therefore, the time-dependency of these parameters as well as the variation among assessors, could be related to their poor correlations. It was also suggested by Kohyama et al. (2003) that time-extracted EMG parameters are strongly influenced by the assessors' variability specifically in the elderly group. It should also be noted that most of the energy-extracted parameters (such as MV and ChW) provide significant correlations ($p < 0.05$).

As discussed in Section 3.2.2, the model foods with low stickiness (10-90 and 35-90) had higher shear stress values compared to the model foods with higher stickiness (50-75, 50-120, 65-75 and 65-120), which are also associated with firmer structures. Similar behaviour was reported for the bread bolus by Gao et al. (2018), who related bolus structure to energy dissipation and muscle activity. The authors suggested that a denser structure leads to higher muscle activity, which was then associated with low energy dissipation. It can be inferred that the stickier model foods in the present study also had a low tendency to dissipate energy (see Figure 3-6 and Figure 3-7) and consequently required a higher effort, muscle activity and number of chews to produce a swallowable bolus. Obviously, reducing the stickiness of the model foods would have an opposite effect on oral processing behaviour.

From the correlation of the rheological parameters at the applied frequency domain with the EMG features, it can be concluded that G′ and the complex viscosity provide significant correlations with most EMG parameters except ChR, $IChT$ and ACh. These correlations are less affected by changes in frequency and can predict stickiness of model foods in relation to EMG parameters. On the other hand, tan delta is strongly frequency dependent and can only provide significant correlations at only a narrow frequency domain. Considering the results of the current research, it can be suggested

that the time-extracted parameters cannot provide strong correlations with rheological parameters.

3.3.3 Electromyography in comparison to the texture analyser measurement

Both EMG and TA methods are instrumental measurements, and it might be challenging to find significant correlations between them. The current section examines which parameters from these two methods show a significant correlation in relation to stickiness. There is still the question of whether these parameters are necessarily related, as TA measures material properties and EMG is a measure of physiology. It will be further investigated how these parameters correlate with each other.

Table 3-33 shows the Spearman correlation coefficients relating EMG features and the texture analyser parameters. Significant differences are indicated by a single asterisk (*) and double asterisk (**) at significant levels of $p < 0.05$ and $p < 0.01$ respectively (both two-sided). It can be seen that both pre-area and the total-area represent the maximum number of significant correlations with 7 EMG variables, while the distance to the adhesive peak and the initial gradient show no significant correlations with any of the EMG features. Matsuyama et al. (2021) correlated textural properties of hydrocolloid-based model samples (with different contents of gellan gum, xanthan gum and/ or locust bean gum) with EMG variables. The adhesiveness was measured by a TPA test by considering the negative area of a force-distance curve. The results showed no significant correlation between the measured adhesiveness with swallowing ease and the uniformity of bolus.

When correlating TA and EMG results, it should be considered that with TA the measurements were made on the intact physical properties of the samples, whereas with EMG the recorded data come from muscle activity during oral processing. During the oral processing the dominant texture perception changes and subsequently reduces the applicability of such correlations (Matsuyama et al., 2021) which might explain why most of the correlations are neither excellent nor -existent.

Table 3-33. Spearman correlation coefficients relating EMG features, to texture analyser parameters. significant differences are presented by single asterisk (*) and double asterisk (**) at significant levels of p < 0.05 and p < 0.01 (both two-sided), respectively.

EMG Features	Initial gradient	Force of the adhesive peak	Distance to adhesive peak	Pre-area	Total-area
ChW	-0.74	-0.88*	-0.46	-0.95**	-0.92**
WR	-0.65	-0.83*	-0.56	-0.95**	-0.90*
BD	-0.41	-0.57	-0.62	-0.69	-0.62
pW	0.49	0.70	0.72	0.92**	0.87*
NCh	-0.60	-0.75	-0.61	-0.89*	-0.83*
ChR	0.80	0.68	-0.48	0.29	0.34
IChT	-0.49	-0.28	0.69	0.13	0.02
ACh	-0.78	-0.67	0.33	-0.34	-0.37
ChT	-0.67	-0.81	-0.54	-0.89*	-0.83*
MV	-0.69	-0.85*	-0.53	-0.95**	-0.90*
AV	-0.65	-0.83*	-0.55	-0.96**	-0.92**

The EMG features of $BD, ChR, IChT$ and ACh do not show significant correlations with any of the TA parameters. These parameters also failed to provide significant differences among model foods, reflecting their low ability to discriminate model foods (see section 3.3.1). All these parameters except ChR are time extracted values. This means that they can be affected by the slow or fast chewing behaviour of assessors. One possible solution to reduce the assessors' impact was to follow a standardised chewing protocol. Although, this method could have increased the significant correlations of above parameters, this would be a deviation from the aim of the current study to investigate natural chewing behaviour. Therefore, it can be suggested that time extracted EMG features cannot be used to study the relationship between EMG and TA parameters.

Among the EMG features, ChW, WR, MV and AV stand out as having the highest significant correlations with the TA parameters. These are in line with the results presented in section 3.3.1 that these parameters distinguished the low sticky model foods from all other samples. The three parameters ChW, MV and AV are the energy extracted values (voltage unit) compared to time extracted features. Therefore, they are more related to the physical characteristics of model foods than being affected by the assessors. Two parameters of ChW and WR have been obtained from the efforts made during the chewing. This might explain the strong and significant relationship of these parameters with pre-area and total- area. Notably, pre-area and total-area are also the parameters related to work of adhesion compared to a single value such as distance to adhesive peak. Regarding the MV and AV as the maximum and average voltage indications, respectively, significant relationships have been found for other textural attributes such as hardness (Yoshida et al., 2009, Taniguchi et al., 2013, Laird, 2017), while the available data on the stickiness are very limited.

Total-area and pre-area have the same number of significant correlation coefficients for exactly the same EMG features, but it is evident that pre-area presents stronger correlation coefficients at all significant levels. As the pre- area was introduced in the present study and can be easily extracted from the results of TA, it may be suggested that this parameter be applied to different samples when performing compression tests to characterise its applicability to other foods. These results are in contrary with the findings of Kohyama and Hayakawa (2007) and Kohyama et al. (2008) who reported that the stickiness (as the total area of a TA experiment)

obtained at low strains (up to 50% strain) did not significantly correlated with any of the EMG variables. Although the TA experiments in the current study are not based on strain levels, the conversion of the force and distance compression values used in the TA experiments correspond to very low strain levels. A possible explanation for this is the differences in the nature of the samples between the above studies and the current study, where the former used solid foods (e.g., carrots and raw radish) and the latter used semi-solid model foods. Most of their samples did not show stickiness as determined by a TPA test. However, the model foods in the current study had a wide range of stickiness, ranging from very low to very high values. Although, Kohyama et al. (2008) reported that both raw and cooked carrots were not sticky in the compression test, in another study, stickiness increased compared to raw carrots when carrots were cooked and further pureed (Wee et al., 2018). Comparing the above samples with our model foods in the current study, it can be seen that the pureed carrot has more textural similarities with the model foods than the raw or even cooked carrot. The increased stickiness can be highlighted as one of these similarities. Another explanation could be the differences in composition between the model foods and the samples from the study by Kohyama et al. (2008).

In a different study by Park et al. (2020) adhesiveness was suggested to be the main textural aspect of semisolid food for aged adults (mean 72.5±6.9 years) by requiring the maximum effort for pharyngeal swallowing mainly for boiled mashed pumpkin, potato, and sweet potato. In the same age group, adhesiveness also correlated with swallowing difficulties and the amount of oral residue. As discussed in section 3.2.1, it is important to note that the possible presence of low molecular weight sugars in the model foods, mainly as a result of longer heating times for model foods with higher sugar content, maximised stickiness.

It is important to note that several studies have used expectorated boli to measure food stickiness, mainly focusing on comparing instrumentally measured stickiness with EMG characteristics (Iguchi et al., 2015). As most of these studies are beyond the scope of this section, they are not discussed here. In a related study, Devezeaux de Lavergne et al. (2015a) reported that the measured adhesiveness (total-area from TA analysis) of the boli of individuals with short and long oral processing time was different. The short chewer manipulated the food sample less and consequently the parameters involved in the development of adhesiveness during chewing, such as

saliva secretion, had a much smaller effect on the bolus than the long chewer, who produced stickier boli. The authors suggested that oral processing time could lead to different swallowing triggers, which is in line with Puerta et al. (2020) who suggested that the extent to which food is broken down during chewing is as important as its mechanical properties to extensively understand bolus development and texture perception.

It should be considered that careful selection of instrumental parameters is critical when performing correlations with EMG parameters. Since the latter is obtained from oral processing with a dynamic character, the former is obtained from a non-destructive texture test. It can therefore be suggested that both destructive and non-destructive TA experiments should be conducted in future studies to investigate whether manipulation of food texture with possible addition of saliva would improve the relationship with EMG parameters.

In summary, both pre-area and total-area suggested to be the most reliable TA parameters in relation to EMG features. They showed the most significant correlations with the energy extracted EMG features which are more influenced by the physical condition of the model foods and less by the chewing behaviour of the assessors. This is an important finding as both pre-area and total-area are easily extracted parameters. The present results confirm that the pre-area could provide stronger correlation compared to the total-area which is the successful outcome of the current research.

3.4 Principal Component Analysis of the data

In the current study, two instrumental methods texture analyser and rheology, sensory evaluation and EMG were used to measure the stickiness of model foods. Although the use of multiple methods contributes to a comprehensive understanding of stickiness as a complex textural feature, the interpretation of the data can be challenging. Principal Component Analysis (PCA) is a method for visualising data to improve the interpretability of multidimensional data. In this section, PCA was employed to elucidate the interrelationships among the data variables and provide a more comprehensive understanding.

PCA was performed for all measured parameters (TA, rheology, sensory and EMG - 53 parameters in total) with Direct Oblimin rotation (Figure 3-16). Using the scree plot, 2 principal components were selected to represent the PCA results (see appendix 6.7 for detailed data). Figure 3-16 shows that the dimensions explain 85.3% of the variance, with Principal Component 1 (PC1) accounting for 68.9% and PC2 16.4%. The component matrix shows that most parameters are explained by PC1 (see appendix 6.6 for component matrix from PCA). The total variance explanation table (6.7) shows that 7.5% of the variance is explained by PC3. By having a closer look at the component matrix (Appendix 6.6), PC3 cannot explain any parameter more comprehensively than PC1 or PC2. However, PC3 gives higher values for some parameters (e.g., Complex viscosity F10-4, total-area, and pre-area) compared to PC2, where PC1 has the highest values. Therefore, while PC1 remains the primary component for explaining these parameters, it is important to consider PC3 as it may contribute to a more comprehensive understanding of the underlying factors influencing these parameters.

Figure 3-16 shows that most of the parameters are clustered into two groups (inside dashed circles). The parameters in the same dashed circle have significant correlations ($p < 0.05$). These two circles are on the extreme positive and negative sides of PC1 which, means they have strong negative correlations with each other. For other parameters with lower values of PC1, such as the initial gradient, the correlations are weaker than force of the adhesive peak with all the parameters in

the circles. The same applies to the parameters that are more effectively described by PC2.

On the right-hand side of the plot, the three rheological parameters loss modulus, storage modulus and complex viscosity (with some frequency exceptions) are grouped with the TA parameters total-area and pre-area. It is particularly interesting to note that pre-area has a stronger correlation with all these parameters, indicated by a lower distance on the PC1 component (x-axis) with the main cluster of parameters. On the other hand, the cluster of parameters on the left side of the plot consists mainly of EMG features with overall stickiness. Notably, each cluster consists of parameters with the same origin. The parameters from instrumental measurements are clustered in the right circle, while the parameters related to oral processing (sensory evaluation and EMG) are mainly clustered on the negative side of PC1.

There are some other interesting aspects to the PCA plot. The three EMG parameters ACh, $IChT$ and ChR are on either extreme side of PC2 (with low values of PC1) which, means that these parameters are more effectively captured by PC2 than by PC1. The reason for this might be the nature of these parameters as they are time-extracted parameters compared to energy-extracted parameters.

With regard to the scatteredness of different frequencies of G˝ and tanδ, it should be noted that the Pearson correlations of these parameters are highly frequency-dependent (see related sections on correlations). Since tanδ is highly related to loss modulus, its frequency dependency would depend hugely on that. The frequency dependency of the loss modulus can be related to the structure of the model foods and its degree of stickiness. The stickier the model foods, the more their loss modulus also depends on frequency. Furthermore, the scattered frequencies provided mainly weak correlations for both G" and tanδ, indicating their poor ability to predict the stickiness of model foods.

Examination of the Pearson correlations performed in the previous sections with the parameters highly described by PC2 revealed that the parameters of the initial gradient, G˝1, G˝10 and tanδ 10^{-5} are not strongly correlated with any of the parameters. This is also true for ACh, $IChT$ and ChR, which are only weakly correlated.

Two parameters of TA, namely the initial gradient and the distance to the adhesive peak, are also at higher levels of PC2 compared to PC1. Although the Pearson correlations of these parameters with all other parameters showed some strong relationships, they mainly represented weak and non-significant correlations with other measurements, making them parameters with low ability to predict parameters related to stickiness.

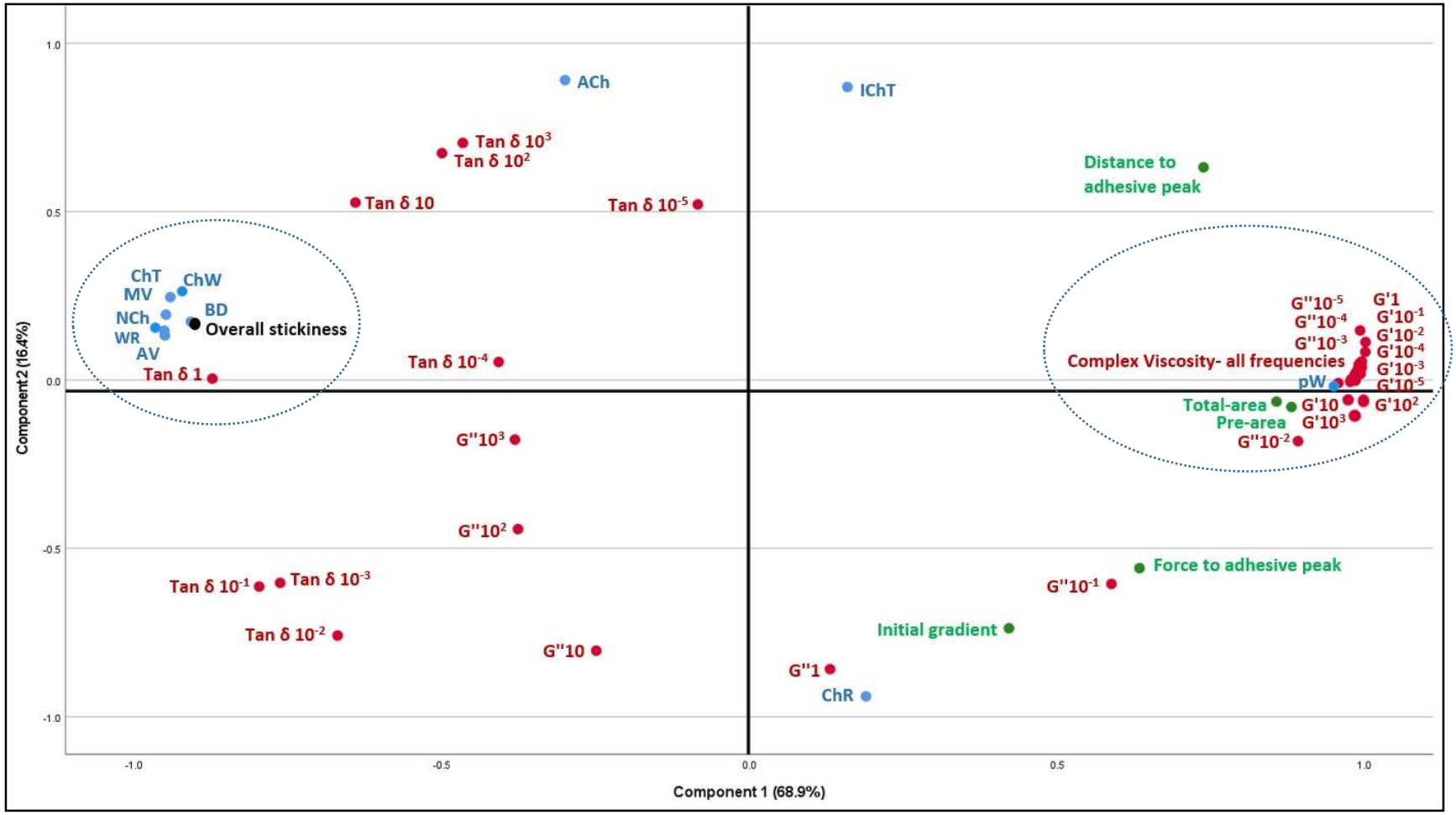

Figure 3-16. (A black-and-white version of the figure is available in Appendix 6.9). Principal Component Analysis (PCA) of all measured parameters (TA, rheology, sensory and EMG). Principal component 1 (PC1) represents 68.9% and PC2 16.4% of the variance in the data. the parameters of each method have the same colour. TA parameters are in green, rheology parameters are in red, overall stickiness is in yellow and EMG parameters are in blue. The frequencies of rheology experiments are labelled with numbers, e.g., G´10^{-2} represents the loss modulus at the frequency of 10^{-2}. Dashed circles indicate two clusters of parameters with the highest relationships within each circle. The parameters on the right side of the plot are negatively correlated with the parameters on the left side.

It can be interpreted that PC1 is the axis of most of the relationships between the instrumental measurements of TA and rheology with sensory evaluation and EMG features. Therefore, PC1 can be used to explain most aspects of stickiness in relation to different measurements. On the other hand, PC2 cannot be directly interpreted with a clear relationship to stickiness and its function is quite difficult to explain.

It should be noted that unlike other texture characteristics (such as hardness, which is the initial property of the food sample), stickiness is a texture property that can result from the chewing process (e.g., bread bolus) (Jourdren et al., 2016). Or, in other words, it arises from food manipulation (mainly for less hydrated foods such as biscuits). It has been suggested that the perception of stickiness occurs after some manipulation of the food (e.g., 60% of chewing) by producing a bolus (Pascua et al., 2013, Young et al., 2013). A similar study by Maeda et al. (2020) measured the adhesiveness and cohesiveness of some steamed rice samples. Bolus adhesiveness was measured as the negative area of the first negative section of a TPA test at 0%, 50%, 100%, 150% oral processing time points. The results showed a significant decrease in adhesion force from 0 to 50%, while it remained almost constant thereafter until 150%. While cohesiveness showed a different behaviour, increasing from 0 to 150%, the authors reported a significant correlation between adhesiveness and cohesiveness. It was recommended that the changes in adhesiveness were time-independent, while both adhesiveness and cohesiveness were affected by increasing the number of chews. This time-independent behaviour of adhesiveness is particularly interesting as a similar pattern was observed in the model foods of the current study. Although the stickiness of the model foods decreased somewhat, the perceived stickiness remained constant until swallowing. In addition, Maeda et al. (2020) suggested that bolus stickiness may be influenced by salivary secretion, which in turn is related to the lubricant content required before swallowing (Wee et al., 2018). Moreover, Schmidt et al. (2018) stated that stickiness cannot be explained by adhesiveness alone as a surface property, but that cohesion is also required for a comprehensive understanding of stickiness. This is an important point when talking about

stickiness as a complex textural feature that develops during chewing and is also influenced by different oral processing parameters.

