Developing Latent Fingermarks on the Surface of Unfired Ammunition by using Time-of-Flight Secondary Ion Mass Spectrometry

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Abstract

It is a statement widely agreed upon that the friction ridge detail on a finger is unique to each digit and to find two that are exactly alike, between any two humans, is highly improbable. For this reason, fingermarks are considered of great importance to the judicial system as a match between two marks can confirm a shared identity. Due to the potential for identification, attempts to improve development processes of fingermarks is an area of interest for many researchers. One area that has been scarcely investigated is the development of latent fingermarks on ammunition by using time of flight secondary ion mass spectrometry.

This study aimed to adapt processes that have been used in other research to generate a complete image of a fingermark on the surface of a round. The fulfilment of this aim was facilitated by conversations with East Midlands Special Operations Unit (EMSOU) and their desire to develop images of fingermarks on evidence in the form of unfired 12mm Mark Webley rounds. With the success of this study, the evidence from this cold case can now be examined. Furthermore, where other techniques might fail, ToF SIMS has been shown as a technique that can be used to yield a clear fingermark.

Full scans of the round from ToF SIMS yielded high-quality images with several examples of minutiae clearly visible over the fingermark. These images were compared with those generated through ink or conventional fuming and staining techniques. In each case, the ToF SIMS image was superior, showing a greater level of detail.

The main sample, a 12 mm Mark Webley round, of this experiment was analysed on three separate occasions. The first occasion was one day after the mark was left on the surface, the second, seven days, and the third, seven months. ToF SIMS recorded no loss of detail over the time course of this experiment with all three images from each analysis period looking virtually identical.

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COVID-19

During the course of this research, October 2019 – March 2021, a pathogen designated COVID-19 plagued the UK causing many delays and issues with research all over the country. This project was one such case where the interruptions caused by this pandemic affected the nature of this investigation. The original plan was to conduct 5 tests over the course of 6 months. The 5 time stamps would have been 1 day, 1 week, 1 month, 3 months, and 6 months. As the laboratories at Nottingham University were not open from March 2020 – July 2020, the 1 month, 3 month and 6 month tests could not be completed. However the 6 month scan was replaced by a 7 month scan. Not only did the pandemic cause issues with scheduling tests on the ToF SIMS instrument in pharmacy, it affected the support I was able to received as well as lack of access to tools and resources with the physics labs. Please consider these circumstances when reading this report.

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1. Introduction

1.1 Motivation

Since 2014, crimes involving firearms have steadily risen by 28.5% with March 2014 recording 4856 cases and September 2020 recording 6242 cases [1]. As part of the process of bringing criminals to justice, the visualisation of fingermarks on surfaces is used to identify and convict the perpetrators of these crimes. It is vitally important that the methods used to yield images of these fingermarks are accurate and reliable, as this kind of evidence has become a staple in the justice system and has been used since the 1860s [2].

Fingermarks are found in three distinct forms: plastic, patent and latent. Plastic marks have a 3-dimensional element to them and might be made by pressing a finger into a substance that can be deformed i.e. wax. Patent marks are visible and are found at crime scenes formed in blood etc. Latent prints are invisible to the naked eye and require the use of chemicals or physical processes to reveal the mark. Patent and latent fingermarks are deposited onto surfaces when a person touches it with their finger and the substances leave a mark, with the materials deposited replicating the ridge detail on a finger [3].

In an attempt to improve the reliability and accuracy of fingermark analysis and comparison, new methodologies have been developed in order to enhance these latent fingermarks. The clearer a fingermark is, the more likely it is to be matched with another fingermark or fingerprint taken from another source, person of interest, or scene. Furthermore, the importance of fingermark analysis cannot be understated, as mark identification remains one of the most important tools for a forensic scientist, due to the persistent and unique nature of friction ridge detail [6, 7].

Certain surfaces and materials are more likely to host a latent fingermark that can be more easily visualised. This, in part, is due to the wettability and porosity of these materials. When the secretions from sweat glands encounter some surfaces the friction ridge detail is not preserved. This can make fingermark analysis using conventional methods very difficult [8]. There is no one method that are adequate for all surfaces. Metallic substrates are notoriously difficult to work with when trying to develop a fingermark [54]. Scientific police researchers agree that powders and chemicals are currently the best options when trying to visualise a latent fingermark on a metallic substrate, but these surfaces are usually hard to develop and the results are often poor [8, 54]. As no optimal technique is available for use on firearms, knife blades and ammunition, researchers a still searching for a reliable method for these metallic surfaces. This difficulty is due to numerous factors. Houck suggests that the lack of success with cartridge cases may be due to their small size, the method of handling, surface treatment, and mechanical abrasion [5]. Other factors may be conditions of the crime scene, the age of the fingermark, and the nature of the interaction between the residue and the substrate [9]. Metals and alloys are one class of problematic materials as the aqueous part of the mark has a short life span. It is this part of the mark that conventional techniques rely on to yield a visible mark.

Metals and their alloys are a key material used to make weapons that are used in violent crimes e.g. knives, guns, and ammunition. One such technique that has been used to improve the quality of a mark present on problematic surfaces is Time of Flight Secondary Ion Mass Spectrometry (ToF SIMS). Thandauthapani et al. used ToF SIMS to great effect, yielding a superior result when compared to conventional superglue fuming and subsequent staining. This method has been used to generate images of fingermarks on the flat of a blade [9]. Other more complex shapes have not yet been tested due to the current design of ToF SIMS machines. The space inside the main chamber is limited and the area that can be scanned is only able to move in a linear fashion and cannot rotate. This means that latent marks on curved surfaces, such as handles, grips and ammunition, cannot be visualised.

In this thesis, ToF SIMS is used in in a similar way but is adapted with the use of a bespoke designed piece of equipment. This means that the entire surface of an unfired round was scanned to reveal any latent fingermarks that may be present. The success of this adaptation will aid forensic scientists by unlocking the ability to find new evidence on curved surfaces.