It is of great importance to remember that applying correlations and combining the results of different methods are far more effective in finding possible explanations for oral perceptions in relation to instrumental measurements (Steffe, 1996). And the results would be more practical for understanding food texture and then using this knowledge to develop specific foods (Kohyama et al., 2015, Le Reverend et al., 2016). The application of this approach has improved the prediction of food creaminess and fattiness (Terpstra, 2008). Similarly, Aguayo-Mendoza et al. (2019) expressed that the consumption time for semi-solid products can be obtained from the rheological and mechanical aspects of food materials. The application of PCA to the data of the current study showed strong correlations between several parameters. These results can be useful for stronger prediction of stickiness through instrumental measurements, as they are a more convenient methods compared to sensory or EMG. Additionally, they contribute to the improved design of food textural properties concerning customer acceptance and specific needs. Moreover, these parameters have implications for other physical attributes, such as mitigating the surface stickiness of food materials in packaging applications.

4 Conclusion

In this study physiological, sensory and instrumental measures of stickiness were established and cross correlated.

The three instrumental approaches to measure stickiness,

- Bulk modulus proved technically challenging and was not possible with the equipment or resources available.
- Texture analyser provided significant correlations with sensory stickiness. In addition to previously described instrumental measures of stickiness, two new quantities were defined: the pre-area and the initial gradient. The pre-area proved to be a parameter that provides significant correlations with rheological and EMG parameters.
- Rheometry stress relaxation experiment was conducted at strain values below 0.1%. The viscoelastic response of the model foods was a function of their water and sugar content. As the stickiness increased, the shear stresses decreased, and the stickier model foods relaxed faster than the less sticky samples. It was suggested that this behaviour was related to the energy dissipation of the model foods leading to greater manipulation of the stickier model foods.

Participants involved in the sensory study, simultaneously undertook electromyography and 22 characteristics were extracted from the muscle activity. The sensory evaluation showed that assessors were able to significantly differentiate the stickiness of the model foods ($p < 0.05$). By increasing the sugar content and thus the stickiness of the model foods, the assessors needed more time to cope with oral processing. The extracted EMG data showed the highest number of correlations for Chew Work and Chew Time.

The combination of instrumental, sensory and physiological methods using PCA proved to be a valuable approach for mapping the stickiness as a complex textural attribute. For example, pre-area and total-area show

significant positive correlations with most rheological parameters and significant negative correlations with most EMG features and overall stickiness ($p < 0.05$).

Despite the fact that the sensory stickiness of the model foods did not show significant changes during oral processing, all of the samples were still successfully swallowed. These results suggest that although the consistency of many foods tends to become stickier as they are swallowed, stickiness may not be the main trigger for swallowing in the model foods used in this study. This implies that other textural attributes or oral processing parameters might have a more pronounced role in triggering swallowing.

The findings of this study have the potential to enhance the prediction of stickiness through instrumental measurements, offering a more convenient alternative to sensory or EMG methods. Moreover, these results can inform the optimization of food textural properties to align with customer acceptance, particularly for products that benefit from a desired moderate level of stickiness.

5 Future work or suggestions

This chapter identifies the limitations of the current study and makes some suggestions for future research based on the findings of the present study.

- In the current study, the parameter of pre-area from texture analysis was introduced. It was explained in section 3.2.1 that the reason for developing this parameter was to consider only the surface stickiness of the model foods and to exclude rheological properties. Since in the present study all measured parameters were a combination of different texture attributes/properties, it can be suggested to correlate the pre-area with other surface measurements of stickiness (e.g., tactile perception of stickiness with the finger). In this way, there is the possibility of obtaining a stronger correlation of the data from TA and especially the pre-area with other measurements.
- Further research should be carried out to investigate the initial gradient parameter. This parameter should be investigated using different food textures to verify its potential applications. Another area can be the development of a method that finds the linear part of the curve and then automatically selects the points instead of determining them manually, as was the case in the current research.
- It is crucial to take saliva into account for future instrumental studies of stickiness. Although saliva has only been used in a small number of research on stickiness, it should be highlighted that saliva serves as a bridge between the physical characteristics of food and the sensory experience of eating. It is feasible to establish a relationship between instrumental measures and sensory assessments by looking at the changes in saliva that occur during the eating process.
- The material of the probe surface for conducting TA experiments may be an important parameter to consider for future studies. Stickiness may be perceived differently when the material of the probe is changed.

- The phenomenon of glass transition temperature (Tg) is another parameter that is becoming increasingly popular among researchers to study stickiness. Tg can be particularly useful for starch-based model foods, as water movement through interaction with water depends on Tg. As discussed in section 3.2.1, the interaction of starch and water significantly affects the viscoelastic behaviour of model foods and thus the sensory and instrumental perceived stickiness. Therefore, it would be useful to include Tg in future studies measuring stickiness.
- Although bulk modulus is traditionally used for agricultural products, it can also be used to measure the internal strength of food. For bulk modulus measurement, it is essential to have a suitable vessel that can withstand up to 30 MPa to meet the different compression requirements. Since no shear stress occurs in the bulk modulus, higher compression rates are required compared to the human jaw (about 869 kPa). Lack of technological development was mentioned as one of the possible reasons why bulk compression is not widely used in food science. It would be very helpful to develop standard equipment for measuring bulk modulus and then studying the texture of food.
- As the model foods in the present study were complex systems, interpretation of the results was sometimes difficult. Therefore, it may be suggested to use a design of experiments to investigate the effects of each composition factor and cooking time on each measured parameter. In this way, the addition and effect of each specific ingredient can be discussed in more detail.
- Conducting the rheological tests outside the LVE region could contribute to a stronger correlation with other methods, especially the sensory evaluation of the model foods. The main reason for this suggestion is the structural break down of foods during chewing, which is on the opposite side of the LVE as the limit of unrecoverable structural damage. One of the issues to consider when conducting tests outside the LVE region is the complexity of data analysis and the influence of other textural properties on the measurement of stickiness.

6 Appendices

6.1 Taste blocking - *Gymnema sylvestre*

Gymnema sylvestre widely known as “Gurmar” is a plant belonging to the *Asclepiadaceae* family which originates in parts of India (Shanmugasundaram et al., 1990, Potawale et al., 2008), east Asian countries, Australia and tropical Africa (Saneja et al., 2009). By chewing the leaves of this plant, the human oral sweet perception can be suppressed for a short period of time (Warren and Pfaffmann, 1959, Kurihara, 1992, Manohar et al., 2009).

According to Glaser et al. (1984), it was in 1847 that this effect of *Gymnema sylvestre* leaves was first introduced to a scientific association. It has subsequently been suggested that the gymnemic acid found in the leaves is the active component responsible for the sweetness blocking effect (Kurihara and Nirasawa, 1994). Gymnemic acid blocks the sweetness receptors on the palate and tongue (Devi and Jain, 2015) and the leaves of *Gymnema sylvestre* have been shown to be able to blocks the sweet taste of a range of natural and artificial sweeteners (Hayes, 2008).

According to Yarmolinsky et al. (2009), sweet taste is one of the five basic tastes along with bitter, sour, salty and umami. Sweetness is perceived through G-protein-coupled receptors (Lindemann, 1996). Two types of these proteins are active in taste perception (Lawless and Heymann, 2010). Type 1 taste receptors (T1R) are responsible for sweet and umami taste perception (Kochem, 2017) whereas type 2 receptors perceive bitter taste (Nelson et al., 2001). The other two tastes (sour and salty) are detected by ion channels (Lindemann, 1996). Figure 6-1 illustrates T1R perceiving sweetness.

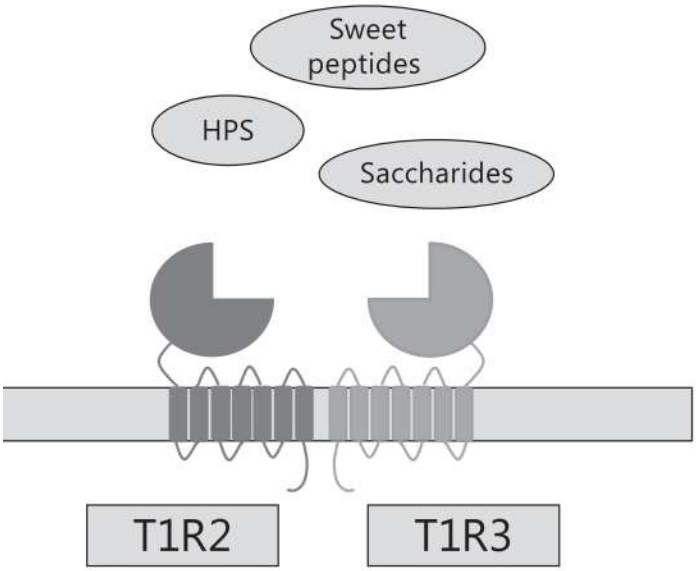

Figure 6-1. Type 1 receptors subunits 2 and 3 bind to sweet peptides, HPS (high potency sweeteners) and saccharides (Kochem, 2017).

Sweet taste is said to have a “bright” effect on consumers and provides them with joy while consuming a sweet food product (Ganchrow et al., 1983, Frank et al., 1992, Lindemann, 1996). This is why a sweeter product might be more attractive to assessors than one that has similar texture but less sweet. Subsequently, sweetness can have an interference in their judgement about a food product when this factor is not the attribute of interest. This is known as a “dumping effect”, where a strong attribute in the food product that is not intended to be reported as the purpose of the study, has an effect on the assessors’ perceptions and rankings (Lawless and Heymann, 2010).

Gymnema sylvestre has a wide range of applications. It has been used as a traditional sweetness blocking and antidiabetic agent (Shanmugasundaram et al., 1990, Devi and Jain, 2015). It has also been used as an anti-obesity agent in form of tablets to be dispersed in the mouth or a food additive to reduce the palatability of sweet products and subsequently reduces the craving for sweetness (Porchezhian and Dobriyal, 2003, Devi and Jain, 2015). Other areas to use *Gymnema sylvestre* include, but not limited to, Hyperlipidaemia effect (Shigematsu et al., 2001), antiallergic property (Sawabe et al., 1992), dental caries treatment (Parimala et al., 2009),

antibiotic activity (Deb Roy et al., 2010) and wound healing effect (Malik et al., 2009).

6.2 Ethical approval for sensory evaluation study

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Faculty of Medicine & Health Sciences
Research Ethics Committee
c/o Faculty PVC Office
School of Medicine Education Centre
B Floor, Medical School
Queen's Medical Centre Campus
Nottingham University Hospitals
Nottingham, NG7 2UH

11 March 2019

Seyedmostafa Kazemeini
PhD Student
c/o Dr Andrew J Rosenthal
Associate Professor of Food Sciences
Division of Food Sciences
School of Biosciences
Sutton Bonington Campus
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LE12 5RD

Dear Mr Kazemeini

Ethics Reference No: 270-1803 – **please always quote**	
Study Title: Effect of stickiness of solid food products on trigger of food swallowing.	
Chief Investigator/Supervisor: Dr Andrew J Rosenthal, Associate Professor, Division of Food Sciences, School of Biosciences.	
Lead Investigators/student: Seyedmostafa Kazemeini, PhD Student, Food Sciences	
Type of Study: PhD project	
Proposed Start Date: 30/10/2018	**Proposed End Date:** 31/05/2018
No of Subjects: 15	**Age:** 18+years

Thank you for notifying the Committee of amendment no 2: 07.02.2019as detailed and the following documents were received:

- FMHS REC Notice of Amendment form and supporting documents version 1.0: 07.02.2019

These have been reviewed and are satisfactory and the study has been given a favourable opinion.

A favourable opinion has been given on the understanding that:

1. The protocol agreed is followed and the Committee is informed of any changes using a notice of amendment form (please request a form).
2. The Chair is informed of any serious or unexpected event.
3. An End of Project Progress Report is completed and returned when the study has finished (Please request a form).

Yours sincerely

Professor Ravi Mahajan
Chair, Faculty of Medicine & Health Sciences Research Ethics Committee

6.3 Results of the preliminary sensory testing

The following table illustrates the results of the preliminary sensory tests using a general linear model (ANOVA) and a Tukey HSD post-hoc comparison. The assigned letters in each column show the significant differences between the model foods where different letters are significantly different ($p < 0.05$).

Model Foods	Assessor1	Assessor2	Assessor3	Assessor4	Assessor5	Assessor6	Average stickiness rating
10-90	0.8^{a}	0.4^{a}	0.2^{a}	0.8^{a}	0.6^{a}	0.4^{a}	0.5^{a}
35-90	2.6^{b}	2.0^{b}	2.6^{b}	2.4^{b}	2.4^{b}	2.5^{b}	2.4^{b}
50-75	4.3^{c}	4.3^{c}	5.1^{c}	5.0^{c}	5.2^{c}	4.7^{c}	4.7^{c}
65-75	6.8^{d}	7.2^{d}	7.1^{d}	6.6^{d}	6.6^{c}	6.9^{d}	6.8^{d}
50-120	8.4^{e}	$8.3^{d,e}$	8.5^{e}	8.4^{e}	8.3^{d}	7.6^{d}	8.2^{e}
65-120	9.7^{f}	$9.2^{e,f}$	9.8^{f}	9.6^{e}	9.4^{d}	9.6^{e}	9.5^{f}

6.4 Profile data for model food*assessor interactions

Tukey HSD post-hoc comparison of model food*assessors based at each time interval ($p < 0.05$).

Model foods	Assessors	Time intervals (s)				
		5	10	15	20	25
10-90	a1	0.3^{a,b}	0.5^{a}	0.8^{c}	0.8^{c}	0.5^{c}
	a2	0.5^{a,b}	0.7^{a}	0.7^{c}	0.8^{c}	0.6^{c}
	a3	0.8^{b}	0.3^{a}			
	a4	0.1^{a}	0.0^{a}	0.1^{a}	0.1^{a}	
	a5	0.8^{a,b}	0.4^{a}	0.2^{a}		
	a6	0.5^{a,b}	0.4^{a}	0.2^{a}		
	a7	0.4^{a,b}	0.1^{a}	0.1^{a}		
	a8	0.2^{a,b}	0.2^{a}	0.3^{a}	0.3^{a}	0.2^{a}
	a9	0.1^{a,b}	0.2^{a}	0.2^{a}	0	
	a10	0.6^{a,b}	0.7^{a}	0.4^{b}	0.4^{b}	0.4^{b}
35-90	a1	2.1^{a}	1.9^{a}	2.5^{a}	2.0^{a}	2.3^{c,d}
	a2	2.5^{a}	2.5^{a}	3.0^{a}	2.9^{a}	3.6^{d}
	a3	1.6^{a}	1.5^{a}	0.8^{a}	3.1^{a}	0.01^{a}
	a4	2.8^{a}	2.6^{a}	2.8^{a}	0.9^{a}	3.4^{d}
	a5	2.6^{a}	2.4^{a}	1.9^{a}	2.7^{a}	0.6^{a,b,c}
	a6	1.6^{a}	1.5^{a}	1.5^{a}	1.0^{a}	0.9^{a,b,c}
	a7	1.2^{a}	1.3^{a}	1.4^{a}	1.8^{a}	1.0^{a,b,c}
	a8	1.0^{a}	1.4^{a}	1.5^{a}	1.3^{a}	1.2^{a,b,c}
	a9	2.6^{a}	1.8^{a}	1.3^{a}	1.5^{a}	0.2^{a,b}
	a10	3.4^{a}	2.9^{a}	2.8^{a}	0.9^{a}	2.2b^{,c,d}
50-75	a1	4.6^{a}	4.8^{a}	5.2^{a,b}	4.8^{a,b}	4.4^{a,b,c}
	a2	3.4^{a}	3.7^{a}	3.8^{a,b}	4.4^{a,b}	5.1^{b,c}
	a3	4.3^{a}	3.5^{a}	2.8^{a}	3.0^{a}	1.9^{a,b}
	a4	7.0^{a}	6.9^{a}	7.1^{b}	7.2^{b}	7.2^{c}
	a5	4.7^{a}	4.1^{a}	2.6^{a}	1.9^{a}	1.1^{a}
	a6	5.9^{a}	4.3^{a}	4.1^{a,b}	3.8^{a,b}	3.0^{a,b}
	a7	3.3^{a}	3.4^{a}	3.4^{a,b}	3.5^{a,b}	2.8^{a,b}
	a8	3.5^{a}	3.6^{a}	3.6^{a,b}	3.6^{a,b}	3.5^{a,b}
	a9	4.7^{a}	4.2^{a}	3.7^{a,b}	3.2^{a}	3.2^{a,b}
	a10	5.5^{a}	5.4^{a}	4.9^{a,b}	4.8^{a,b}	3.2^{a,b}
50-120	a1	7.4^{a}	7.5^{a}	7.8^{a,b}	8.2^{b}	8.0^{a,b}
	a2	6.9^{a}	6.7^{a}	7.0^{a,b}	7.1^{a,b}	6.5^{a,b}
	a3	7.7^{a}	8.1^{a}	7.9^{a,b}	7.5^{a,b}	7.4^{a,b}
	a4	9.5^{a}	9.2^{a}	9.3^{b}	8.7^{b}	8.6^{b}
	a5	7.3^{a}	6.0^{a}	5.3^{a}	3.4^{a}	2.4^{a}
	a6	9.2^{a}	8.4^{a}	8.4^{a,b}	7.2^{a,b}	7.1^{a,b}
	a7	7.4^{a}	7.9^{a}	7.8^{a,b}	7.8^{b}	5.4^{a,b}

	a8	7.5[a]	8.3[a]	8.3[a,b]	8.4[b]	8.0[a,b]
	a9	9.0[a]	9.0[a]	8.3[a,b]	8.3[b]	8.3[b]
	a10	6.4[a]	6.3[a]	6.0[a,b]	5.5[a,b]	5.3[a,b]
65-75	a1	5.6[a]	5.2[a]	4.5[a]	5.0[a]	4.3[a,b,c]
	a2	4.5[a]	4.7[a]	4.7[a]	4.8[a]	4.8[a,b,c]
	a3	7.4[a]	8.1[a]	7.7[a]	7.1[a]	6.9[b,c]
	a4	8.5[a]	8.3[a]	8.4[a]	8.4[a]	8.5[c]
	a5	7.2[a]	6.6[a]	6.1[a]	4.4[a]	2.2[a,b]
	a6	8.2[a]	7.9[a]	7.5[a]	6.6[a]	7.0[b,c]
	a7	4.7[a]	5.1[a]	5.1[a]	4.7[a]	1.3[a]
	a8	7.1[a]	7.3[a]	7.3[a]	7.0[a]	6.9[b,c]
	a9	6.9[a]	6.6[a]	5.8[a]	5.4[a]	5.4[a,b,c]
	a10	5.2[a]	5.0[a]	4.9[a]	4.3[a]	4.8[a,b,c]
65-120	a1	8.9[a]	8.9[a,b]	8.2[a]	6.8[a]	8.6[b,c]
	a2	7.7[a]	7.8[a,b]	8.0[a]	8.1[a,b,c]	8.0[a,b,c]
	a3	7.7[a]	8.2[a,b]	8.2[a]	8.3[a,b,c]	9.4[c]
	a4	9.9[a]	9.9[b]	9.9[a]	9.2[b,c]	9.8[c]
	a5	9.6[a]	9.2[a,b]	8.5[a]	9.8[c]	6.1[a]
	a6	9.0[a]	8.7[a,b]	8.6[a]	7.5[a,b]	7.4[a,b,c]
	a7	8.3[a]	8.8[a,b]	8.5[a]	8.4[a,b,c]	8.7[b,c]
	a8	9.5[a]	9.3[a,b]	9.3[a]	8.7[a,b,c]	9.2[c]
	a9	9.9[a]	9.7[b]	9.6[a]	9.3[b,c]	8.9[b,c]
	a10	7.3[a]	7.0[a]	7.0[a]	9.1[b,c]	6.7[a,b]

6.5 Grouping assessors based on their total chewing time

Tukey HSD post-hoc comparison of assessors based on total chewing time ($p < 0.05$).

Assessors	Model foods											
	10-90		35-90		50-75		50-120		65-75		65-120	
	Total chewing time (s)	Overall stickiness	Total chewing time (s)	Overall stickiness	Total chewing time (s)	Overall stickiness	Total chewing time (s)	Overall stickiness	Total chewing time (s)	Overall stickiness	Total chewing time (s)	Overall stickiness
a1	$15.0^{a,b}$	$0.3^{a,b}$	$23.7^{a,b}$	3.1^{a}	25.0^{a}	5.2^{a}	21.2^{a}	8.1^{a}	16.2^{a}	5.0^{a}	$23.7^{a,b}$	9.3^{c}
a2	$18.7^{a,b,c}$	$0.5^{a,b}$	25.0^{b}	3.3^{a}	21.2^{a}	4.0^{a}	$25.0^{a,b}$	7.2^{a}	$26.2^{a,b}$	4.5^{a}	$23.7^{a,b}$	$8.7^{a,b}$
a3	8.7^{a}	$0.6^{a,b}$	15.0^{a}	1.3^{a}	20.0^{a}	3.9^{a}	$23.7^{a,b}$	8.1^{a}	$23.7^{a,b}$	7.8^{a}	$30.0^{a,b,c}$	$8.4^{a,b}$
a4	$15.0^{a,b}$	$0.05^{a,b}$	$23.7^{a,b}$	3.1^{a}	27.5^{a}	7.5^{a}	33.7^{b}	9.2^{a}	30.0^{b}	8.7^{a}	$37.5^{b,c}$	9.9^{c}
a5	$16.2^{a,b,c}$	$0.4^{a,b}$	27.5^{b}	2.3^{a}	27.5^{a}	5.1^{a}	$30.0^{a,b}$	6.9^{a}	27.5^{b}	6.6^{a}	38.7^{c}	9.7^{c}
a6	$13.7^{a,b}$	$0.1^{a,b}$	$18.7^{a,b}$	2.0^{a}	23.7^{a}	4.8^{a}	$27.5^{a,b}$	8.9^{a}	$25.0^{a,b}$	7.8^{a}	$27.5^{a,b,c}$	9.3^{c}
a7	$10.0^{a,b}$	$0.1^{a,b}$	$21.2^{a,b}$	1.5^{a}	21.2^{a}	3.8^{a}	$22.5^{a,b}$	7.9^{a}	$21.2^{a,b}$	5.3^{a}	22.5^{a}	$8.8^{a,b}$
a8	$21.2^{b,c}$	$0.2^{a,b}$	$23.7^{a,b}$	1.7^{a}	28.7^{a}	4.1^{a}	$27.5^{a,b}$	8.8^{a}	$26.2^{a,b}$	7.5^{a}	$26.2^{a,b,c}$	9.6^{c}
a9	$13.7^{a,b}$	$>0.001^{a}$	$22.5^{a,b}$	1.8^{a}	27.5^{a}	4.3^{a}	$31.2^{a,b}$	8.9^{a}	27.5^{b}	6.8^{a}	$36.2^{a,b,c}$	9.8^{c}
a10	27.5^{c}	$0.3^{a,b}$	25.0^{b}	2.9^{a}	23.7^{a}	5.1^{a}	$26.2^{a,b}$	5.9^{a}	$25.5^{a,b}$	5.1^{a}	$28.7^{a,b,c}$	7.0^{a}

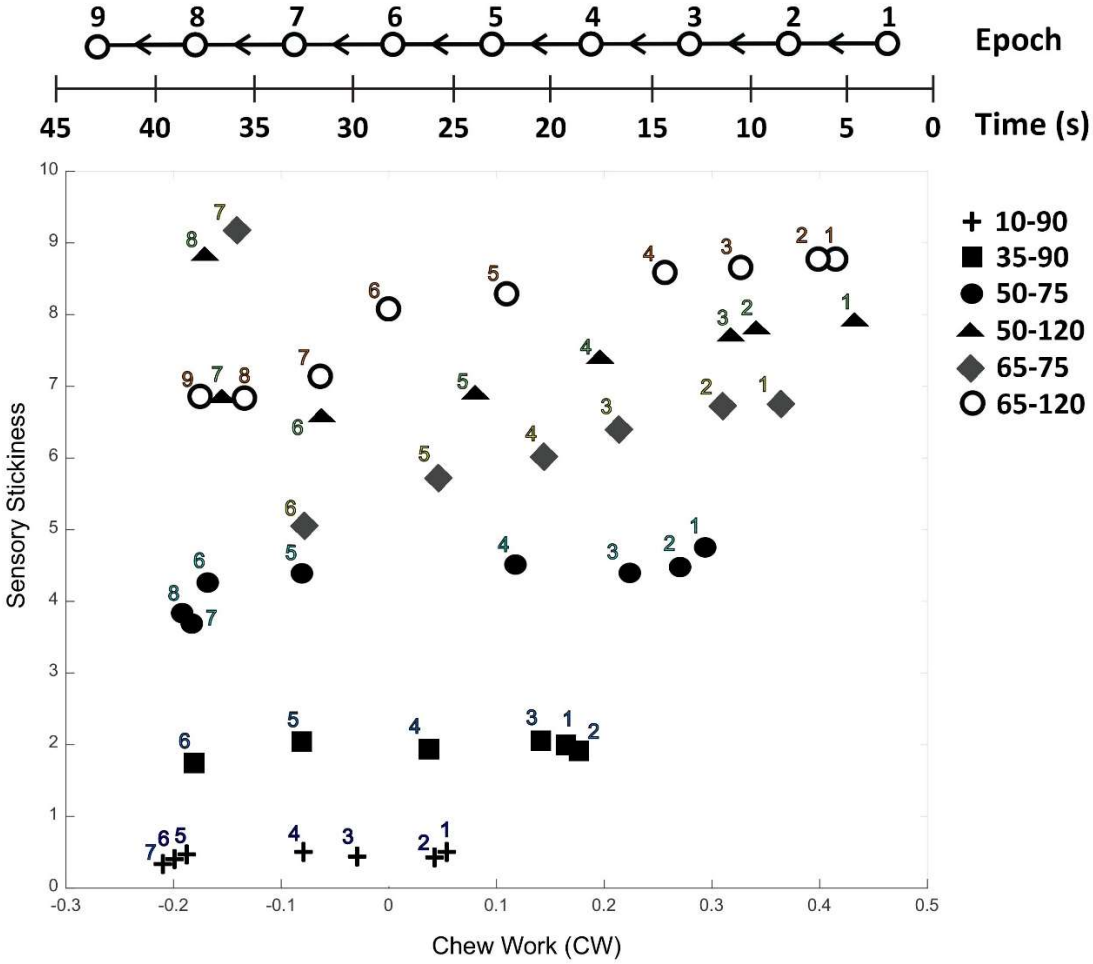
6.6 Texture analyser data

The below table contain different parameters extracted from a single compression test for different batches (values are presented as mean (SD)), (each data point is the average of 9 replications of each batch number of the model food).

Model foods	Batch number	Initial gradient (mN/mm)	force of the adhesive peak (mN)	Distance to the adhesive peak (mm)	Pre-area (mN/mm^2)	Total-area (mN/mm^2)
10-90	1	-95.9 (5.2)	-147.1 (27.8)	6.5 (0.4)	-110.2 (33.7)	-167.7 (72.7)
	2	-87.3 (5.2)	-124 (11.7)	4.5 (1.1)	-81.1 (15.6)	-148.8 (52.3)
	3	-127.4 (43.3)	-112 (37.7)	2.5 (1.6)	-67.8 (46.3)	-98.6 (57.6)
35-90	1	-145.3 (50.4)	-107.4 (42.3)	7.3 (2.3)	-65.9 (30.4)	-133.1 (55.6)
	2	-218.2 (42.6)	-322.4 (82.3)	8.2 (1)	-356 (113.8)	-582.8 (182.8)
	3	-154.5 (4.5)	-123.1 (22.2)	5.6 (0.6)	-69.1 (15.3)	-127.1 (24.2)
50-75	1	-91.5 (7.8)	-582.1 (9.3)	18.6 (1.1)	-5505.7 (295.9)	-13112.5 (803.8)
	2	-66.8 (1.7)	-553.3 (23.5)	13.2 (0.6)	-2659.3 (117.4)	-5693.3 (269.5)
	3	-69.8 (1.6)	-536.6 (19.7)	10.1 (1.3)	-2379.7 (125.6)	-4838.1 (187)
50-120	1	-1357.6 (91.1)	-1670.2 (101.7)	11.2 (0.7)	-7438.1 (542.6)	-31738.5 (4480)
	2	-456.3 (16)	-581.5 (38.1)	9.2 (1)	-2402.7 (172.6)	-9706.6 (841.7)
	3	-356.4 (27)	-542.7 (19.4)	6.6 (0.7)	-2319.6 (104.5)	-9712.1 (700.9)
65-75	1	-660.7 (81.3)	-782.5 (19.6)	12.7 (1.4)	-4219.4 (615.1)	-20645.9 (1576.7)
	2	-113 (7.8)	-367.2 (6.8)	14.7 (0.6)	-3818.2 (177.7)	-9343.6 (641)
	3	-118 (4.6)	-356.9 (8.9)	11 (1)	-3737.8 (206.5)	-9389.6 (475.9)
65-120	1	-5217 (344.8)	-3951.4 (209.4)	10.1 (0.7)	-11867 (1154)	-26020.8 (4343)
	2	-854.1 (26.7)	-769.7 (63.3)	8 (1.7)	-2714.7 (384.7)	-12158 (3245.4)
	3	-781 (22.1)	-737.1 (23.3)	5.2 (0.8)	-2619.3 (145)	-12736.8 (788)

6.7 Black and white version of Figure 3-15

Mean sensory stickiness of all the assessors and their normalised chew work at different time epochs:



6.8 Principal Component Analysis data

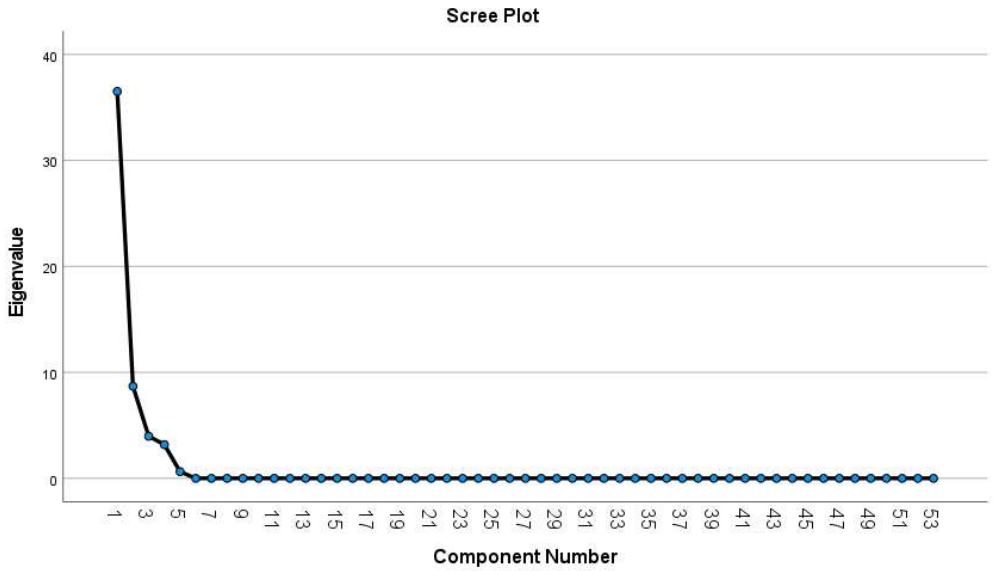
- Total variance explanation:

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total
1	36.520	68.905	68.905	36.520	68.905	68.905	36.399
2	8.693	16.402	85.307	8.693	16.402	85.307	9.295
3	3.975	7.500	92.807				
4	3.183	6.006	98.813				
5	0.629	1.187	100.000				
6	1.406E-14	2.654E-14	100.000				
7	8.134E-15	1.535E-14	100.000				
8	1.341E-15	2.530E-15	100.000				
9	1.280E-15	2.415E-15	100.000				
10	1.103E-15	2.082E-15	100.000				
11	1.035E-15	1.953E-15	100.000				
12	9.438E-16	1.781E-15	100.000				
13	8.789E-16	1.658E-15	100.000				
14	8.223E-16	1.551E-15	100.000				

15	7.491E-16	1.413E-15	100.000				
16	6.479E-16	1.222E-15	100.000				
17	5.955E-16	1.124E-15	100.000				
18	5.122E-16	9.664E-16	100.000				
19	4.861E-16	9.172E-16	100.000				
20	4.521E-16	8.529E-16	100.000				
21	4.218E-16	7.958E-16	100.000				
22	3.339E-16	6.301E-16	100.000				
23	3.017E-16	5.692E-16	100.000				
24	2.217E-16	4.183E-16	100.000				
25	2.122E-16	4.004E-16	100.000				
26	1.830E-16	3.454E-16	100.000				
27	1.557E-16	2.938E-16	100.000				
28	7.106E-17	1.341E-16	100.000				
29	4.952E-17	9.343E-17	100.000				
30	2.987E-17	5.635E-17	100.000				
31	-4.681E-17	-8.833E-17	100.000				
32	-7.226E-17	-1.363E-16	100.000				
33	-1.328E-16	-2.506E-16	100.000				
34	-1.877E-16	-3.541E-16	100.000				
35	-2.171E-16	-4.096E-16	100.000				
36	-2.372E-16	-4.476E-16	100.000				
37	-2.626E-16	-4.954E-16	100.000				
38	-2.843E-16	-5.364E-16	100.000				
39	-3.672E-16	-6.929E-16	100.000				
40	-3.997E-16	-7.542E-16	100.000				
41	-4.331E-16	-8.171E-16	100.000				

42	-4.835E-16	-9.122E-16	100.000				
43	-5.475E-16	-1.033E-15	100.000				
44	-6.197E-16	-1.169E-15	100.000				
45	-6.609E-16	-1.247E-15	100.000				
46	-6.804E-16	-1.284E-15	100.000				
47	-7.578E-16	-1.430E-15	100.000				
48	-8.370E-16	-1.579E-15	100.000				
49	-9.061E-16	-1.710E-15	100.000				
50	-9.729E-16	-1.836E-15	100.000				
51	-1.131E-15	-2.134E-15	100.000				
52	-1.633E-15	-3.080E-15	100.000				
53	-2.064E-15	-3.894E-15	100.000				

- Scree plot:



- Component matrix from PCA. The cells highlighted in red shows parameters primarily explained by PC1 and PC2 ($p < 0.05$)

Parameters	Component		
	1	2	3
Complex viscosity F10	0.988	0.084	0.163
$\boldsymbol{pW}$	0.987	0.103	0.138

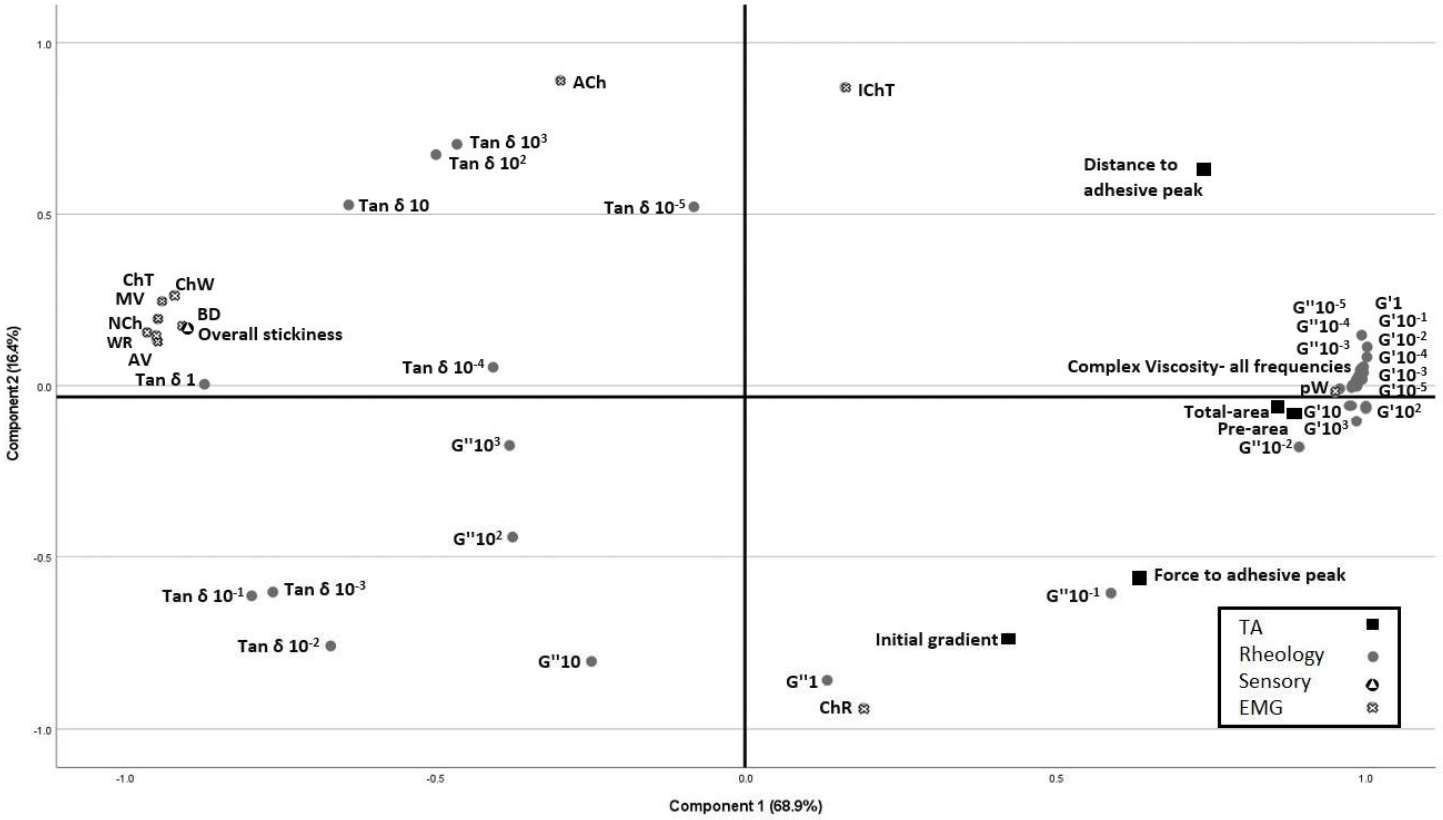
Complex viscosity F10^{-3}	0.986	0.148	-0.346
NCh	-0.986	0.090	-0.037
Complex viscosity F10^{-2}	0.985	0.120	-0.073
G' F10^{-3}	0.983	0.116	-0.088
Complex viscosity F100	0.983	0.063	-0.038
G' F10	0.983	0.073	-0.056
G' F1	0.983	0.083	-0.050
G' F10^{-4}	0.982	0.114	0.120
G'' F10^{-5}	0.982	0.178	0.196
G' F100	0.982	0.068	-0.334
G' F10^{-1}	0.982	0.096	-0.360
G' F10^{-2}	0.982	0.108	0.301
G' F1000	0.982	0.065	-0.314
Complex viscosity F10^{-4}	0.981	0.111	-0.478
Complex viscosity F10^{-1}	0.981	0.092	0.309
G'' F10^{-3}	0.979	0.082	0.278