1.2 Background

In 1902, a landmark in forensic science was established when an individual was convicted based on fingermark evidence. The case was that of a burglary involving Harry Jackson who stole several billiard balls from a property in London. After eliminating other persons that could have left the fingermark at the scene of the crime, the finding of a match was successful. The jury were persuaded that there was reliability to this evidence and Jackson was sentenced to 7 years imprisonment as a direct result [10]. This conviction set a precedent in the UK. Since this case, the use of fingermark evidence has become such a staple in the justice system that the total number of cases in the UK relying on fingermark evidence are not recorded [11]. Further proof that the UK still is using fingerprints to identify suspects and link identities to crimes, are the statistics from the same publication stating over 26 million identified prints and 2.2 million yet to be identified prints are held on the IDENT1 system (the central biometric database for the UK) [11].

Friction ridge detail is found on the tips of fingers, palms of the hands, and the base of the feet and toes [25]. It forms during the development of the foetus in the womb at roughly 4 months into pregnancy [4]. From the moment these ridges develop, they do not change over the course of a lifetime, save the addition of some wrinkles due to ageing. They even regrow with the exact same pattern if the skin is removed [4]. The exception to the unchanging nature of friction ridge detail is due to external factors such as scarring and psoriasis. These can remove or obscure ridge detail resulting in a new, unique print [26].

As the courts began to rely on fingerprint evidence as a means to identify or rule out suspects, a need for more reliable techniques arose. One of the earliest examples of the improvement of finger mark identification was by a French chemist Paul-Jean Coulier by using iodine stains and magnifying glasses [12]. This technique was applied by using the iodine to stain the mark and make the pattern more visible for inspection. Using magnifying glasses in tandem with this chemical process allowed for better analysis of a finger mark.

Today, more sophisticated techniques are used to visualise a fingermark and a wealth of other reagents and powders are used depending on the surface or the composition of the fingermark [3]. 3 main processes are used, preparation, optical, and chemical and physical. The home office describes these processes as “category a”, as other processes, categories b through f, are not as commonly used and have varying results. These three are the most reliable and will be used in different situations according to this literature [60]. The current conventional and popular technique using cyanoacrylate (superglue) fuming is used in this paper as well as using contrasting dyes such as basic yellow 40 (BY40) [13, 14]. Furthermore, as part of the effort to improve the way fingermarks are developed, the Henry system was introduced in order to better classify fingerprints. This system was introduced in 1901 by Sir Edward R. Henry and it is still in use today [15], the explanation of this system is provided in chapter 2. The way we visualise, analyse, and classify fingermarks and fingerprints is being improved each day to aid the work of the police, with the current Automated Fingerprint Identification system (AFIS) as a cornerstone [16].

Bumbrah et al. suggests that a new trend is emerging, with the visualisation of latent fingermarks achieved by using sophisticated instrumentation [17]. One such technique is the use of ToF SIMS. This analytical method enables the detection of the constituents of a latent fingermark so the mark can be visualised. The tests conducted using ToF SIMS, yielded a high-quality image of a latent fingermark where conventional methods failed to yield a satisfactory result [18]. This method of visualisation, while being superior, also allows for further testing by other methods as the mark is unchanged by this technique.

The UK currently uses a process of fingerprint comparison called "ACE-V". This acronym stands for analysis, comparison, evaluation, and verification [49]. It is conducted by highly trained fingerprint experts that are responsible for the comparison of fingermarks found at a crime scene or later developed with fingerprints taken from persons of interest.

The first step, analysis, is the process of locating and annotating minutiae that can been seen when looking at a fingermark. It may be at this stage that the fingermark is deemed not usable and the process ends here. If enough minutiae are present and visible, the comparison stage can begin. Detail is split into three different levels as seen in figure 1.1. The first level of detail is overall ridge flow, this level of detail is fairly easy to see even on a low-quality fingermark. 6 examples are given of this level in figure 1.1. The second level of detail is that of minutiae found in the ridge detail of the skin i.e bifurcations and endings of friction ridges, see figure 1.1 for more examples. The third level represents the greatest level of detail in the friction ridge characteristics such as the shape of the ridges and the placement and shape of the sweat pores also seen in figure 1.1[4].

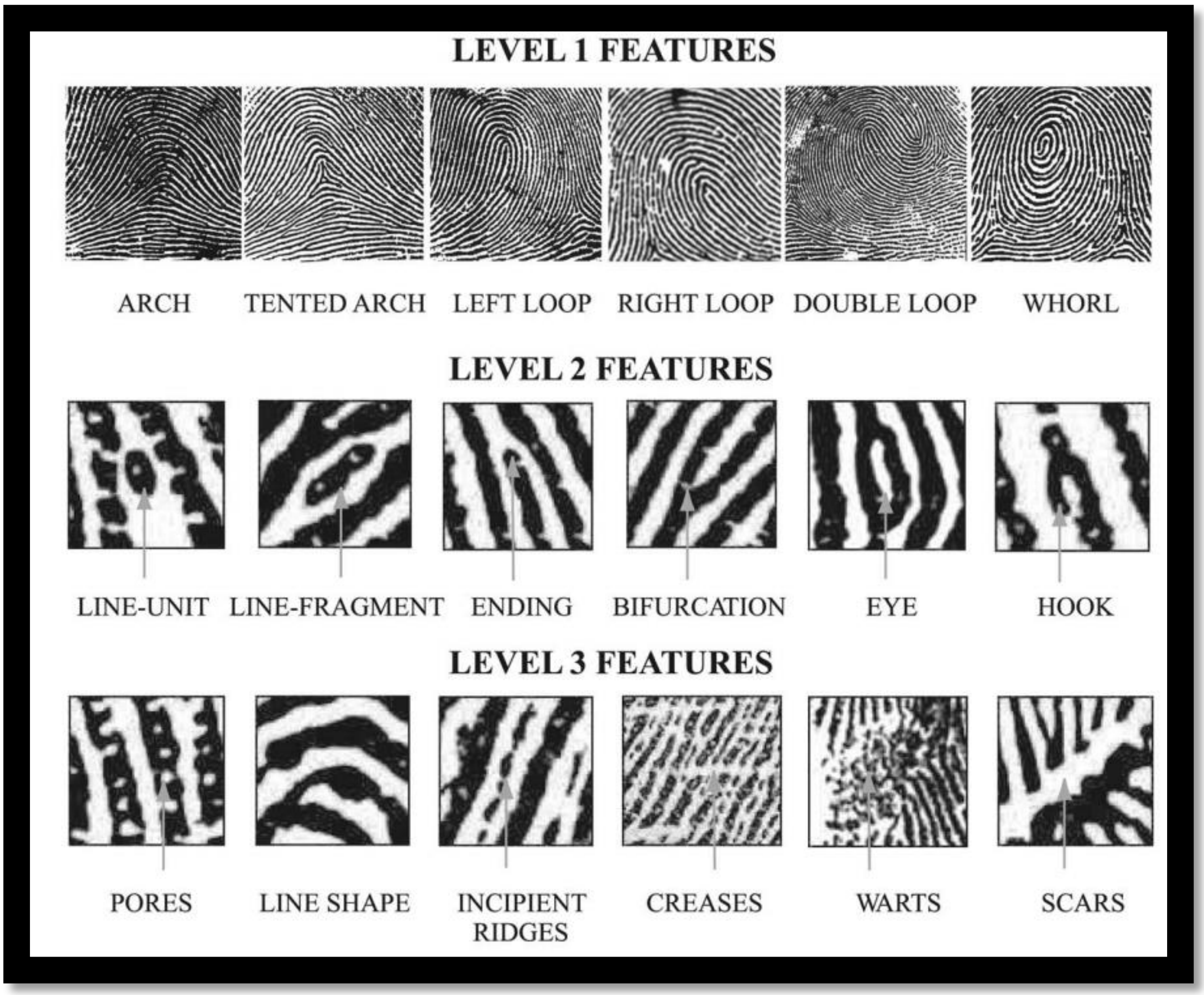

Figure 1.1: This figure shows a list of features found at each level of quality with level 1 being common and of the least significance and level 3 being of the highest quality and greatest significance. Image is reprinted from *Pores and ridges: High-resolution fingerprint matching using level 3 features* [50]

1.3 Time of Flight Secondary Ion Mass Spectrometry

As the subject washed their hands and avoided touching other parts of their body with their hands prior to touching the round, it is reasonable to suggest that the only contents on the substrate would be the secretions from eccrine sweat glands. As previously stated, the majority of eccrine sweat is water, which has a short lifespan on a non-porous surface. This means that by the time the first scan is initiated (24 hours after deposition of the mark), the only materials left on the surface will be the small amounts of organic and inorganic compounds found in eccrine sweat. This is another reason why ToF SIMS has been successful when developing latent prints on metallic surfaces. The sensitivity of this method is extremely high as it can detect a monolayer of material [55].

To be able to visualise a fingermark on the surface of a casing, the detection and differentiation of the residue and contaminants is needed. This is the basic premise behind developing fingermarks on any surface. Aiming for a higher quality image than what would be developed when using conventional methods, ToF SIMS was used to visualise the fingermarks on the surface of the 12 mm replica Mark Welbey round. ToF SIMS measurements were carried out using an ION-TOF ToF SIMS 4 instrument.

ToF SIMS uses mass spectrometry to great effect by analysing the internal and external products of a finger. The chemical image is produced by moving an energetic ion beam across the surface of the sample while measuring the ions that are ejected. A 25keV Bi_3^+ ion beam was the primary beam used in this experiment. These bismuth ions are accelerated by the use of electrodes set at the extraction voltage desired [56]. The beam of primary ions is focused using a magnetic field and the location of impact is decided by moving the sample around on a motorised stage. To ensure that the primary and secondary beams do not collide with air molecules, the chamber is airtight, and an ultra-high vacuum is created using a series of pumps. The secondary ions that are ejected from the sample after the primary beam is incident are collected by an extractor and then accelerated towards a detector. The secondary ions are accelerated with the same energy meaning the time taken for the ions to reach the detector will be different depending on the mass of the ion [57]. These secondary ions are separated according to their mass-to-charge ratio (m/z) and the relative abundance of each ion is recorded.

The kinetic energy of the ion is the energy set by the accelerator. Therefore, the mass of the ion can be calculated using equation 1.

$$Kinetic\ Energy = \frac{1}{2}mv^2 \tag{1}$$

Where *m = mass* and *v = velocity*. If the time taken to traverse a known length is recorded for a particle travelling at a constant velocity. The velocity can be easily calculated using equation 2.

$$Velocity = \frac{s}{t} \tag{2}$$

Where *s = distance travelled* and *t = time taken*.

Once the instrument is calibrated using known ion peaks, m/z are assigned easily to the secondary ions that are produced [58].

ToF SIMS detects all ions present on the surface analysed and the intensity of the peak will depend on the amount of substance detected. For areas where the ridges of the fingerprint have been in contact with the surface of the round, it is expected that high intensity peaks will be present for ions found in eccrine secretions. As the areas where the ridges have been in contact will have a high count of these ions and the areas with no contact will have low or a zero count of these ions, a fingermark will be able to be seen.

ToF SIMS needs a flat sample if it is to be able to analyse the surface effectively. Substrates with a non-flat topology are problematic as the angle that the primary beam is incident on the surface will vary. This is a major issue for a sample in the shape of a cylinder as is the case for testing ammunition for latent fingermarks. Fortunately, there is a small window where the primary ion beam will still be in focus. This depth of field is a very small distance and for the model of ToF SIMS that has been used, this is 0.002 mm. Using simple circle formulas and trigonometry, the width across a cylinder of radius 6.05 mm that will remain in focus if the depth of field is 0.002 mm is 0.3 mm.

Diagram A in figure 1.2 shows a simplified image, detailing the problem that occurs when ToF SIMS is used on a curved surface. Due to the difference in height of the area to be scanned, some portions will not be in focus as the secondary ions will not be collected by the detector. Diagram B in figure 1.2 is an example of this phenomenon as the intensity lessens towards the edge of the scan due to these edges being out of focus.