Complex viscosity F1	0.975	0.066	0.143
ChT	-0.975	0.182	0.140
Complex viscosity F1000	0.975	0.059	0.150
MV	-0.975	0.130	0.156
WR	-0.970	0.083	0.159
G" F10^{-4}	0.968	0.211	0.163
AV	-0.967	0.068	0.168
ChW	-0.958	0.200	0.175
Complex viscosity F10^{-5}	0.956	0.054	0.010
G' F10^{-5}	0.950	0.044	-0.069
OverallStick	-0.931	0.112	0.176
BD	-0.925	0.108	0.321
G" F10^{-2}	0.916	-0.121	0.309
Pre-area	0.891	-0.021	0.318
tanδ F1	-0.871	-0.053	0.498
Total-area	0.864	-0.007	0.634

tanδ F10	-0.716	0.479	0.646
Force of the adhesive peak	0.715	-0.511	-0.002
tanδ $F10^{-1}$	-0.705	-0.659	0.430
G" $F10^{-1}$	0.676	-0.560	0.250
tanδ $F10^{-3}$	-0.672	-0.646	0.020
tanδ $F10^{-4}$	-0.414	0.026	0.224
G" F1000	-0.354	-0.200	0.486
ChR	0.327	-0.917	0.454
$IChT$	0.031	0.871	0.388
ACh	-0.430	0.861	0.388
G" F1	0.257	-0.841	0.261
G" F10	-0.130	-0.811	0.151
tanδ $F10^{-2}$	-0.556	-0.795	0.070
Initial gradient	0.529	-0.701	0.122
Distance to adhesive peak	0.643	0.673	0.163
tanδ F1000	-0.568	0.666	0.197

tanδ F100	-0.598	0.634	0.125
tanδ F10^{-5}	-0.160	0.510	0.168
G'' F100	-0.310	-0.463	0.206

6.9 Black and white version of Figure 3-16



6.10 Publications-1

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Muscle activity during oral processing of sticky-cohesive foods

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ABSTRACT

We investigated muscle activity during oral processing of sticky model foods. Chewing Time extracted from the EMG data distinguished the most sticky and least sticky model foods from the others, but was not a good discriminator between the other models. Mean chew work declined by 25.4%, while the median frequency shift (which is related to muscle fatigue) increased by 54.9% during oral processing for all the model foods, with the effect being greatest for the stickiest foods.

We conclude that the degree of stickiness is not a trigger for swallowing and changes in the other bolus properties, such as softness, may influence muscle activity to a level at which we can swallow.

1. Introduction

The oral trajectory of many foods leads towards a sticky-cohesive bolus, and these sensations remain dominant until the point of swallow for example wheat flake type breakfast cereals [38,45], nut butters [22,50], crackers and Melba toast [48], granola (with yoghurt) [57] and bread [26]. While a sticky-cohesive sensation may be dominant, their magnitude may well change and the purpose behind this study was to investigate whether there might be a threshold for swallowing based on the magnitude of sticky-cohesive sensations, above which we are unable to swallow.

Sticky-cohesive definitions are closely related to each other [10,17,43]. The association of sticky cohesive terms was highlighted by Rosenthal and Thompson [51] and they mentioned that many researchers linked these terms together. Chewing along with the incorporation of saliva are suggested as the main reasons for a cohesive bolus forming in low water, high carbohydrate content foods [22]. Cohesiveness and stickiness (adhesiveness) are features of bolus formation, the former referring to the tendency of a food material to stick to itself, while the latter refers to the material to sticking to external surfaces such as mouth [36].

Stickiness is a multi-dimensional textural attribute and its quantification is a dynamic mechanism and involves complicated processing [1,2,31]. Different parameters such as viscosity, water content, temperature and compression have been suggested to affect the perceived stickiness both sensorially and instrumentally [1,2,16,17,53,56].

Sensory assessment of stickiness can be perceived either by finger touch or in the mouth [6,12,24,42,54].

Hutchings et al. [24], examined stickiness perception of nuts by using the Time Intensity method. Although, no reference samples were provided as anchor points, a high variation was reported between the assessors which was in accordance with other researchers who used such anchors. They suggested that other factors might be responsible for stickiness rankings by the assessors. Stickiness is largely influenced by the levels of low molecular sugar in food systems, e.g., caramel [1,3,25,39,58]. Specifically, prolonged heating times lead to increased hydrolysis of sugars and subsequently higher amounts of low molecular sugars are produced [3]. Surface Electromyography (sEMG or EMG) is a non-invasive method for studying human muscle activity in which the electrical responses of active muscles are recorded. Surface electromyography provides an *in vivo* evaluation of chewing process and its relationship with food texture [11,18,28,30]. sEMG signals are acquired from bipolar electrodes placed adjacent to the masticatory muscles and represents their electrical response during chewing. Studies show that some sEMG parameters can be related to food texture attributes, mainly to assess sensory characteristics [20]. Various studies have used sEMG features to extract quantifiers of the mastication behavior (such as number of chews, chewing frequency and muscle activity) and then correlate them with the texture of the food such as gum [47], meat [4,14], bread [15,19], biscuits [8], pasta [4] and potatoes [15].

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Many different parameters can be extracted from the sEMG data to study the food mastication. Some of the most common sEMG parameters used in eating studies are chew rate, chew time, total number of chews, chew work, number of swallows, maximum voltage (peak-to-peak amplitude), mean voltage, the interval between single cycles and the clearance duration [7,9,32,41,47,52,55].

It is important to note that a large number of existing studies in the literature use a combination of parameters or modify some of the parameters. For example, some researchers undertook the summation of the muscle activity [21,33,44,52] while others divided the mastication process into defined segments such as the whole chewing sequence, the first chew, and the last five chews [34,35,55].

Increased understanding of the oral processing of sticky-cohesive food products and the parameters responsible for trigger of swallowing of such foods can help in formulating food products with specific textures and applications (e.g., dysphagic people).

2. Materials and methods

2.1. Model food preparation

All model foods included granulated white sugar beet (Sainsbury's, UK), native wheat starch (Foo Lung Ching Kee, Hong Kong), citric acid (Sigma-Aldrich, Dorset, UK) and tap water.

The model foods were based on a formulation for Turkish delight but adjusted to achieve the full range of stickiness encountered in our diet. The stickiest of the model foods was comparable to toffee, while the least sticky model food was akin to table jelly.

Model food preparation commenced by heating the mixture of sucrose and water to the desired temperature (Table 2). These temperatures were determined by the complete dissolution of sucrose in the water. When fully dissolved, citric acid was slowly added and the solutions were then heated for 40 minutes (for model foods A, B, C, D) and 60 minutes (for model foods E and F).

Starch and the remaining water were then slowly added (over a period of about 2 minutes). Further heating was undertaken such that A and B had an additional 90 minutes, while the 50g and 65g sucrose foods had an additional 75 and 120 minutes. In order to have a proper mixing, the solutions were stirred manually every two minutes. Finally, model foods were poured into the flexible tubes - these tubes allowed the assessor to hold the sample in the mouth behind the front teeth and then pull the tube out thus squeezing the entire sticky mass into the mouth. Model foods were stored at ambient temperature (20 °C) overnight to fully set.

2.2. Sensory evaluation

Ten assessors (4 females, 6 males) aged between 21 and 27 years old were recruited from students of University of Nottingham, participated in two training sessions followed by two data collection sessions. Assessors were chosen from healthy individuals (self-reported) with at least 28 natural teeth. Exclusion criteria were participants who had dentures, crowns or dental prostheses. Male participants were asked to have a clean shaven before the sEMG sessions so the attachment of the sEMG probes to the skin was possible. Assessors were instructed not to smoke or drink coffee or other strong drinks at least 2 hours before attending their session.

Table 1
Model food formulations .

	A	B	C	D	E	F
Sucrose (g)	10	35	50	50	65	65
Starch (g)	8.5	8.5	8.5	8.5	8.5	8.5
Citric acid (g)	0.3	0.3	0.3	0.3	0.3	0.3
Water (ml)	81.2	56.2	41.2	41.2	26.2	26.2
Desired temp. (°C)	79	84	99	99	108	108
Heating time (min)	90	90	75	120	75	120

Table 2
Terms, definitions and reference samples used in training sessions .

Term	Definition	Reference sample
Enveloping	Leaves residual material on side surfaces of teeth	Werther's original; Creamy toffees (Germany)
Stringy	Forms strings as you pull teeth apart	
Tacky	Adheres to teeth, resists separation	Rowntrees; Fruit gums (Nestle, UK)
Cohesive	Pieces reform together	Maoam; Joystixx Swizzles (Dunhills, UK), Drumstick squashies (Swizzels, UK)
Tooth packing	Packs in teeth - related to quantity that packs	Rowntrees; Fruit pastilies (Nestle, UK)

Five descriptive terms developed by Mayhew 2018 were used in order to define stickiness. In order to prevent any complication of terms, just the definitions were given to the assessors and the actual terms have not been mentioned.

For the training sessions, at least one reference sample was selected from the UK market products in order to provide assessors with an example food of each definition. Terms, definitions and reference samples used in training sessions are given in below Table.

Compusense cloud (Compusense Inc., Canada) running on iPad computer tablets was used for instructions and rating the model foods. Assessors were asked to measure stickiness using an unstructured line (from left side, non-sticky, minimum (0) to the right side, very-sticky, maximum (10) scale in time intervals of 5 seconds after starting point of chewing to the point of swallowing. This was achieved by tapping the iPad screen to mark the line as a measure of perceivable stickiness. After swallowing, assessors were asked to measure the overall stickiness of each model food.

As stickiness is often associated with sweet sugary foods, we blocked the perception of sweetness through the use of a *Gymnema sylvestre* mouthwash. This was prepared using the method of Meiselman and Halpern [40]. Ten milliliters of the mouthwash was served in a 30ml pot. Each assessor was given the mouthwash twice.

2.3. Instrumental measurements of stickiness

A TA.HD Plus texture analyser (Stable Micro Systems, UK) fitted with a P/6 stainless steel probe (6 mm diameter, flat end and circular cross section) mounted on a 5 kg load cell was used for running a tack tests. The test protocol was to bring the probe into contact with the model food moving at 1 $mm.s^{-1}$ and then to apply 10 g force. The probe was then withdrawn from the surface at 10 $mm.s^{-1}$ and the peak area was measured[29]. Nine replicates were taken for each model food.

2.4. sEMG acquisition

The signal was collected by bipolar Ag/AgCl electrodes (Duotrodes 6145, Myotronics, USA) with a center to center distance of 19mm, from the masseter and temporalis muscles of both sides from the assessors' face. The digastricus muscle was considered less useful and it was not considered in the analysis because of the low signal-to-noise ratio (SNR) that can result in false detection errors. An epoch of 5 seconds was chosen to analyze the EMG features along time to match with the epoch used in the sensory analysis. The EMG signal vector was considered as the signal between the first chew to the swallow event. Therefore, the last epoch of each individual is relative to the epoch when the swallow occurs.

The equipment used was a Noraxon Telemyo transmitter (TeleMyo 2400T G2) with a sampling frequency of 1500 samples/second and a digital resolution of 16 bits. Each of the six model foods was served in all

of the four sensory/EMG sessions.

2.5. Signal processing

Figure 1 shows an example sEMG trace after filtering and without normalization. Simultaneous signals from different muscle groups are shown (Left and right Temporalis, Left and right Masseter muscles, as well as the Left and Right Digastricus).

Each chew can be divided into segments using the Double Threshold Onset Segmentation (DTOS) algorithm, which consist in finding the segment in two steps. First, a threshold based on the noise baseline defines the onset (beginning) and offset (end). The threshold (th) is calculated as:

$$th(BL) = \mu_{BL} + k * \sigma_{BL}, \tag{1}$$

where BL is the baseline noise extracted from the signal vector, μ_{BL} is the mean of the baseline, σ_{BL} is the standard deviation of the baseline and k is a predefined factor. If the signal overpass the threshold a onset is detected and when the signal return to a value under the threshold the offset defines the signal segment starting and ending boundaries (signal window). The second step is to verify if the window length (W) between the onset and offset is over a predetermined critical value (W_{crit}). This is because short signals may be associate to signal artifacts which does not represent a chew signal. Therefore, if we call t_{on} and t_{off} the instant of time that the onset and offset are detected, we can say that a segment is detected if:

$$W = t_{off} - t_{on} > W_{crit}. \tag{2}$$

The factor k was heuristically defined iterating in the range of [1:20] with 0.1 steps till the number of found segments converged to the expected value of number of chews using the double threshold (amplitude and time).

Figure 2 depicts the process of segmentation. The sEMG signal is rectified, smoothed and the signal from the masseter and temporalis are summed together. The threshold defines the begin (onset) and end (offset) of each burst. Each pair of onset and offset is used to limit the signal window and are the base to extract signal features.

2.6. Feature extraction

The features used in this work are mainly based on temporal aspects of the sEMG activation: the burst duration, the cycle duration and interchew time. Moreover, the sEMG information within the window (during the burst duration) is also used to construct the features, as the area of the sEMG signal within section is related to the muscle activation (chew work). These aforementioned metrics are illustrated in the Fig. 3.

Denoting the onset and offset of the i-th chew during a trial as t_{on}^{i} and t_{off}^{i}, respectively, the burst duration of the i-th chew (BD_i) can be written as:

$$BD_i = t_{off}^{i} - t_{on}^{i}. \tag{3}$$

The same way we can define the Cycle Duration of the i-th chew (CD_i) as the period between the begin (onset) of the burst and the begin of the next one:

$$CD_i = t_{on}^{i+1} - t_{on}^{i}. \tag{4}$$

The Interchew Time of the i-th chew (ICh_i), which is the period between bursts, is calculated from the period between the end of a burst and the begin of the consecutive burst:

$$IChT_i = t_{on}^{i+1} - t_{off}^{i} = CD_i - BD_i, \tag{5}$$

and it is equivalent to the difference between CD_i and BD_i.

Considering the whole mastication process, the total time (T) is the period between the commencement of the first burst and the end of the

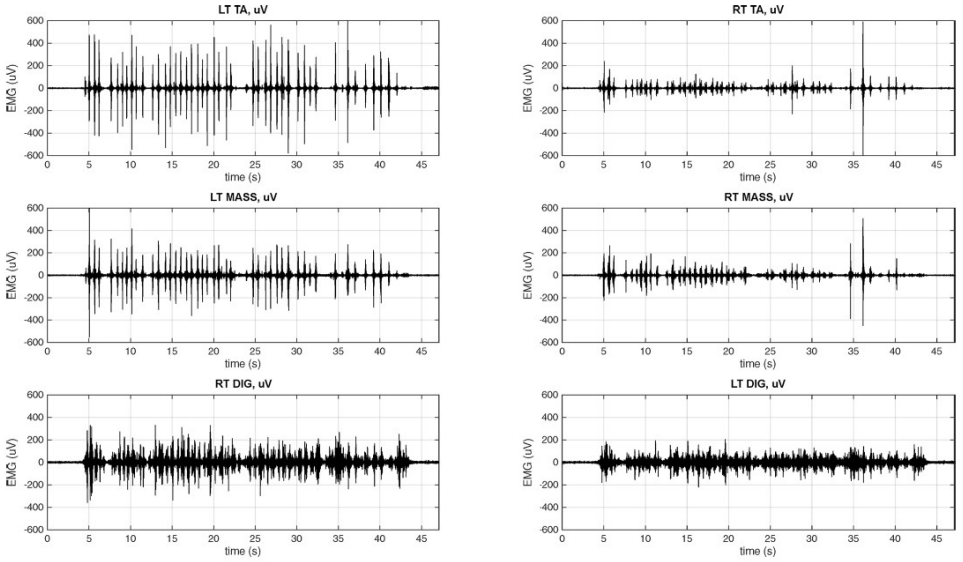

Fig. 1. Plot of a sEMG signal sample from a trial after filtering (without normalization): Left Temporalis (LT TA), Right Temporalis (RT TA), Left Masseter (LT MASS), Right Masseter (RT MASS), Right Digastricus (RT DIG) and Left Digastricus (LT DIG).