Each strip in diagram B in figure 1.2 measures 0.1 mm across with the central 3 strips being the clearest. Strips 1 and 5 are affected by the curvature of the casing and the depth of field of the ToF SIMS detector is unable to keep the consistency of measurement across the entire 0.5mm wide scan. Diagram B in figure 1.2 shows one of the preliminary tests that confirmed that the maximum width that can be viewed across a cylinder of this diameter is 0.3 mm.

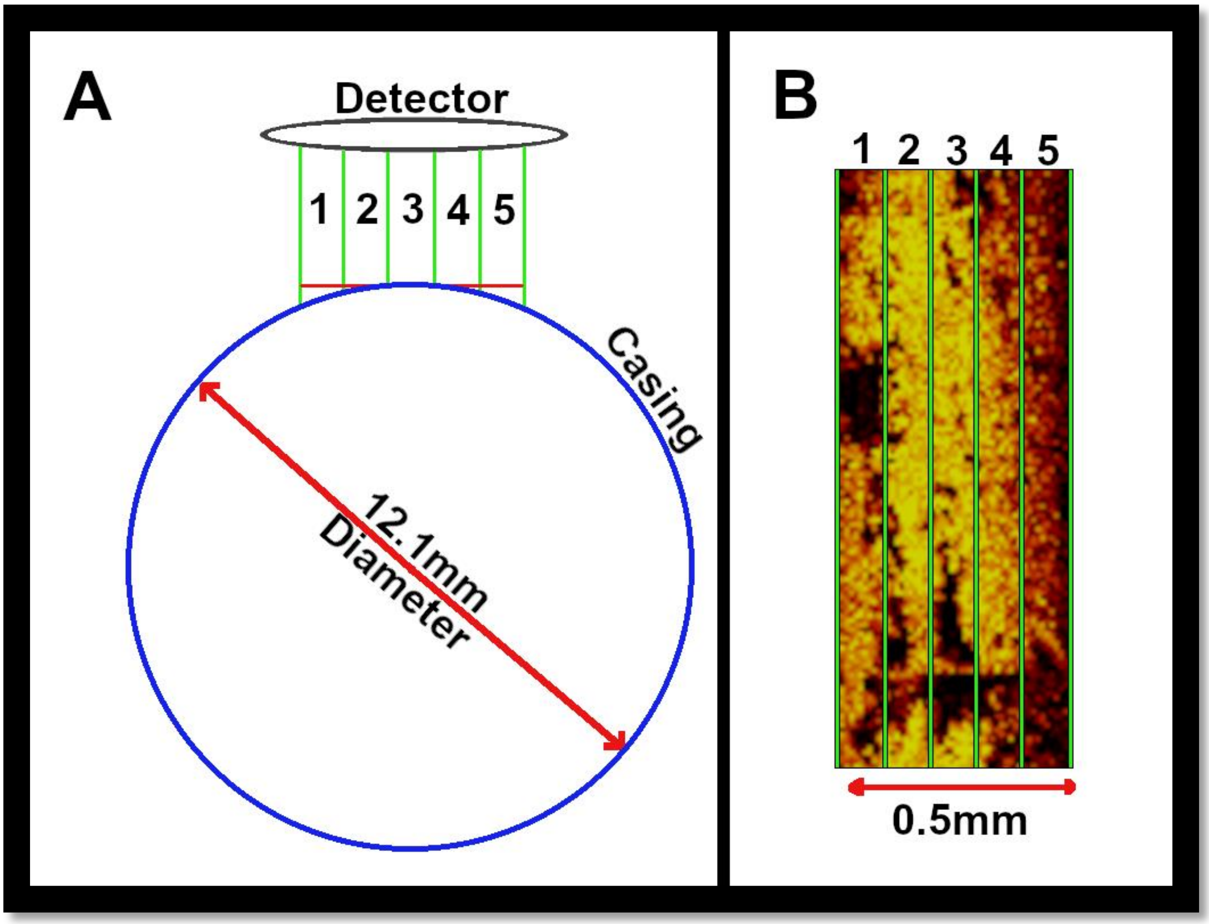

Figure 1.2: Diagram A shows a simplified image of how the surface of a cylinder changes the distance and angle to the detector across the width. Diagram B shows confirmation that the depth of field effects the detail of a scan measuring 0.5 mm wide with the central 0.3 mm region clearly in focus.

2. History and visualisation methods

2.1 Fingermark formation and classification

A fingerprint is the impression left on a surface when the friction ridges of a finger leave the same detail on a surface. It is known as a fingermark, sometimes abbreviated to “mark”, if this pattern is generated in non-controlled conditions [19]. The current favoured method for recording fingerprints in the UK is using IDENT1 which replaced the National Automated Fingerprint Identification System [20]. This transfers the pattern of the detail quickly, efficiently, and with minimal cost all while achieving a high-quality image of the ridge detail of each digit and the palm of both hands. Where this method cannot be used, prints are taken manually using ink, a pad, and a piece of card or paper [21]. When prints and marks match, it is highly probable that the two originated from the same source with the probability that two different people have the exact same print is roughly 1 in 64 billion [24].

Most latent marks are made through contact with surfaces when only sweat or gland secretions are present on the finger [4]. The eccrine, sebaceous, and apocrine glands excrete their namesake fluids with only eccrine glands found on the fingers. This is not to say that sebaceous or apocrine secretions would not be found on a finger as people tend to touch areas of their body where these glands are located [5]. Of the three distinct ways friction ridge detail is left on a surface, latent fingermarks are the hardest to visualise as they are usually invisible to the naked eye. This, in part, is due to their composition. Latent fingermarks occur when the sweat from the pores of the body, coats the friction ridge details and transfer to the substrate when in contact. It is this sweat that makes up the latent fingermark. There are 3 primary types of sweat glands: eccrine, sebaceous, and apocrine [2]. Eccrine sweat glands can be found all over the body and the composition of the sweat that comes from these glands is mostly water with only 1% of the substances found as organic and inorganic compounds [27]. Sebaceous sweat glands are found in areas that are surrounded by hair follicles and the sweat is composed of organic components (mostly triglycerides and fatty acids) and apocrine sweat glands are found in the armpit and genital regions [28]. It might be thought that latent fingermarks are solely made of eccrine sweat, and it is unlikely that sebaceous and apocrine sweat will be present due to the location of each gland. However, in practice, eccrine, sebaceous, and apocrine sweat are all found as part of latent fingermarks due to the fingertips picking up these sweat types from contact with the body [2]. Furthermore, other materials may be present from contact with cosmetics and food. Even diet and supplements can also affect the composition of secretions from the sweat glands of the body [29]. A table with a list of components of each sweat type in table 2.1. Each of these kinds of sweat responds differently to the substrate it encounters due to numerous factors. These include, but are not limited to, contaminants, ambient conditions, and the properties of the material.

Type of gland	Inorganic components	Organic components
Eccrine	Chlorides Ammonia Phosphate Sulphate Metal ions (Sodium, Magnesium, Calcium, Iron, Copper, Manganese) Halide ions(Iodide, Bromide, Flouride)	Amino acids Urea Lactic acids Sugars Creatinine Choline Uric acid
Sebaceous		Fatty acids Glycerides Sterol Wax esters Hydrocarbons Squalene Alcohols
Apocrine	Iron	Proteins Cholesterol Carbohydrates

Table 2.1: The main inorganic and organic components of the three different kinds of sweat secreted by the human body. Reproduced from *The role of wetting effects on the development of latent fingermarks* [30]

While unique, similarities can be found in the friction ridge patterns between two people. As more prints were being recorded, a classification system was introduced by Sir Edward Henry at the beginning of the 20th century. This allowed a more efficient sorting method so that a match could be easier to find within a pool of thousands, at the time, of unique prints. Referring to Purkinje's 9 types of fingerprints shown in fig. 2.1, The classification method assigned specific non-zero values to each of the 10 digits if a whorl pattern was present and a zero value if the pattern was not a whorl. The combinations of these values allowed for 1024 groupings which meant the ever-growing base of prints could be more easily organised [4].

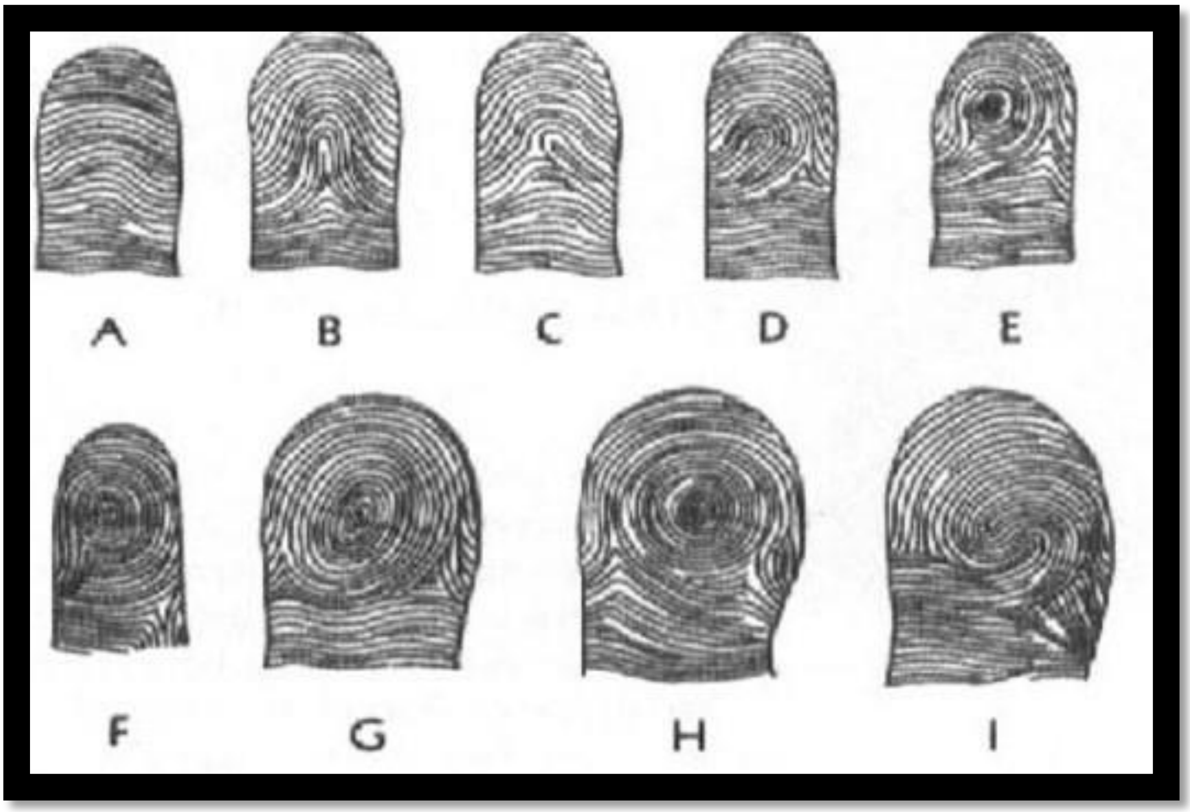

Figure 2.1: This figure shows the 9 types of fingerprints classified by Purkinje. A: Transverse curves, B: Central longitudinal stria, C: Oblique stria, D: Oblique sinus, E: Almond, F: Spiral, G: Ellipse or elliptical whorl, H: Circle or circular whorl, and I: Double whorl. Image is reprinted from *The Fingerprint Sourcebook* [4]

In the present day, the home office is tasked with the sorting and identification of millions of unique prints and so a more sophisticated method is needed. Whilst being a method for scanning and comparing fingerprints, IDENT1 is also a database that houses 26 million unique prints for over 8 million individuals [11]. Most modern classification systems are a derivative of the Henry system with a basis in Purkinje's 9 types of print [31] and IDENT1 uses this to great effect when attempting to match two patterns.