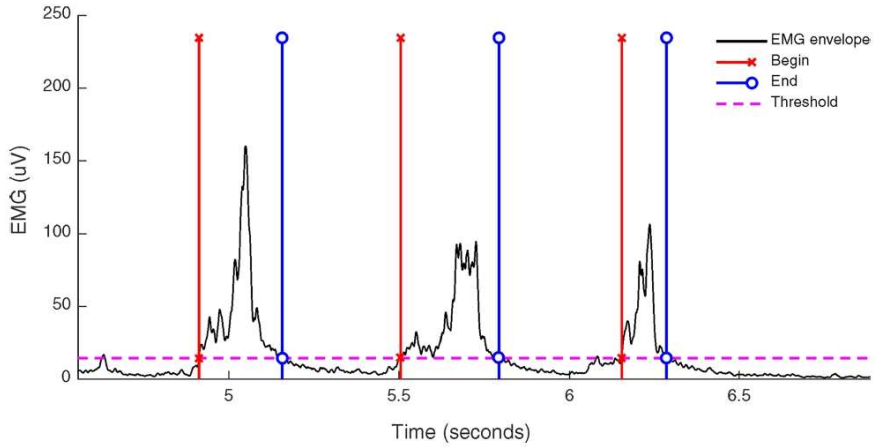

Fig. 2. Signal onset detection and segmentation step. A threshold defines the amplitude that identifies the onset and offset of the signal.

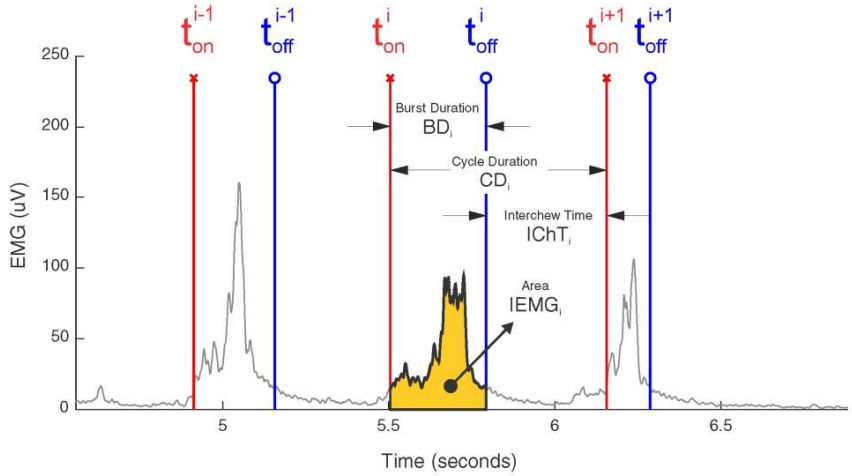

Fig. 3. Signal metrics used to calculate sEMG metrics. The Burst Duration (BD) is equal to the period where the EMG is active, the Interchew Time (IChT) is the period where the EMG is inactive, the Cycle Duration (CD) is the sum of BD+IChT and the Integrated IEMG (IEMG) is the area under the signal within the BD range.

last:

$$T = t_{off}^{M} - t_{on}^{1}, \tag{6}$$

where M is the number of detected chews or the cardinality of the feature vector mathematically expressed by $M = card(t_{on})$.

The area of the sEMG signal within the burst section of the i-th chew ($IEMG_i$), which represent the chew work, is equal to the integration of the sEMG signal (sum of each amplitude within the section). Considering that the i-th chew have length equal to N_i, and being the signal represented by $x_i = \{x_{i,1}, \cdots, x_{i,k}, \cdots, x_{i,N_i}\}$, the area within an analysis window is:

$$IEMG_i = \sum_{k=1}^{N_i} |x_{i,k}|. \tag{7}$$

This metric is a descriptor of a single chew during the process (notated as the i-th chew). To describe the whole mastication process of a food sample a features using the aforementioned metric is calculated.

Before the feature extraction, the signal amplitude was normalized in the range of [0:1] using the maximum values of each assessor. This procedure maintains the signal of every assessor in the same range, which may mitigate bias between individuals. Also, the signal of all four

muscles considered in the analysis were summed up together to form a single signal that represent the mastication process.

The Total Chew Work (ChW) is defined as the IEMG (Area) of all chews in the process González et al. [20]. The first bite is not accounted because it have distinct characteristic from the following chews [37]. This way, considering a mastication process with M chews, the ChW features can be calculated as:

$$ChW = \sum_{i=2}^{M} iEMG_i \tag{8}$$

The Median Frequency (MF) is a frequency at which the EMG power spectrum is divided into two regions with equal amplitude, which is defined by:

$$\sum_{j=1}^{MF} P_j = \sum_{j=MDF}^{M} P_j = \frac{1}{2}\sum_{j=1}^{M} P_j \tag{9}$$

where P_j is the EMG power spectrum at the frequency bin j. This feature is sensitive to the firing of the motor unit, level of recruitment and synchronization [46].

The decline in the spectral characteristics is related to fatigue [13], being the shift of the median frequency a commonly used feature and considered a gold standard for muscle fatigue assessment [27,46].

Therefore, the Median Frequency Shift of the i-th chew in relation to the first bite can be calculated as an indirect measure of muscle fatigue:

$$MFS_i = MF_i - MF_1. \tag{10}$$

Beyond the ChW and MFS features, there are a wide set of parameters that could be derived from the aforementioned metrics [20,34,37]. The Chew Work Rate (WR) is defined as the ChW over the Cycle Duration and the Proportional Work (PW) is the IEMG of a chew over the total ChW. Both metrics are used to measure the muscle effort of a chew. The Average Duration of Chews (ACh), Number of Chews (NCh) and Chew Time (ChT) are metrics directly calculated from the number of detected chews and cycle duration. The Chew Rate (ChR) is the number of chews per unit of time, and it is a measure of chew frequency. The interchew Time (IChT) and Burst Duration (BD) are indirect measures of the phases of the chew cycle: BD measures the period when the muscle is active and IChT the period when the muscle is inactive. The Maximum Voltage Peak Amplitude (MV) and Average Voltage Peak Amplitude (AV) are related to the sEMG activation and are calculated from the peak of the signal during contraction. Amplitude measurements are related to the ChW. Table 3 summarizes the features used in this work.

Some metrics are derived from the whole process (e.g. Number of Chews) and others can be extracted from the individual chew (e.g Chew Work). Features can also be extracted from epochs, which are defined time intervals which could contain one or more detected chews. Features that can describe the characteristics of a single chew or an epoch should be more useful to explore the chewing behavior and how the physiology of the oral processing change over time.

2.7. Statistical analysis

Multivariate general linear model was used with the Tukey HSD post-hoc comparisons in order to highlight significant differences of sensory evaluation results. sEMG features were compared using Friedman's test, which is a non-parametric version of balanced two-way ANOVA, for each pair of model food. A non-parametric test was applied as the null hypothesis of data normality was rejected by a Kolmogorov-Smirnov with $p < .05$. The multiple comparison procedure was performed using the Bonferroni post-hoc correction. The Cohen's d size effect was calculated to elicit differences between the model foods.

Similarity between features was analyzed using correlation as a metric. This was calculated for each pair of features and a tree of hierarchical clusters was defined based on the single linkage algorithm using the inner squared distance.

3. Results and discussion

A linear regression ($R^2 = 0.96$) was obtained when correlating the overall sensory stickiness with the instrumental data. This reinforces the validity of the sensory data.

Fig. 4 shows the sensory response of the assessors to the six model foods. Each assessor evaluated each model food on four occasions thus each result consists of up to 40 evaluations. Analysis of variance (ANOVA) and the Tukey HSD post-hoc test showed which of the model foods were significantly different from each other. Each rectangle has one or more model food codes along with the mean assessor response and standard deviation. Sample codes contained in the same box are not statistically different from each other ($p < .05$) - on some occasions the same sample code appears in more than one box in a column, depending on statistical relation to other samples. The percentage value adjacent to each sample code shows the actual number of assessments included in the statistics - this is because some of the model foods samples were not assessed every time, for example in the column headed "30 sec", some of the assessors will have swallowed the sample before 30 seconds had elapsed and therefore the statistics were based on a smaller number of values.

During the sensory sessions the assessors had two task, to provide temporal values of relative stickiness at every 5 second interval (the six left hand columns of Fig. 4), and to provide an overall value of sample stickiness (the right hand column of Fig. 4).

It can be seen that sample F is the stickiest model food with sample A being measured to have the least stickiness. Generally the stickiness increases with rising sugar content. However, model foods D and E did not follow this pattern, probably due to the action of the citric acid on the hydrolysis of starch and inversion of sucrose during the longer heating period of preparation. It is also possible that greater water losses occurred during lengthy heating, which would fit with Brennan and Mohamed [5] who showed that higher soluble solids led to higher viscosity resulting in a higher stickiness perception. Unlike Brennan and Mohamed who worked with relatively dilute syrups, the gels used in this study were definitely viscoelastic materials.

The results show that at the first stickiness rating point (five seconds), all the model foods (except for D and F) are perceived significantly different. The same pattern occurs after 15 and 20 seconds. Curiously at 10 seconds after the first chew, all the model foods are ranked as significantly different by the assessors. In reality the mean and standard deviation for model foods D and F are close to each other during the first 20 seconds - presumably borderline on statistical significance, sometimes being above the critical value and sometimes below.

We should bear in mind that at long time periods there are less assessors still chewing (Fig. 4, % values) as many have swallowed already, and the reduction in data points reduces the discrimination of the test. This is most evident at longer times (e.g. 25 and 30 seconds) where the action of saliva may have normalized the overall sugar content.

The statistical analysis of the sEMG data is summarized in Fig. 5. The matrix on the left shows which pair of model foods (A to F) are being compared where yellow squares mark which model foods are being compared.The matrix on the right shows the effect size (Cohen's d) in a color scale (where the scale is defined on the side color bar) for each pair of model foods. The columns in matrix are the sEMG features extracted. As a reference, a d of 0.5 can be considered medium difference, while a value of 0.8 represents a large effect size. Moreover, values over 1.2 represents large differences and huge effect sizes if over 2. The asterisk marks (*) and double asterisk marks (**) on the Table represent a significant difference at the significance level of 0.05 and 0.01, respectively.

Results suggest that features based on the sEMG information within the windows (ChW, WR, pW, MV and AV) represent better the

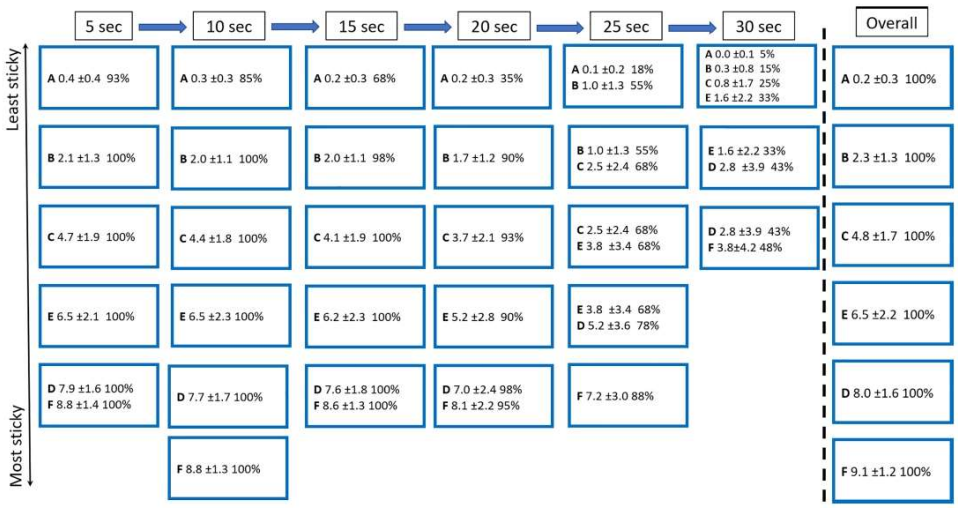

Fig. 4. Levels of stickiness with time and overall stickiness (right hand column). Sample codes in the same box are not significantly different at $p < .05$. Values are mean +/- one standard deviation. The % figure comes from the number of values included in the calculations (based on a maximum of 10 assessors with four assessments each).

differences between model foods than features based on sEMG periods and counting (Ach, ChR, IChT and BD). The feature ChT, which express the total chewing time presented also significant difference for most comparisons. This observation is especially interesting because the ChT feature is relatively simple to extract when compared to other features. The ChW feature is the most distinctive feature. Furthermore, model foods A and B which were measured by assessors the least sticky samples, are more distinctive than the other model foods.

Fig. 5 shows that with respect to the Chew time (ChT - oral residence times), model food A is distinctly different ($p < .05$) from all the other model foods. Similarly model food F has a significant difference with respect to Chewing time from all but model food D, this is in keeping with Fig. 4 which suggests that model foods D and F are similar to each other. Model food D also shows a difference from model food B, yet all other comparisons of model foods have indistinguishable oral processing times. This is not only true for Chew Time, for none of the other parameters can distinguish C from D, or C from E, or D from E, or D from F (see Fig. 5). While the Chew Time appears to separate nine of the fifteen pairs of samples, Figure 6 shows considerable overlap between the Chew Time of the different model foods, making it perhaps a poor discriminator for the different model foods.

A dendrogram plot of the hierarchical binary cluster tree of the extracted features is depicted in Fig. 7. The height represents the distance between the two features being connected. The color is defined by a threshold set at 50% of the maximum height.

From the dendrogram, one can observe that features related to the sEMG signal information (ChW, WR, AV and MV) are close to each other. All those features are related to muscle work information. The Number of Chews (NCh) and the Chewing Time (ChT) are correlated, which is expected as both measures the chew process duration. The average duration of Chews (ACh) and Interchew Time (IChT) are closely related to each other, meaning that the information between these two features are somehow redundant.

We collected a vast amount of data from the sEMG and classified it in terms of: Average Duration of chews; Average Voltage Peak Amplitude; Burst Duration; Chew Rate; Chew Time; Chew Work Rate; Inter-chew Time; Maximum Voltage Peak Amplitude; Number of Chews; Proportional Work; and, Total Chew Work. The inability to distinguish some pairs of model foods (C:D, C:E, D:E or D:F) is likely due to the fact that each food follows a trajectory and therefore, while the model foods all start with different consistencies - requiring different degrees of chewing, with time the muscle activities converge as the boli approach the point of swallow.

There were physiological differences in the sEMG results, indicating inter-assessor variation. By merely looking at the range of values we were able to see some assessors had high levels of Chew Work while others are much more restrictive in their Chew Work activity. It could be that these particular assessors tend to suck the samples rather than chewing them - hence the low values of chew work. Of course this kind of eating behavior could influence their perception of stickiness. Chewing behavior has been the subject of many studies e.g. Rosenthal and Philippe [49] who found gender differences in the rate of chewing hard candy particles, though not altering the overall eating time.

To combat the wide levels of variation between the assessors we sought to normalize the sEMG data for each assessor by subtracting the mean value for the variable concerned and then dividing that by the range of that variable.

$$normalized\ value = \frac{(value - mean\ value)}{range} \tag{11}$$

This has the effect of spreading the variable being investigated across a graph with the mean value for that assessor at the zero point of the normalized axis. By doing this, we can overlay the response to the same variable for all the assessors, the common zero point being each individual's mean and their ranges scaled to their maximum values. Fig. 8 is a scatter plot for the sensory stickiness and normalized chew work data of all the 10 assessors. Scatter is still evident, yet the different samples form overlapping clusters of values and provide a sense of sensory stickiness for the six model foods.

The clustering of samples into groups is apparent in Fig. 8, yet the

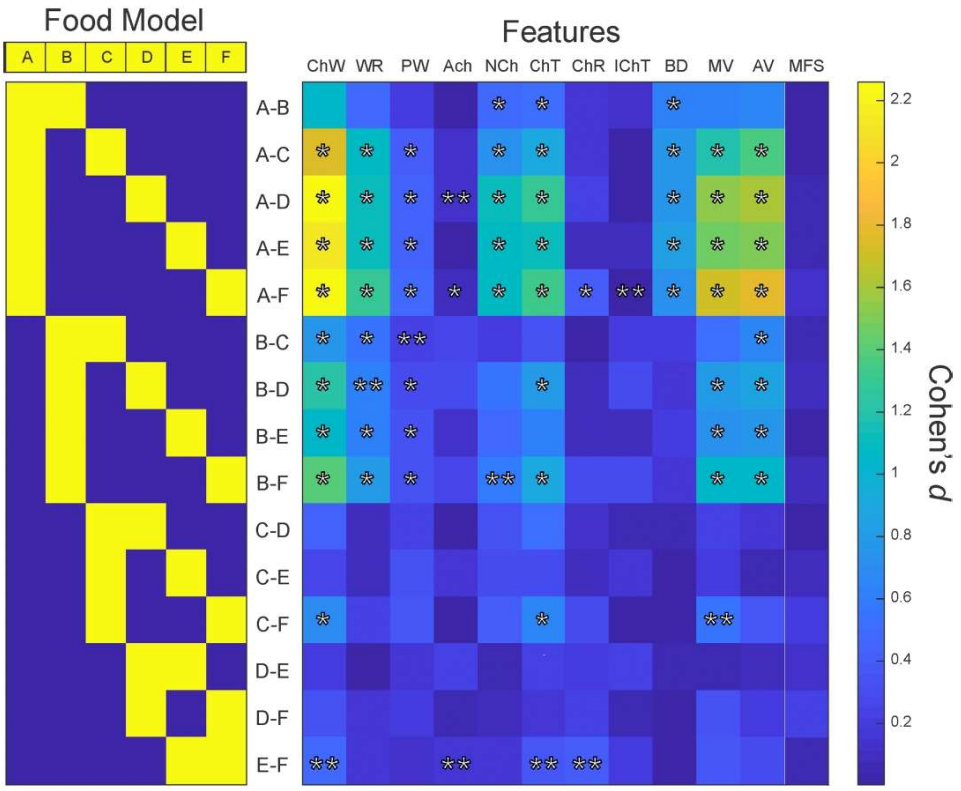

Fig. 5. Effect size (Cohen's *d*) between each pair of model foods. The asterisk marks (*) and double asterisk marks (**) on the Table represent a significant difference at the significance level of 0.05 and 0.01, respectively. All abbreviations are summarized in Table 3.

Table 3
Twelve features extracted and abbreviation.

Feature Full Name	Abbr.
Total Chew Work	ChW
Median Frequency Shift	MFS
Chew Work Rate	WR
Proportional Work	PW
Average Duration of chews	ACh
Number of Chews	NCh
Chew Time	ChT
Chew Rate	ChR
Interchew Time	IChT
Burst Duration	BD
Maximum Voltage Peak Amplitude	MV
Average Voltage Peak Amplitude	AV

variable "Chew Work" does completely separate samples A-B, C-D, C-E, D-E and D-F (refer to Cohen's *d* - Fig. 5). The level of overlap between the data points does show this, and while significant differences may not exist, there are clear groupings of the data. We do need to be cautious when considering this data, for there is a time element lost in the scatter.