2.2 Conventional methods used to visualise latent fingermarks

2.2.1 Optical Methods

Depending on the substrate and the composition of the mark found, certain visualisation methods will have more success than others. One of the first things that is considered before choosing one of the many modern visualisation techniques is whether the substrate is porous or non-porous [32]. Kent goes on to designate optical and physical development processes that should be used depending on the composition of the latent print as well as the current state the surface is in i.e wet or dry. Where possible, Kent encourages the drying of the sample at a maximum of 30°C and then the decision tree he shows should be followed in order to achieve the best image of the latent mark possible [32].

Optical methods are often considered first when detecting or enhancing fingermarks. They are also involved in several other processes with photography usually being the last step. They are non-destructive as there is no contact with the mark and depending on the wavelength of the light source, they can enhance the visualisation of the mark greatly if certain chemicals are used prior [33].

Using white light is the simplest optical technique and it is the first point of call in any examination of a latent fingermark. The scattering of the light on the surface where a mark is present may lead to a pattern that is visible by a camera and therefore captured for analysis. It can be that altering the angle at which the light is incident on the surface may aid the exposition of a mark especially in cases where the substrate is textured or reflective [30].

Fluorescence is often considered when examining fingermarks. Under visible light, some components of sweat, tyrosine, phenylalanine, and tryptophan, are fluorescent [32]. Other reasons that an untreated mark may fluoresce are that it is contaminated with other materials not excreted by sweat glands such as cosmetics or industrial chemicals [32]. Otherwise, fluorescent dyes, reagents, and powders are used to stain, react, or adhere respectively with the mark so fingermarks that are invisible under normal light conditions with be revealed. Examples of the dyes that are commonly used are basic violet 3 (BV3), basic yellow 40 (BY40), basic red 14 (BR14), and acid yellow 7 (AY7) [4]. Examples of reagents are crystal violet (CV), sudan black (SB), and 1,8-diazafluoren-9-one (DFO) [43]. Examples of fluorescent powders are those that contain fluorescein or rhodamine B [34].

Ultraviolet (UV), Infrared (IR) and near infrared (NIR) light are sometimes used in place of visible light in order to successfully visualise a fingermark. By using cameras or filters that a sensitive to UV, IR or NIR, more fingermarks can be visualised as some materials are fluorescent under these bandwidths of light. This can be either the mark itself, substances in the mark that fluoresce, or a chemical that is used to develop the fingermark, so it is visible in the same way [35, 36, 37].

2.2.2 Physical Methods

The physical methods can be broken down into further categories: powders, chemical reagents, lifts, and fuming. Unlike optical processes, these methods involve directly interacting with the surface where the latent fingermark is found. Disturbing, chemically altering, or adding to the composition of the mark. Once these processes have been used on a surface, it is no longer considered in its original state and the mark will have been enhanced when using the most appropriate technique according to Kent [32].

Dusting for prints with powders is a frequently used technique by crime scene investigators. It relies on the powder sticking to the components of the fingermark. A soft brush is loaded with a fine power which is then softly swept over an area in the hope of revealing a latent fingermark. If there is a mark present, still with water present from eccrine sweat or oily, sticky components from sebaceous and apocrine sweat, the power will leave the brush and stick to these materials. This action will reveal a mark on a surface that can then be analysed using an optical technique to finalise the visualisation of the fingermark [38]. The powder used may be resin, metal, or plastic based depending on the substrate type. Fluorescent powders are also an option for use on multi-coloured surfaces where optical methods using visible light may fail [39-42].

As previously mentioned, one of the more commonly used reagents for visualising fingermarks is DFO and this chemical is typically used to develop marks on porous surfaces [43]. This and other reagents are applied by either spraying or swabbing the surface where a fingermark is located. The other option is to fully submerge the substrate in the reagent. The reagents target the amino acids present in latent fingermarks. As the reagent comes into contact with these substances, a reaction occurs, presenting a change of colour where there is residue [35]. This colour change allows the use of optical methods to capture the image of the fingermark.

Fuming methods work in a similar way to reagents, but instead of targeting amino acids, the fumes form white deposits on the fingermark ridges [44]. The use of cyanoacrylate, better known as superglue, fuming is the most commonly used fuming method and has been used since the later part of the 20th century [45]. This method is initiated with the heating of a small amount of superglue, usually 2-5g depending on the size of the substrate being fumed, to over 100°C. Once the superglue begins to emit white fumes, the dish containing the superglue is enclosed with the substrate suspended above the dish [46]. In this air-tight container, the superglue fumes come into contact with the substrate and the white deposits form on the fingermark ridges, this is a sign of polymer growth. Over fuming is a potential issue with this method, as allowing too much polymer growth can lead to the ridges growing and detail of the fingermark being lost [47]. Depending on the colour and material of the substrate, dyes are then applied to the mark in order to add contrast so a better image can be seen. Several dyes are options for this process. BY40 and BR14 are two highly useful dyes commonly used in the UK [14, 48]. They are frequently chosen as they add contrast to the mark and have fluorescent properties so this process can be followed with fluorescence examination. These dyes are particularly effective at developing contrast on multicoloured or shiny backgrounds.

3. Experimental Techniques

3.1 Preparation of samples

One replica 12 mm Mark Webley round was used for all three tests over a 7-month period. The casing of the Mark Webley round was the main subject of the scan and so the metallic surface was cleaned to remove any materials that may have a similar composition as the secretions from a finger.

The round was firstly subjected to a sonicated bath of dilute acetic acid solution. The round was placed upright with the base of the case in contact with the bottom of the glass beaker. 200 mL of this solution was added to the beaker, so the round was fully covered for the sonication lasting 15 minutes. After these 15 minutes had elapsed the round was removed from the beaker and any remaining liquid was evaporated from the surface using compressed nitrogen gas. During this process, the round was held by tweezers gripping the bullet portion of the round. This process was then repeated in the same way with the weak acid replaced with methanol and then deionized water. After the final use of compressed nitrogen gas clear the round of any residue, the round was stored upright, with the base of the casing in contact with the glass, in a glass cylinder with the opening of the cylinder covered with aluminium foil. The round was always handled with tweezers by gripping the bullet portion of the round, and stored indoors within a container at ambient temperature, 10-20°C, and humidity, 70-90%, when not in use. These processes ensured that there would be no contact with the measured surface from any foreign objects or substances other than the fingers used to deposit the finger marks.

One subject was used for the depositions of fingerprints for this experiment. This subject was a male donor aged 43. This volunteer cleaned their hands by using ordinary hand soap and washing them for a minimum of two minutes using the NHS guide for washing hands [53]. This was to ensure that all residue, oils and substances from other sources such as other people as well as other secretions from other body parts, i.e. face and hair, were not present. A thorough clean of the hands and more importantly the fingers achieved this. After cleaning their hands thoroughly, their hands were left to dry naturally in air and secretions were left to build up naturally over the course of two hours before fingerprints were deposited on the rounds. All touching of face, hair, door handles, and other objects were avoided during these times so it is reasonable to suggest only the eccrine secretions would be present.

The thumb right hand of the subject was used to deposit a mark. After lining up their finger appropriately, the subject pressed down their finger on to the surface of the casing for a total of three seconds applying a force of 2 newtons for the duration. A stopwatch was used to time the duration and a top pan balance was used to record the force applied.

3.2 Settings for ToF SIMS

ToF SIMS builds the long strips by scanning smaller square patches along the full length of the require area. Even though only a width of 0.3 mm was needed, a width of 0.5 mm was scanned instead. This greatly reduced the time and cost for scanning the full surface of the round. If a patch size of 0.3 mmx0.3 mm was used then a total of 47 patches would be needed whereas, if a 0.5 mmx0.5 mm patch was used then only 28 would be needed. As the time taken for each patch does not greatly differ when altering its size, using a patch size of 0.5 mmx0.5 mm was more desirable. The excess width on either size of the scan was simply cropped during the processing of the images post scan.

As the diameter of the round was roughly 12.1 mm, and the width of usable data was 0.3mm, a total of 190 scans of thin strips were needed as there needed to be overlap between the strips. An overlap of 0.1 mm was chosen as there would be 10x1400 pixel area that a program could use to stitch the strips together. ToF SIMS scanned a strip, the round rotated through a small angle and the process repeated till the entire surface was analysed. To make sure there was an overlap of 0.1mm between scans, a movement of 0.2 mm across the surface of the round was need. This movement translated to a small rotation of 1.89° which would be provided by the high precision stepper motor that the round was connected to. The stitching program was written using Python in Microsoft visual studio. When the stitching program was run, a full fingermark was revealed for each of the sets of images depending on the ion data that was extracted.

The ToF SIMS was set to scan the 14 mmx0.5 mm area in 0.5 mmx0.5 mm patches with a patch resolution of 50x50 pixels. Each pixel for the scans were measured 0.01 mmx0.01 mm resolution. The distance between the peaks of the ridges on a fingerprint is typically measured to be in the region of 0.5 mm [59] so this resolution would enable the fingermark to be visualised. The scan was set to complete 20 frames per patch meaning with the generated images using these frames to determine the relative intensity of each pixel.

These were the settings used for the 1-day, 1-week and 7-month scan.