In reality each of the model food's cluster of points includes values from the beginning, middle and end of oral processing, moreover the duration of oral processing is different for the model foods. This becomes apparent in Fig. 9 in which each of the assessor response for each of the samples is shown (the colored dots represent different time epochs of five seconds). The scatter data in Fig. 8 is difficult to summarize and so we have redrawn it as line plots in Fig. 9. Each model food enters the oral trajectory at the right hand end of that curve where the epoch is 1 and then moves towards the left. While there is a slight decline in the overall assessor reported stickiness during oral processing, it is only about 20% the initial values for each model food. Yet the span of chew work is considerable for all the foods. Fig. 9 shows some anomalous points at the left hand end of the curves for model foods C, D, E and F, whereby erroneous data points appear towards the left hand end. Just as the sensory data in Fig. 4 is not collected from all the assessors towards the end of oral processing (as some have swallowed), this is also true for the sEMG data and where data is sparse we lose. the moderating effect of averages, hence the impact of individual values becomes more apparent. While we have included such points in the Fig. 9 we have not joined them to the rest of the series with a line. Despite these anomalous values at the left hand end of the Chew Work data, there are clear declining

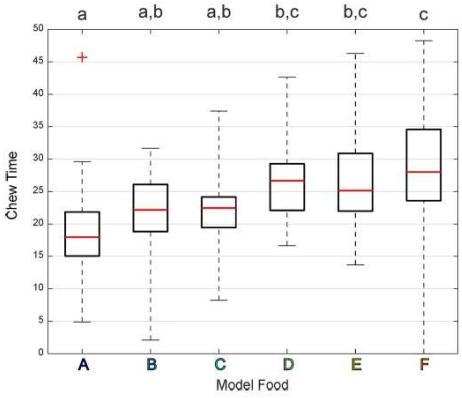

Fig. 6. Boxplot of the Chew Time for each Model Food. The central line in the box indicates the median, the box limits are the 25th (bottom) and 75th (top) percentiles. The whiskers represent extreme data. Outliers (point greater than 1.5 times the interquartile range) are marked as +. Samples that do not share a lowercase letter are significantly different ($p < .05$) based on posthoc pairwise comparisons.

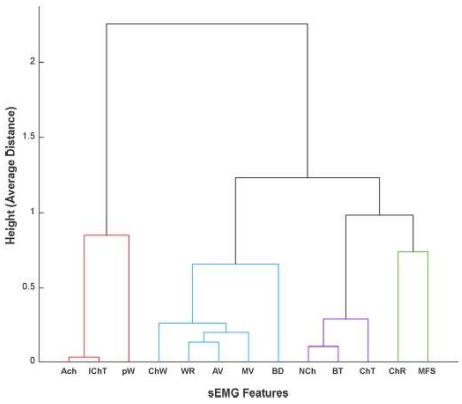

Fig. 7. Dendrogram of the hierarchical cluster tree of the extracted features. The height represents the distance between the two features being connected. All abbreviations are summarized in Table 3.

trends in the normalized chew work for all the model foods.

This research was prompted by observations of researchers such as Lenfant et al. [38] and Rosenthal and Pang [48] who found that low water, high carbohydrate foods gradually become cohesive and sticky towards the end of oral processing. Both of the above mentioned studies used Temporal Dominance of Sensations (TDS) to ascertain the dominant sensations during oral processing. Yet it is well known that while TDS helps us identify the dominant sensation, it does not identify the level of that sensation. The hypothesis of this work was that once sticky-cohesive textures become dominant, the degree of oral stickiness might decline through further oral processing and that when it reached a particular level, it could act as the trigger for the swallowing process. It is however clear from Fig. 9 that with these particular model foods the level of stickiness does not appreciably change during the oral trajectory. Yet regardless of the unchanging stickiness, the chew work does decline during the mastication process. As Fig. 9 shows, a time sequence from right to left, we can see that at the end of oral processing, the chew work has declined to a normalized value of -0.1 for all of the model foods.

In addition to not quantifying a dominant sensation, TDS is limited by the attributes identified for the assessors to choose from. Where an attribute might exist as a bipolar scale with an anchor point at both ends, TDS requires only one of the anchors to be named. Thus in studies such as Lenfant et al. [38] the attribute "hardness" was used and was found to predominate at the early stages of oral processing. But of course on a linear scale where hardness forms one end, softness might be the anchor at the other end. Yet researchers often focus on the positive-decisive attributes with a tendency not to address the negative-weaker anchor points as attributes. Thus while oral processing of bran flakes and dry crackers may start with hard dominant sensations, the reported loss of hardness cannot be tracked towards an increase in softness as the attribute was not available to be chosen. Instead another positive-decisive attribute "stickiness" was offered, and became dominant at the end of the oral trajectory. In this study we found that the level of stickiness of our model foods only slightly changed during oral processing. Despite this, the Chew Work declined substantially, suggesting a change in the consistency of the oral contents towards the point of swallow. Certainly during oral processing there is secretion of saliva and chew work will no doubt mix and soften the bolus. We speculate that perhaps it is a change in softness, which while not identified as dominant in the aforementioned studies, leads to the point of swallowing.

Another related measure of muscle activity is the Frequency Shift which is known to correlate with muscle fatigue. Fig. 10 shows the frequency shift for the different model food samples. Whereas in Fig. 9 time commences on the right where the muscles are fresh in Fig. 10 the first time epoch is at zero on the horizontal axis and subsequent epochs are shown initially on the right where fast twitch muscle fibers are recruited to overcome the varying consistencies of the model foods. Progressive epochs of time move the trajectory through zero on the horizontal axis after which the muscles progressively become fatigued. Model food F which is known to be the stickiest and most cohesive causes the muscles to fatigue the most, while there is a systematic reduction in frequency shift for the less cohesive model foods. Thus model food A only recruits a small number of fast fibers after the first epoch and over the duration of oral processing does not result in much fatigue.

In setting up the model foods, our intention was to control the degree of stickiness. It is however virtually impossible to modify one textural characteristic in isolation of the others. Without doubt the cohesiveness also changed as perhaps did the firmness. We modified the food to examine the oral behavior, yet it is the bolus which is swallowed and the transformation of oral processing further changes the characteristics of the bolus.

If during oral processing the chew work is lessening, then something is happening to the contents of the mouth which allows the muscles activity to decline. In the case of these model foods, the materials start sticky and cohesive, they start as concentrated relatively low water materials. During oral processing, while the stickiness did not decline, they do change in such a way as to require less chew work to be applied. This could be due to increased hydration through the secretion of saliva. Hutchings and Lillford [23] theoretical model sets two triggers for swallowing, the degree of structure and the level of hydration. While these cohesive foods do not change in particle size - remaining as a coherent sticky mass, they do become hydrated through the secretion of saliva. Despite no appreciable change in stickiness, these model foods do change in consistency. The initial firm, cohesive foods tend towards much softer bolus through the mechanical action of the jaw muscles and secretion of saliva. While the Hutchings and Lillford [23] model focuses

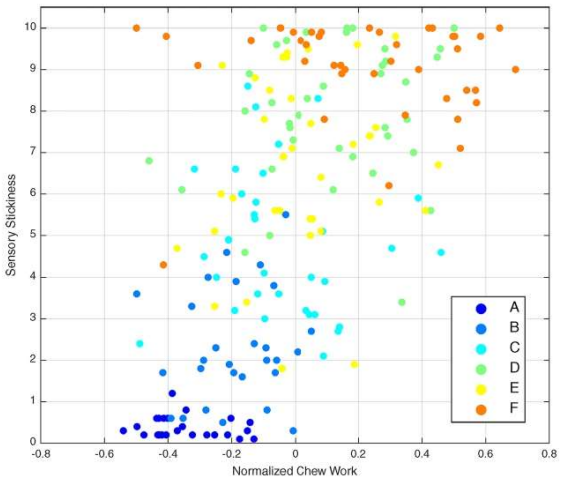

Fig. 8. Sensory Stickiness versus Normalized Chew Work, showing the extent of the overlapping scatter.

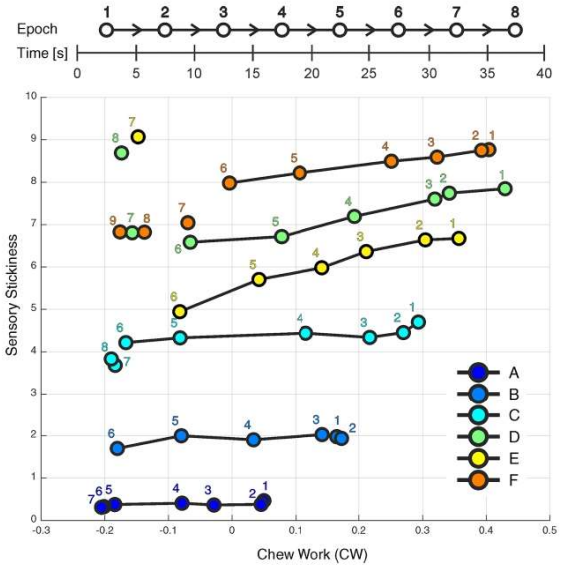

Fig. 9. Normalized Chew Work for model foods along with their mean stickiness at different time epochs.

on the properties of the food and its derived bolus, perhaps the trigger is actually physiological and related more to muscle activity.

4. Conclusion

We have disproved our starting hypothesis, that swallowing is triggered by a threshold level of stickiness. In fact our model foods did not appreciably lose their stickiness during oral processing. We did however observe a reduction in the chew work and an increase in muscle fatigue during the oral trajectory. These changes were greatest for the stickiest samples. We surmise that while stickiness is not changing, there are other textural changes in the consistency of the bolus associated with

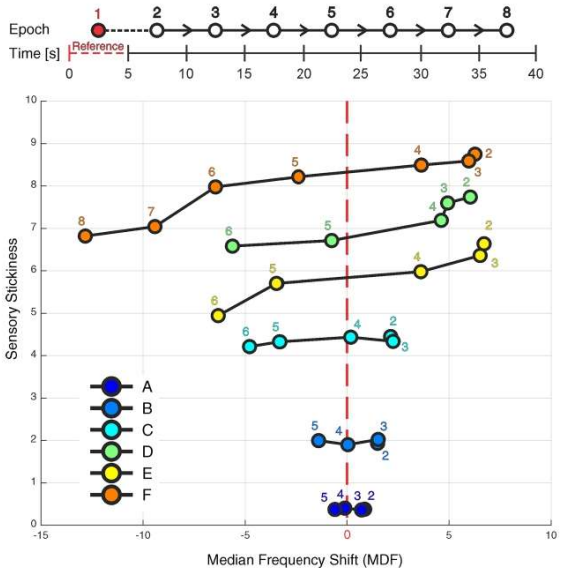

Fig. 10. Sensory Stickiness in function of the Median Frequency Shift (MFS) in relation to the first epoch (reference) for each model food.

an increased level of saliva secretion with time. Such changes might include a softening of the bolus.

Ethics

This study was approved by the University of Nottingham Medical Ethics committee (ethics reference no. 270 - 1803). All participants gave informed written consent.

Declaration of Competing Interest

The authors state that they do not have any conflict of interests.

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6.11 Publications-2

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Observations on the Instrumental Measurements of Liquid Food Stickiness †

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Abstract: While we encounter sticky liquids in our daily life and are able to discriminate between them, instrumental measurements of stickiness are difficult to match to those that relate to our perception. In this paper, we examine some of the factors that influence instrumental measurements of stickiness in liquid foods. The shortcomings of using the maximum peak or the area under the curve are discussed, and a hitherto unused measure, the gradient of the force–distance curve, is suggested as a measure of tension per unit contact area. The zero-perimeter virtual probe, which compensates for the changing meniscus and mass of liquid below it, is introduced. This zero-perimeter approach allows us to extrapolate measures of stickiness such as the gradient of the force–distance curve or the area below that curve. Despite the zero-perimeter correction, there is still a speed dependency on results from instrumentally measured stickiness (for all indexes considered). The speed of the test is responsible for the type of failure (cohesive or adhesive) reported by other authors.

Keywords: stickiness; zero-perimeter virtual probe; initial gradient; maximum peak; area under the curve

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1. Introduction

Stickiness is an important characteristic of many food materials, and it has a major influence on numerous industrial processes. While in some products it is a desirable property, for example in bonding oats together in a breakfast bar, in many it causes problems such as doughs sticking to conveyor belts or material not separating from a depositing mould. Related scientific terms include adhesiveness and cohesiveness, though again, the meanings of these terms are poorly defined. This is compounded by some highly influential papers on texture measurement, which have (perhaps inappropriately) attributed parts of a texture analyser curve to a particular property [1]. Ultimately, understanding what contributes to stickiness is a worthy aspiration, and publications such as Adhikari and coworkers [2] attempt to quantify the different forces involved. They report the study of Brennan and Mohamed, which attempted to correlate the sensory stickiness of sugar solutions with a number of physical measures, finding viscosity and surface tension gave the best relationships.

Fiszman and Damasio [3] surveyed a variety of instrumental food stickiness tests, illustrating the diversity of measures that researchers have used. The normal approach taken is to press a probe onto the food surface and then pull it away while measuring the force. The two most widely used measures are the area under the force–time/distance curve and the peak force to detach the probe from the surface [3].

Possibly, as a result of the engineering limitations of the early texture analysers, many of the publications on stickiness measurement deal with force–time curves. Hoseney and Smewing [4] show the effect of varying the withdrawal speed on the force required to pull a probe away from a surface (Figure 1).

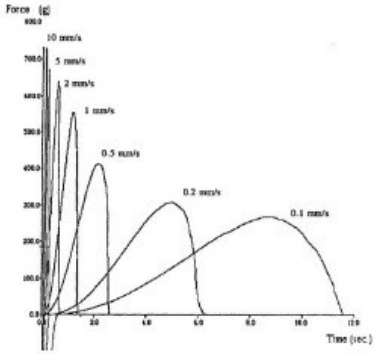

Figure 1. Typical force–time curves of stickiness at different withdrawal speeds (adapted from [4]).

When a probe is slowly raised as if to try and separate it from the surface of a sticky liquid, a column of liquid clings on below. Close to the liquid surface, the meniscus of the liquid spreads out beyond the perimeter of the probe, and the force pulling downwards on the probe is due to the mass of liquid held above the plane liquid surface. Moreover, as the separation of the probe from the plane surface increases, the shape of the meniscus changes as the probe rises from the surface [5].

The aim of this work was to better understand the forces and factors involved in the instrumental measurement of stickiness. We aimed to quantify the forces involved in the stickiness of syrups with the hope that the lessons learned might be extended to other materials.

2. Materials and Methods

2.1. Sample Materials

Several sticky proprietary syrups were used for tests: golden syrup (Tate & Lyle, London, UK), black treacle (Tate & Lyle, London, UK) and clear honey (Rowse, UK). The syrups were poured into plastic Petri dishes, clamped onto the base of the texture analyser and tested as below.

2.2. Texture Analyser and Probes

Stable Micro Systems (Godalming, UK) manufactured a series of three bespoke acrylic probes. One had a single head, but the other two were multiheaded, with three and six heads all milled out of an acrylic block. The contact surfaces of all the probes were on the same plane.

The geometries of the three probes are given in Table 1.

Table 1. Dimensions of the multiheaded probes.

Number of Heads	Diameter of Heads (mm)	Total Perimeter (mm)	Total Contact Area (mm^2)
1	35.0	110	962
3	20.0	190	962
6	14.0	269	962

A TA.HD texture analyser (Stable Micro Systems, Godalming, UK) was used with a 5 kg load cell. The TestMaker application (Stable Micro Systems, Godalming, UK) was used to write a sequence whereby the probe was brought into contact with the liquid surface. It had to remember that position then push 0.3 mm into the material followed by a 10 s pause to try to allow the liquids to achieve good contact. The probe was then pulled back to the remembered position, and there then followed a further 2 min pause. The probe was then withdrawn from the surface at a defined speed until detachment was achieved. Photographic images of the probe liquid contact were taken with an iPhone 7 Plus.

3. Results and Discussion

The spread of the peaks in Figure 1 was resolved by transposing the horizontal time axis to distance. Figure 2 has the same experimental design as undertaken by Hoseney and Smewing [4], with a range of separation speeds, which almost span the capability of the texture analyser used. In Figure 2, we have drawn the curves as negative peaks; however, throughout this discussion, we will refer to the peak as the maximum force (as referring to a minimum force makes little sense).

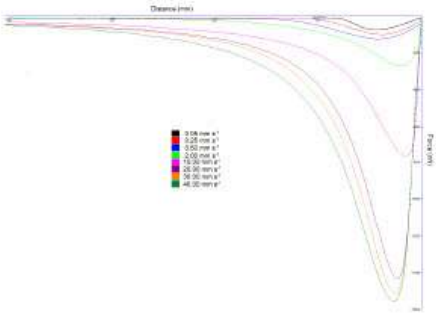

Figure 2. Typical force–distance curves depicting stickiness at different withdrawal speeds for a single-headed probe.

By plotting the distance of separation as opposed to time on the horizontal axis, we observe that the distance to the negative peak is roughly the same regardless of the speed of the probe. This is consistent with Hoseney and Smewing [4], though not at all obvious from Figure 1. In contrast, the distance that the probe moves from the liquids' plane surface to the maximum negative peak is both visibly similar and intuitively the same for all probe speeds in Figure 2. In Figure 1, the areas under the curves are greater for the slower speeds, with the units of this quantity being force–time. In Figure 2, the units are force–distance, and the areas under the curves are greatest for the higher speeds. Moreover, if researchers assign stickiness to the area under the curve, then the derived units of force–distance are energy or work (J).

Figure 3 shows a typical curve for the separation of a probe from the surface of a sticky liquid (at a relatively low speed). When the separation is about 1.5 mm, the liquid is still in good contact with the edge of the probe, though the surface develops a curvature (inset of Figure 3a). Before the peak is reached, the force starts to lessen (inset of Figure 3b) as the curve starts to flatten; this is followed by the narrowing of the column of the liquid joining the probe to the body of liquid below. The curvature of the glucose syrup as the probe pulls away exhibits a concave necking (inset of Figure 3d), holding firmly to the probe perimeter and narrowing within the liquid itself (this is shown diagrammatically in Figure 4d). In contrast, the inset image with a broken line (inset of Figure 3c) is for

golden syrup at the peak, and while the column of the liquid thins, it does not hold fast to the perimeter of the probe but forms a narrowing cylinder attached to the base of the probe (this is shown diagrammatically in Figure 4b). After the peak is passed, the force continues to reduce as the thickness of the column of the liquid progressively thins (e.g., inset Figure 3e).

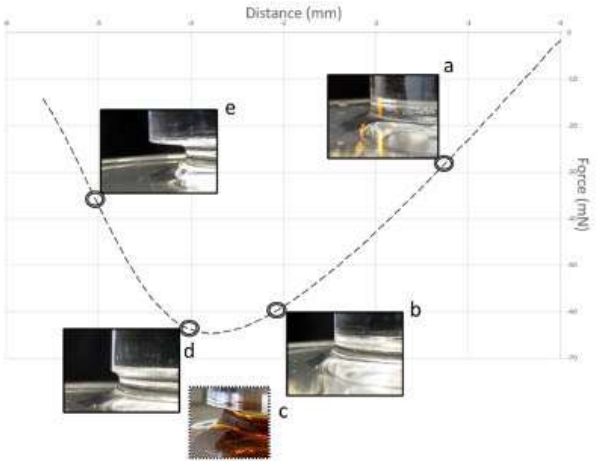

Figure 3. Force–distance curve for glucose syrup with images of the probe contact surface. The inset images with a broken border are golden syrup at its force–distance peak.

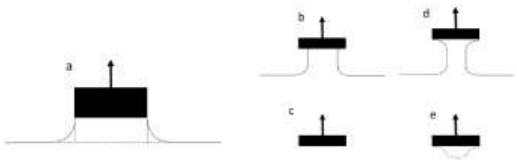

Figure 4. Schematic view of the stickiness test of liquids. (**a**) Starting state and early stages of the pull—linear portion of the curve; (**b**) narrowing liquid column—loss of adhesion; (**c**) adhesive detachment; (**d**) narrowing liquid column—necking; (**e**) cohesive failure.

Stickiness has been attributed to the interaction of liquid viscosity and the interfacial properties between the probe material and the liquid [2]. Figure 3 shows a linear region at the start of probe withdrawal, during which time the perimeter is fully in contact with the liquid. The inset images in Figure 3 show the curvature of the liquid observed during these early stages of probe withdrawal. The forces acting downward on the probe are due to the surface tension but also the mass of the liquid below the probe and within the truncated annular region beyond the cylindrical perimeter of the probe (Figure 4a). During the linear part of the force–distance curve, during the stages of probe withdrawal from the surface, the curvature of the liquid adhering to the perimeter of the probe is a complex relationship, which changes as the probe progressively moves up [5].

In an attempt to compensate for the effects of curvature, we developed a practical (nontheoretical) solution that utilises multiheaded probes with a common surface area in contact with the liquid. The dimensions of these probes are shown in Table 1. Despite having the same contact area in touch with the liquid, if we sequentially undertake the

stickiness test outlined above with each of these probes, we produce three different curves that do not superimpose on each other (Figure 5). While the curves in Figure 5 are all of a similar basic shape, the curve features, such as distance to reach the maximum negative peak, force at the maximum negative peak, area under the curve and gradient of the linear portion of the curve, are all slightly different. However, if we plot these features against the total probe perimeter, we unsurprisingly obtain good straight lines.

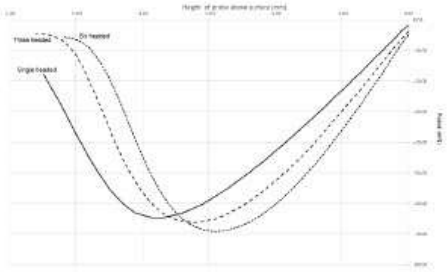

Figure 5. Force–distance curves for glucose syrup with constant area (varying perimeter) probes. The probe withdrawal speed is 0.01 mm·s^{-1}.

Furthermore, if we extrapolate such lines to zero, we effectively obtain the force we would obtain from a zero-perimeter probe. Of course, such a probe does not exist, yet the force exerted on such a virtual probe would be solely due to the mass of the liquid in a cylinder below a 962 mm^2 probe.

Our zero-perimeter virtual probe overcomes problems with the unpredictable meniscus; however, we can see from Figure 3 that, strictly speaking, the known contact area (962 mm^2) is only valid during the linear part of the curve, and this in reality excludes the use of parameters such as the peak force or the area under the curve. However, we continued to make use of the parameters of peak force and the area under the curve as measures of stickiness, albeit with our zero-perimeter curve. At least the zero-perimeter probe excludes the effect of the curved meniscus and as such better defines the system.

If the purpose of a stickiness test is to determine a defined material property, then the dimensions of the geometry are important. That is to say, in an ideal world we would be able to express the stickiness per m^2. Yet, once the curve begins to deviate from a linear force–distance behaviour, the sample geometry has changed. Ideally, we should obtain our measure of stickiness from the linear portion of the curve, otherwise we are measuring a property of ill-defined dimensions. Certainly, during the linear part of the force–distance curve, there is neither separation of the liquid from the perimeter nor necking of the sample, thus the instrumental readings obtained will relate to the contact area of the probe. Moreover, if we consider that property our zero-perimeter virtual probe, we can overcome changes in the meniscus. On this basis, using a property of this linear region such as its gradient would perhaps give us a better measure of stickiness and one that can be related to the probe geometry. Appropriately, the units of this gradient are force per unit distance (Nm^{-1}); in other words, a tension, which is possibly a better match for stickiness than either the peak force or work used to separate the probe. At low test speeds, the gradient of the linear portion of the force–distance curve is straightforward to measure; however, as is apparent from Figure 2, at higher speeds, and especially with the more viscous liquids, the curve is almost vertical. It is as if the liquid cannot flow or yield, with the stress exerted by the texture analyser being unable to be dissipated, thereby rapidly rising. Although in Figure 2 these high-speed curves appear to superimpose upon each other, if the horizontal axis is enlarged, we can discriminate between them.

Figures 1 and 2 emphasise the importance of probe withdrawal speed on the curve obtained. Making use of our multiheaded probes, we were able to estimate the various parts of the curves of our zero-perimeter virtual probe, being withdrawn from different sticky liquids at a range of speeds. Reminding ourselves that the purpose of these tests is to measure stickiness as a characteristic property of the material, we attempted to examine the relationship between the initial gradient, the maximum negative peak force and the area under the curve of the zero-perimeter virtual probe as functions of withdrawal speed.

To cope with the wide range of withdrawal speeds employed to collect the data in Figure 6, we plotted the withdrawal speed on a log axis. Figure 6b,c appears to show a discontinuity, remaining relatively low at slow speeds and then increasing logarithmically after some critical value, which is specific for each liquid.

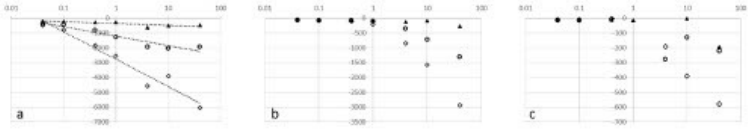

Figure 6. Influence of probe withdrawal velocity on the parameters of the zero-perimeter virtual probe curves. (**a**) Area under the curve; (**b**) peak force; (**c**) initial gradient. ∘ Golden syrup; ◇ black treacle; ▲ honey.

Kilcast and Roberts [6] introduced the idea of adhesive and cohesive failure based on whether a sticky liquid leaves a residue on the probe or separates cleanly when a probe is pulled out of a sticky material. We concur with Noren, Scanlon and Arntfield [7], who observed that the cohesive/adhesive failure behaviour actually depends on the speed at which the test is undertaken. We speculate that as the probe is withdrawn from the liquid surface, at low speeds, the liquid is able to flow back to the liquid bulk, allowing it to cleanly separate from the surface of the probe, progressing from Figure 4a,b and finishing at Figure 4c—i.e., adhesive failure. In contrast, once we exceed the critical speed that marks the discontinuity in Figure 6b,c, the separating probe moves faster than allows the liquid to flow back to the bulk with the result that the probe liquid behaviour progresses from Figure 4a,d ending with Figure 4e—i.e., cohesive failure.

Earlier, we commented on the almost vertical force–distance curves of the high-speed tests, and the idea that the material cannot dissipate the stress through flow or relaxation is consistent with stresses building within the material until that material cannot support further stretching and undergoes catastrophic failure. If the separation of the probe from the surface of the liquid is faster than allows the liquid to flow, we get cohesive failure. In this study, we are dealing with viscous syrups. Many foods are glassy materials, which are often considered to be supercooled liquids. Such materials have immensely high viscosities, which would be difficult to flow in the time frame of the test protocol outlined here. Thus, we would expect glassy materials to undergo cohesive failure. Some other foods are viscoelastic (e.g., doughs), and in such situations, we expect the failure to depend on the predominance of viscous and elastic elements present. In the case of viscoelastic materials, both the ease of flow and the elastic limits of the material will dictate whether the texture analyser imposed force can be adequately dissipated in the time frame of the test or whether we will end with cohesive failure.

In our daily life, we experience the phenomenon of stickiness in the liquids we interact with, and perhaps the approach we should take from a physical testing point of view is to employ separation speeds akin to those employed in the manual manipulation of materials with our fingers and jaw motion. Shama and Sherman [8] used a similar approach in recommending shear rates with which to evaluate the viscosity of liquids if they are to match our human experience.

4. Conclusions

Clearly, the data collected to measure stickiness of liquids have a huge dependence on the speed and geometry of the test being undertaken. We might even go so far as to say the results are artefacts of the test method employed.

Our zero-perimeter virtual probe overcomes problems with the unpredictable meniscus. The probe is only in full contact with the probe during the linear region of the force–distance curve, and as such, perhaps the gradient would give us a better measure of stickiness and one that can be related to the probe geometry. In contrast, while the peak force and the area under the curve are not able to relate the force to the geometry of the probe, the zero-perimeter virtual probe does reduce some of the variability in results arising from the curved meniscus.

Author Contributions: S.M.K. and A.J.R. designed the experiments, analysed the data and wrote the manuscript together. A.J.R. conceived and designed the multiheaded probes. S.M.K. undertook the data collection. All authors have read and agreed to the published version of the manuscript.

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6.12 Publications-3

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RESEARCH ARTICLE

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Artifacts and errors in the measurement of the stickiness of liquid foods with tack tests

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Abstract

We encounter stickiness in many areas of our daily life and as humans, we are able to discriminate different levels of stickiness. Yet attempts to measure stickiness with instruments have been challenging. One of the commonest approaches has been the "tack test" in which a probe is brought into contact with the sticky food and then pulled away while measuring the resisting force—various indices, such as the maximum force or the area below the force curve have been used to describe stickiness. This work rationalizes results from tack tests for liquid foods and helps us explain the influence of probe geometry. Photographic evidence of the way that the liquid adheres/detaches from the probe suggests that the terms "cohesive" and "adhesive" failure depend on the speed of the test. Application of a fixed deformation with time shows rapid loss of adhesive force suggesting that liquid samples flow from the probe. We propose that stickiness of liquid foods is entirely due to the liquid's viscosity and surface tension, and that measurements of tack for liquid foods—while highly reproducible—are entirely artifacts of the test method employed and are in effect snapshots in time of non-equilibrium processes.

KEYWORDS
artifacts, instrumental measurements, stickiness, tack, viscous flow, zero-perimeter virtual probe

1 | INTRODUCTION

We encounter stickiness in many situations in our daily lives. From a food processing perspective, it can be both a curse (with doughs adhering to a conveyor belt) or a blessing (with oats forming clusters in breakfast cereals). From a texture measurement perspective, stickiness is often referred to as adhesiveness and a number of empirical measurements of both stickiness and adhesiveness exist (Fiszman & Damasio, 2000). One such test is the "tack test" in which a probe is brought into contact with food and then the tensile force is measured as the probe is pulled away (Budelmann, Schmidt, & Meiners, 2020; Werner, Jones, & Paterson, 2007). Such a test was undertaken by Hoseney and Smewing (1999) who showed a dependence of test speed on the results obtained (Figure 1).

Tack has been the subject of numerous studies in food and material sciences. Its complexity of measurement has been highlighted by several of the researchers including Bormashenko, Whyman, and Pogreb (2009), Bosc, Ferrari, and Michon (2008), Budelmann et al. (2020), Noren, Scanlon, and Arntfield (2019), and Werner et al. (2007). Tack can be related to the area of contact just before detachment occurs (Hui, Lin, & Baney, 2000). Tack provides a measure of adhesiveness or stickiness and relates to separation of a surface from a sticky material. A related term is cohesiveness which has been defined as "the strength of the internal bonds making up the body of

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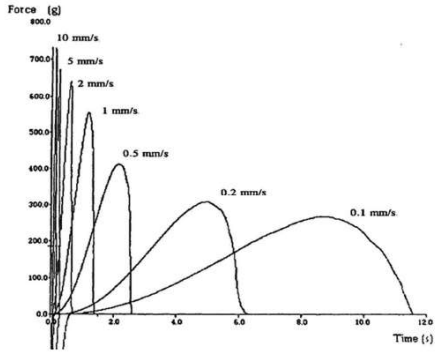

FIGURE 1 Typical force-time curves of stickiness at different withdrawal speeds (adapted from Hoseney & Smewing, 1999)

the product" (Szczesniak, 1963). The difference between cohesion and adhesion was suggested by Kilcast and Roberts (1998) who investigated the failure of samples in tension. Failure at the contact surface between the probe and the food has been termed adhesive failure, while samples that bind tight to the probe and break apart within the food itself are said to exhibit cohesive failure. It is notable that Noren et al. (2019) found that the type of failure one obtains in tack testing depends on the conditions used, with high separation speeds leading to cohesive failure while slow speeds result in adhesive failure.

"Adhesiveness" has become a standard term in the food texture lexicon partly through its assessment in texture profile analysis (TPA) (Friedman, Whitney, & Szczesniak, 1963). Despite its popularity, this test protocol has been critically scrutinized and discredited in recent years (Nishinari, Fang, & Rosenthal, 2019). In TPA, adhesiveness relates to the area below a force-time curve resulting from the removal of a plunger from a squashed sample. There are parallels to this measure and the tack test as some researchers have used the area below a force-time curve as a measure of stickiness. Michalski, Desobry, and Hardy (1997) reviewed the theories of adhesion. Factors such as the material from which the probe is fabricated, sample-probe contact angle, nature of the sample, and influence of temperature and pH have been identified as influential (Bormashenko et al., 2009; Bosc et al., 2008; Gay & Leibler, 1999; Russell & Kim, 1999).

Ultimately a better understanding of what contributes to stickiness is a worthy aspiration and the aim of this work. Publications such as Adhikari, Howes, Bhandari, and Truong (2001) attempt to quantify the different forces involved. One might expect sensory stickiness to be related to the physical properties of the materials involved and such measurements are well documented in the literature, for example, Golden syrup is reported to have a density of 1,430 kg/m^3 and a viscosity of about 210 Pa/s (Beckett, Mader, Phillips, Rust, & Witham, 2011). Brennan and Mohamed (1984) correlated sensory stickiness of syrup solutions with a number of physical properties (specific gravity, surface tension, viscosity, tack test, and back extrusion). They found good correlations between sensory stickiness with both viscosity and surface tension. Interestingly they did not seem to find a correlation between sensory stickiness and the instrumental tack test—this might relate to the conditions they were using as may be seen later in this paper. The insight that surface tension influences the stickiness, points us to the complexity of the curvature of the liquid surface which adheres to the probe (Henriksson & Erisson, 2004). As the probe is pulled away from the surface of the liquid, a strand of liquid is pulled with it. At the start of a tack test, a meniscus exists between the plane surface of the liquid and the flat end of the probe (Figure 2, upper schematic). The force pulling down on the texture analyzer is due to the mass of liquid directly below ($h\pi d^2/4 \times$ density of the liquid), the mass of liquid in the conical annular region bounded by the meniscus ($V_m \times$ density of the liquid) as well as the surface tension of the liquid. Of course, the contact angle between the liquid and the probe influences the curvature of the meniscus and therefore the material from which the probe is manufactured has an impact. As the probe is raised the shape of the meniscus changes. As the probe is pulled up a strand of liquid clinging to the probe rises from the liquid reservoir below. At low levels of "h" the strand has a concave surface (xy plane), yet as the probe is raised the liquid begins to thin in the xz plane. The narrowing is not uniform, because initially it tends to stick to the probe perimeter, narrowing in the middle of the strand and giving the characteristic "sticky" strand. Obviously, the geometry of the probe is important as small diameter probes have a tighter curvature than those with a large diameter and this in turn complicates the curvature of the liquid surface. Kazemeini and Rosenthal (2021) devised a non-theoretical, practical way of compensating for the surface curvature by measuring tack with three probes shown in the lower part of Figure 2. The multi-headed probes were designed to have a constant surface contact area in the same plane, yet with varying total perimeters. The intention in using these probes was to enable any measurement (e.g., maximum force) to be taken from the tack test to be plotted against total perimeter and the resulting data extrapolated to a perimeter of zero.

Of course, a zero perimeter probe does not exist, but the trend in parameters extracted from the tack test curve showed linear response with changing perimeter from this probe series (Kazemeini & Rosenthal, 2021). In addition, the zero perimeter virtual probe has other advantages, as there is no perimeter there is no contact angle and so the material of probe construction becomes irrelevant, moreover, the force acting on the texture analyzer is purely due to the mass of liquid directly below the plane surface of the probe whose geometry is known.

The underlying hypothesis behind this work was whether adhesiveness of liquid foods can be reliably related to some measure resulting from a tack test.

2 | MATERIALS AND METHODS

2.1 | Sample materials

Several sticky proprietary syrups were used for tests: Golden Syrup (Tate & Lyle, London, UK), Black Treacle (Tate & Lyle), clear honey

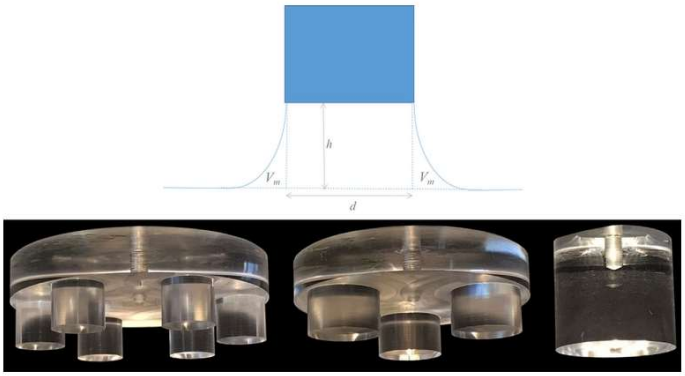

FIGURE 2 Schematic of liquid meniscus during the tack test and Kazemeini and Rosenthal's constant area probes

(Rowse, Wallingford, UK), and glucose syrup (Nordic Sugar, Malmö, Sweden). Syrups were poured 10 mm deep into petri dishes. These were clamped onto the base of the texture analyzer and tested as below with the probe series described in Figure 2. All experimental work was undertaken at 20°C in a temperature-controlled laboratory.

2.2 | Probe geometries

Three probes were fabricated from acrylic. Each with a contact area of 962 mm^2. These consisted of a single-headed probe which was 35 mm diameter (110 mm perimeter). A triple-headed probe, with each head being 20 mm indiameter (giving a total perimeter of 190 mm) and a six-headed probe witheach head being 14 mm in diameter (hence 269 mm total perimeter).

2.3 | Tack tests

A TA.HD texture analyzer (Stable Micro Systems, Godalming, UK) was used with a 5-kg load cell. The TestMaker application (Stable Micro Systems), was used to write a sequence whereby the probe was brought into contact with the liquid surface, the position was then stored in the instrument's memory, the probe was then pushed a further 0.3 mm into the material followed by a 10-s delay to try to allow the liquids to achieve good contact. The probe was then pulled back to the remembered position and then allowed to equilibrate for 2 min. The probe was then withdrawn from the surface at a defined speed between 0.05 and 40 mm/s (representing the range of speeds available to the texture analyzer) until detachment was achieved. Tests were carried out in duplicate and a macro was used to extract peak force, peak area, the height of the probe from the surface at the peak, and the initial gradient of each curve. For each test speed the values of these parameters were plotted against probe perimeter and extrapolated the intercept (i.e., zero perimeter). Photographic images of the probe liquid contact were taken with an iPhone 7 plus.

Data acquisition ranged from 10 points per second (pps) for the slow tests, to 200 pps for the higher speeds.

2.4 | Fixed distance step change tests

A TA.XT.plus texture analyzer (Stable Micro Systems) with a 1,000-g load cell was used. The TestMaker application (Stable Micro Systems), was used to write a sequence whereby the probe was brought into contact with the liquid surface, the position was then stored in the instrument's memory, the probe was then pushed a further 0.3 mm into the material followed by a 10-s delay to try to allow the liquids to achieve good contact. The probe was then pulled back to the remembered position and then allowed to equilibrate for 1 min. Having first undertaken a tack test as described above, the texture analyzer was programmed to raise the probe at the same speed as the tack test, to a height within the linear portion of the force-time curve (about two thirds up). The height of the probe was then held constant while the force exerted on the texture analyzer measured over time.

3 | RESULTS AND DISCUSSION

The spread of the peaks in Figure 1 is resolved by transposing the horizontal time-axis to distance. Figure 3 has the same experimental design as undertaken by Hoseney and Smewing (1999), with a range of separation speeds spanning the capability of texture analyzer used. In Figure 3 we have drawn the curves as negative peaks as they are in tension, however, throughout this discussion we will refer to the height of the peak as the maximum force (as referring to a minimum force makes little sense).

By plotting the distance of separation, as opposed to time, on the horizontal axis we observe that the negative peak is roughly at

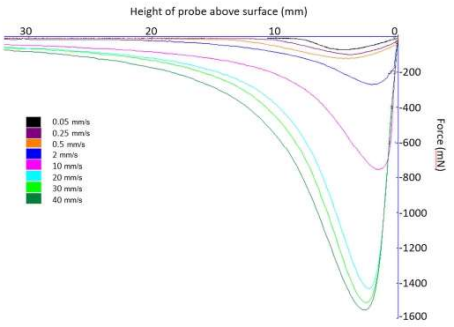

FIGURE 3 Typical force-distance curves depicting stickiness at different withdrawal speeds for a single-headed probe

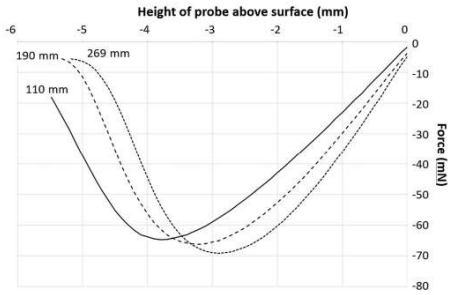

FIGURE 4 Force-distance curves of glucose syrup with constant area (varying perimeter) probes. Probe withdrawal speed is 0.01 mm/s

the same position regardless of the probe velocity. This is intuitively what one might expect and consistent with observations made by Hoseney and Smewing (1999), though not at all obvious from Figure 1. Additionally, the areas under the curves in Figure 1 were greater for the slower speeds with the units of force-time. In Figure 3 the units are force-distance and the areas under the curves are greatest for the higher speeds. Moreover, if researchers assign stickiness to the area under the curve (as some do) then the derived units are force-distance (Nm) which equate to Joules of energy or work.

Fiszman and Damasio (2000) reviewed the parameters that different researchers have used as a measure of stickiness; the commonest of these are peak height and area under the curve. By undertaking identical tack tests with each of the three Kazemeini and Rosenthal constant area probes, we see three separate curves, each with a similar basic shape, yet skewed and shifted along the distance axis (Figure 4). By extracting peak heights or areas under the different curves in Figure 4, we can plot them against probe perimeter and extrapolate to zero. In doing so we are predicting the peak height or area below the curve which we would expect from the zero perimeter virtual probe. Of course, these are predicted for a meniscus-free test. In addition to peak height and area under the curve, there are other differences apparent in Figure 4, such as the height of the probe above the plane surface of the liquid when the maximum force reached, and the gradient of the initial slope (i.e., top right moving towards the bottom left) is steeper as the number of heads and total perimeter increase. While the latter curve parameter has not previously been used as an index of stickiness it does have the advantage of having the units of force per unit distance (N/m) which is of course a tension.

At the start of this study we had (perhaps naively) thought that we might be able to establish an index of stickiness, whereby for any given liquid we could express the stickiness as force per unit area. The zero perimeter virtual probe would perhaps allow us to do this. However, the data in Figure 4 originate at a fixed speed of 0.01 mm/s and it is apparent from Figures 1 and 3 that the speed of withdrawal influences the result obtained. Thus we sought to examine the relationship between probe speed and the maximum peak height, the area under the curve, height of the probe at maximum separation, and the initial linear gradient of the force-distance curve. Through repeated experiments we were able to determine these parameters with the three probes in our series and then extrapolate to a zero perimeter virtual probe; these results are shown in Figure 5. We employed a wide range of speeds available with the texture analyzer, covering 4 orders of magnitude (0.04–40.00 m/s). In order to avoid low-speed values overlying each other in Figure 5, we have plotted the horizontal axis as a logarithmic scale. Bearing this log scale in mind, Figure 5a shows the influence on the peak force and we have fitted a linear best line with increasing speed while Figure 5b, the area under the curve, shows a good log fit for all the syrups being considered. Figure 5c, the height of the probe at the peak, does not seem to show much variation, let alone trend, with increased speed. On seeing this result we undertook a single factor analysis of variance of the probe height (at maximum force) and found no significant difference. At low speeds, the initial gradient values of the different syrups (Figure 5d) all overlie each other, but at higher speeds they deviate from the baseline. However, the scatter in the high-speed values is quite large and no clear relationship can be ascertained.

It will be noted that the initial gradient of the force-distance curve can be very steep at high speeds of separation especially for the more viscous liquids. It is obvious from Figure 3 that many of the initial curves overlap each other and are difficult to discriminate. This is apparent in Figure 5d where the higher speeds of probe withdrawal do not seem to follow a consistent pattern.

Something which intrigued us in Figures 1 and 3 was the fact that all the curves start to flatten out as they reach the maximum. We had expected the peak force to increase continuously until the probe separated from the food. To investigate this further we took photographs of the liquid contact with the probe during the test. Figure 6 shows a typical curve for the separation of a probe from the surface of a sticky liquid (at a relatively low speed). When separation is about 1.5 mm, the liquid is still in good contact with the edge of the probe, though the surface is developing a curvature (inset Figure 6A). Before the

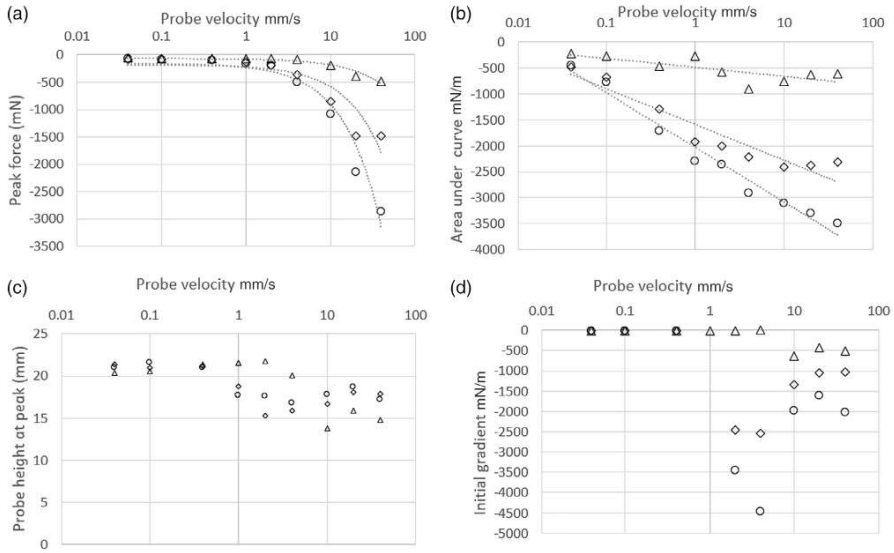

FIGURE 5 Influence of probe withdrawal velocity on zero perimeter virtual probe for (○) black treacle, (◇) golden syrup, and (△) honey. (a) Peak force (mN—linear trend lines are fitted), (b) area under the curve (mN/mm^2—log trend lines are fitted), (c) height of the probe at the peak (mm), and (d) initial gradient (mN/m)

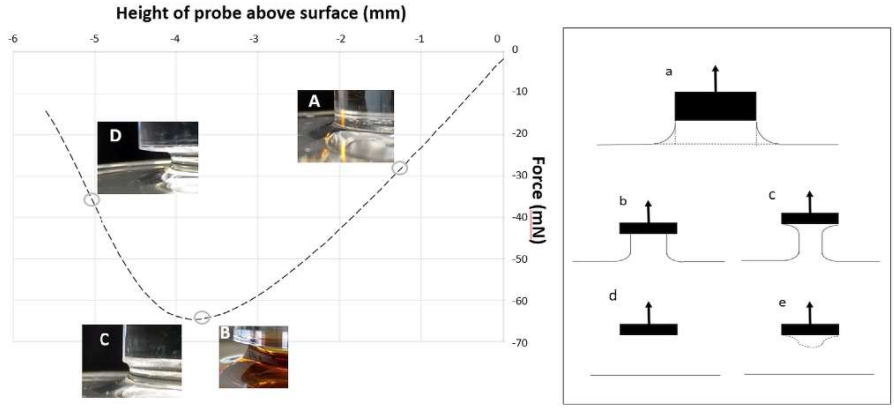

FIGURE 6 Force-distance curve for glucose syrup with inset images of probe contact surface and schematic view of the liquid in relation to the probe in tack tests. (See text for explanation of the codes)

peak is reached, the force-distance curve starts to flatten out, simultaneous with a narrowing of the column of liquid joining the probe to the body of liquid below. The curvature of the glucose syrup as the probe pulls away exhibits concave necking (inset Figure 6C)—holding firm to the probe perimeter and narrowing within the liquid itself (this is shown diagrammatically in Figure 6c). In contrast, the inset image

with a broken line (inset Figure 6B) is for Golden syrup at the peak and while the column of liquid thins, it does not hold fast to the perimeter of the probe but forms a narrowing cylinder attached to the base of the probe (shown schematically in Figure 6b). After the peak is past, the force continues to decline as the liquid progressively thins (e.g., inset Figure 6D). The schematic images (Figure 6d,e) show the amount of liquid adhering to the probe once detachment has occurred. Figure 6d results from the sticky behavior of Figure 6b and depicts adhesive failure (Kilcast & Roberts, 1998), while Figure 6e follows on from Figure 6c and is referred to as cohesive failure.

The inset image, Figure 6A (and the corresponding schematic 6a) shows the curvature of the liquid observed during these early stages of probe withdrawal. During the linear initial gradient of the force-distance curve, there is complete contact between the probe perimeter and the liquid, however, once the curve deviates from linearity there is a corresponding thinning of the liquid strand, either as Figure 6b where the liquid column detaches from the probe as it thins or Figure 6c where the strand thins but remains attached to the perimeter. The significance of these results is that even with our zero perimeter virtual probe the peak height and area below the curve do not correspond to the geometry of the end of the probe. In fact, of the parameters which we measured, the only one which is consistent with the geometry is the initial gradient.

In trying to understand what is going on in the tack test with liquid food syrups we considered the analogy of a "gas jar." Gas jars have rigid walls; they can be filled with water and inverted into a bath of water such that the open end is immersed at the bottom of the bath. If we set the gas jar so that the sealed end is in the same plane as the level of the water outside the jar, then we have (albeit with rigid sides) the geometry of a tack test—the sealed end of the jar is in the same plane as the probe would be in a tack test. By raising the gas jar (while keeping its open end submerged), we are effectively doing what occurs in the tack test, that is the rigid sealed end (or probe) is raised from the level of the liquid. The level of the water in the gas jar does not fall because an equilibrium develops between a small partial vacuum at the top of the jar and the weight of water below. Now consider what would happen if instead of the side walls being rigid they are actually flexible, of course now the weight of the water will cause it to flow but in doing so the walls will contract resulting in a curvature to the sides. Instead of glass walls, we now have the surface tension containing the liquid strand and as the probe moves upwards the strand is stretched while it undergoes simultaneous contraction. At relatively short distances, liquid from the bulk may actually flow into our rising strand, but with increased separation, the strand thins and we get the situation illustrated in Figure 6B/b, C/c where strand thinning is underway. The force exerted on the texture analyzer starts to decline as the strand thins and the curve begins to level off.

As has been stated, Brennan and Mohamed (1984) correlated sensory stickiness with liquid viscosity and surface tension. We hypothesis that the surface tension is pulling the strand tight as the liquid syrup drains out and that it is the surface tension that creates the characteristic narrowing strand of separating sticky liquids. However the rate of drainage from the strand depends on the viscosity and while the texture analyzer is pulling up, if the speed of the upward moving probe is greater than the rate of liquid drainage, a tensile force will be registered by the texture analyzer. Of course at high texture analyzer speeds the resulting tensile force is greater. This corroborates with Figure 3 where high speeds have higher peaks, greater areas under the curve, and steep initial gradients. The idea that the texture analyzer speed might be greater than the ability of the liquid to flow away would give rise to a stress increasing within the strand with the effect of the strand breaking at its thinnest point which in Figure 6c is in the midst of the strand and results in cohesive failure. In contrast when the speed of the texture analyzer allows enough time for the liquid to flow then we obtain the situation depicted in Figure 6b where the liquid flows from the probe resulting in a thinning strand and with reduced contact area attaching to the probe. As it flows the strand thins and results in adhesive failure. In effect, it is the speed of the instrument which is causing either cohesive or adhesive failure (Kilcast & Roberts, 1998). This postulate supports the work of Noren et al. (2019) who observed that the cohesive/adhesive failure behavior actually depends on the speed with which the test is undertaken.

To test the idea that the liquid is flowing from the tip of the probe we undertook a step change, fixed distance test. To achieve this we initially ran a tack test with just the single-headed probe. When the test was complete, we noted the height of the probe at its peak. We then repeated the test halting the texture analyzer about two thirds up the initial linear gradient. After halting the instrument we continued to monitor the force exerted on the texture analyzer with time.

Anecdotal handling of syrups (such as when separating our fingers with the syrup between), might give us the impression that they have elastic properties. The stretching and thinning of the strand and the stickiness of the liquid suggest inherent elasticity. Yet the fixed distance step change test (Figure 7) shows that when the texture analyzer is halted an instantaneous fall in the resisting force occurs, suggesting the material flows off the probe.

These investigations lead us to the conclusion that sticky liquid syrups are viscous materials and that the stickiness is due to the time required for the liquid to flow. When we stretch the liquid between our fingers or in a tack test, we are applying a force which is often

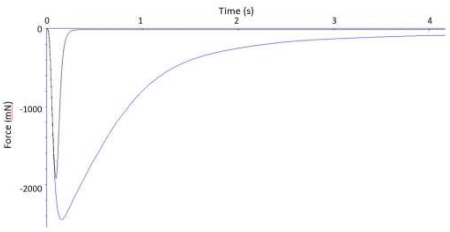

FIGURE 7 Step change test for fixed distance force with time—blue curve is a tack test and black curve is the step change fixed distance test

greater than the rate at which the liquid can flow, and consequently a resisting force is registered. We have in effect an unsteady state process that has not reached its equilibrium. By undertaking a tack test we are imposing a mechanical separation at a rate faster than the liquid is able to dissipate the energy. What we register on the texture analyzer is a snapshot of resistance, yet if we were to operate at a slower speed, the snapshot would be different as the liquid would have more time to flow and therefore the resistance would be smaller. Clearly in the case of liquid syrups, what we are measuring is an artifact, inasmuch as an artifact is defined as "something observed in a scientific investigation or experiment that is not naturally present but occurs as a result of the preparative or investigative procedure" (Lexico, 2021). This is not to say that the results are unreproducible, for they are highly repeatable with small standard deviations for the characteristics which can be extracted from the force-distance curve (e.g., peak height, etc.).

We must remind ourselves that in this work we are dealing with liquids albeit sometimes rather viscous ones. Earlier we comment on the almost vertical force-distance curves of the high-speed tests, the idea that the material cannot dissipate the stress through flow or relaxation is consistent with stresses building within the material until that material cannot support further stretching and undergoes catastrophic failure. If the separation of the probe from the surface of the liquid is faster than allows the liquid to flow, we get cohesive failure. In this study, we are dealing with viscous syrups. Many foods are glassy materials, which are often considered to be supercooled liquids. Such materials have immensely high viscosities which would be difficult to flow in the time frame of the test protocol outlined here. Thus we would expect glassy materials to undergo cohesive failure. Some other foods are viscoelastic (e.g., doughs) and in tack tests, we might expect the failure to depend on the predominance of viscous and elastic elements present. In the case of viscoelastic materials both the ease of flow, stress relaxation, and the elastic limits of the material will dictate whether the texture analyzer imposed force can be adequately dissipated in the time frame of the test or whether we will end with cohesive failure.

In our daily life, we experience the phenomenon of stickiness in the liquids we interact with. Yet here we discredit the use of tack tests to instrumentally measure the stickiness of liquids. If researchers persist in using tack tests then perhaps the approach they should take should be to employ separation speeds akin to those applied in the manual manipulation of materials with our fingers and/or jaw motion. Sharma and Sherman (1973) used a similar approach in recommending shear rates with which to evaluate the viscosity of liquids if they are to match our human experience.

4 | CONCLUSIONS

In examining parameters that contribute to tack tests of liquid foods we have seen that the measured stickiness depends on the geometry of the probe, the speed of separation of the instrument probe from the liquid surface. We have been able to eliminate factors relating to the liquid meniscus such as curvature, contact angle, and probe construction material by converting data from the Kazemeini and Rosenthal probe series to a zero perimeter virtual probe. In addition to the widely used "peak height" and "area below the curve," we have introduced the "initial gradient" of the force-time curve and the "distance to the peak" as indices of stickiness. Photographic evidence discredits the use of "peak height" and "area below the curve" as such values cannot be considered on a per unit area basis. We corroborate other researchers who show that the cohesive and adhesive failure depend on the speed of the texture analyzer. We show that when viscous liquids are subjected to a tack test the results depend entirely on the way in which the test is undertaken, providing a snapshot of the liquids' response to a given speed and geometry.

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AUTHOR CONTRIBUTIONS

Mostafa Kazemeini: Data curation; investigation; writing-review & editing. **Andrew Rosenthal:** Conceptualization; investigation; methodology; supervision; visualization; writing-review & editing.

ETHICAL STATEMENTS

Conflict of Interest: The authors declare no conflict of interests.
Ethical Review: This study does not involve any human or animal testing.

DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are freely available on request from the corresponding author.

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