3.3 Development of the stage

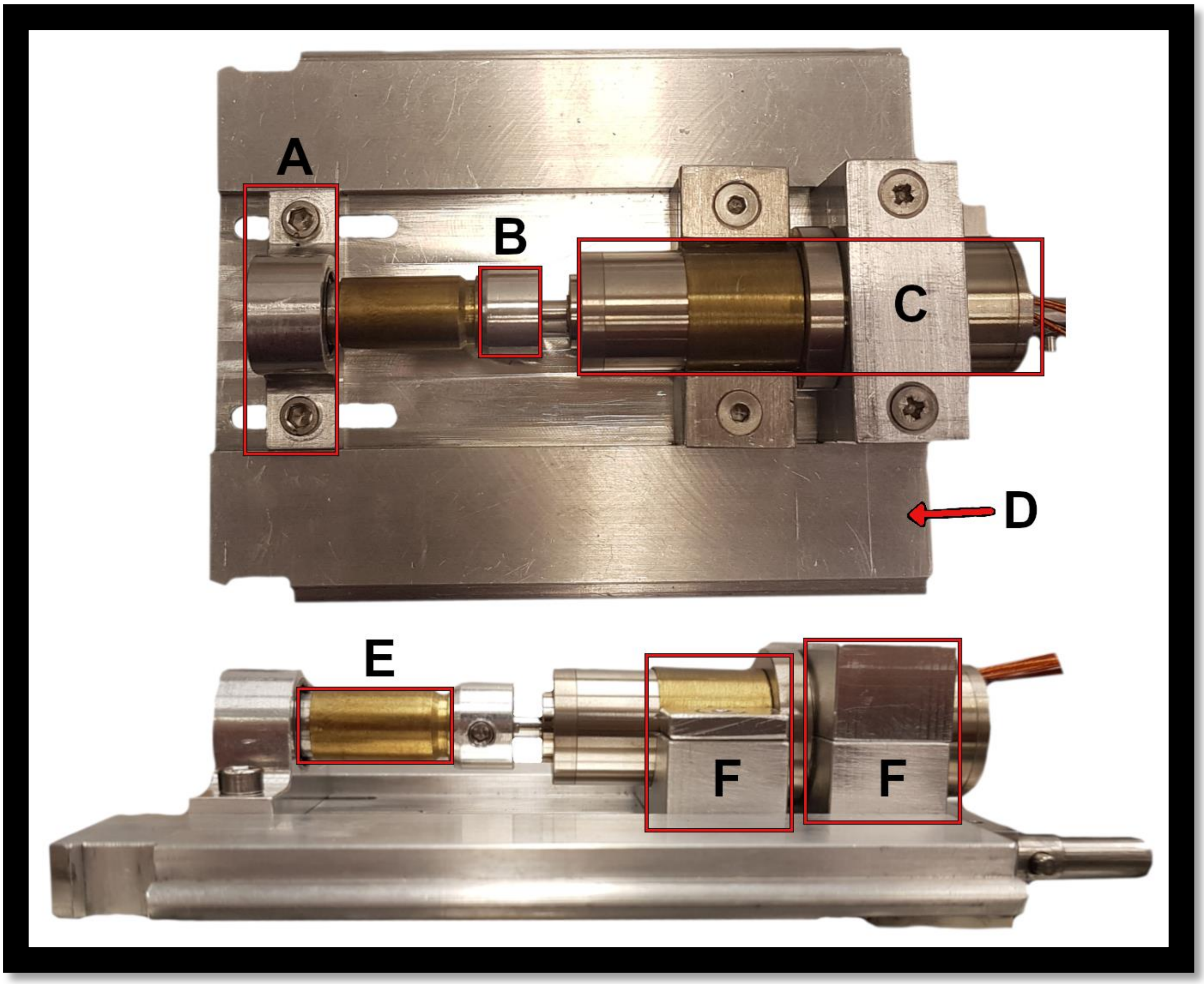

Figure 3.2: This image shows and highlights the various components that were part of the stage used in the ToF SIMS. A top-down and a side on profile have been included to better see each component to give context to the explanation within this chapter.

A Phytron VSS stepper motor with a gear box, C in figure 3.2, was used to ensure a high resolution when specifying an exact angle to rotate. The motor took 40,000 steps to achieve one full rotation. This was due to the stepper motor originally taking 200 steps to achieve a full rotation working in tandem with a 200:1 gearbox ratio. With this high resolution it meant that the smallest angle that could be specified to rotate could be as small as 0.009°. This may be needed for rounds of a smaller size as the ToF SIMS still needs a relatively flat surface to be able to successfully scan. The width needed to achieve this is relative to the diameter of the round, a smaller round means a thinner scan is needed and so a smaller angle of rotation is required. For the 12 mm Mark Webley round, the angle of rotation that is needed is 1.9° which is 210 steps with this motor and gearbox. The motor and gearbox were mounted using brackets at two positions on the stage, F in figure 3.2. This was done to ensure that the axis was parallel to the longest edge of the stage, D in figure 3.2.

The dimensions of the plate were designed so that it would fit the ToF SIMS and so the dimensions of a bottom mounted sample holder were used. The space inside the UHV chamber is limited as the aperture to enter the chamber is narrow and there needs to be adequate clearance between the ion beam emitters and the sample surface. The motor was wider than the Mark Webley round as it measures 14 mm in diameter. As the scan approaches the motor end of the stage then there is risk of collision between the motor and the emitter due to this difference in size. This will be counteracted by not scanning the full width of the casing and leaving a small length of unscanned metal at the base of the round so collisions can be entirely avoided. This unscanned length is determined by the angle of the emitter closest to the motor inside the chamber. A clearance between the emitter and motor is set at 2 mm which means that a length of 2 mm is not scanned at the base of the round.

To make sure the round is kept level, a bearing has been mounted at the bullet end of the round where the tip of the bullet will be fixed. When mounting the round for scanning, a small amount of pressure will be applied so that the round is firmly fixed between the bearing collar and the motor. This will ensure that the round is always kept horizontal, so the round does not move out of focus. These axes have been tested and shown to be tolerant to within 0.005 mm when using a dummy, machined stainless steel round of the same dimensions as a Mark Webley round. Replica and commercial rounds are not as uniform as the ideal round that was created for the purpose of testing the tolerance of the sample stage that was created specifically for this experiment.

Using the Phytron MCC-2 motor controller, the motor could be programmed to turn at regular intervals and even wait for an input before starting a specified command. Unfortunately, there was not a simple and easy way to link the ToF SIMS and the motor controller together so the controller could take an input from the ToF SIMS. This meant that the motor controller would need to be programmed in a way that relied on specific timing.

A total of 209 scans were performed by the ToF SIMS. Only 190 would be needed to achieve scans of the whole surface of the round with the casing rotating 0.2 mm after each strip was analysed. An extra 19 (10%) scans were added to the procedure to ensure that the entire surface of the round was scanned. The casing was liable to slipping or catching when rotating, meaning the desired 1.9° rotation was not always achieved. The rotation would sometimes be a little smaller than desired or no rotation at all might occur. This was largely in part due to the mechanism used to mount the round. Pressure was created at the bearing end of the stage, A in figure 3.2, pushing the base of the casing, E in figure 3.2, on to the plate connected to the driving axis, B in figure 3.2. It was originally thought that the friction between the plate and the base of the round would be sufficient to turn the sample reliably. In practice, the sample would not always rotate as intended. When more pressure was applied so the action of friction was greater, it created a similar issue where the sample would not rotate as the bearings were then under too much load. For this reason, the mechanism of mounting the round to the driving axis would need to be redesigned.

The software used to program the motor controller was the Phytron made Minilog-comm program. Included in the program were features to ensure the motor and controller were behaving correctly before the experiment began. These included a full rotation clockwise, anticlockwise and a reorientation back to the starting position originally set.

3.4 Application of conventional fingerprint enhancement techniques (FET)

Three casings were sent to EMSOU to be forensically analysed for fingerprints using conventional techniques. These 3 casings were cleaned using the method described in section 3.1 and then the same subject deposited after cleaning their hands in the previously described method. One of the casings has the print deposited 7 months before being sent, another, 1 week and the final, 1 day. The 1-day and 1-week samples were deposited 7 months after the first batch of samples that were tested using ToF SIMS. The 7-month-old casing was the same that was used for the ToF SIMS scans and was only sent to the forensics team after all scans had been completed as the superglue fuming techniques would contaminate the surface. The vast difference in time between the two 1-day-old and 1-week-old tests may have influenced results.

All 3 casings were treated in the same way with the same solvents, cycles and treatments applied. Foster and Freeman MVC 5000 superglue cabinets were used and were run on an automatic cycle; this includes a 15-minute humidity cycle (humidity reaches around 75-90%), a 20-minute glue cycle (the glue reaches around 120c on the hot plate before fuming) and then a 40-minute purge cycle using contained carbon filters. 4 g of superglue was used in the fuming cabinet as there were other items as part of the workload that were also being treated. A BY40 stain was applied so an optical scan could take place of the surface.

A BASler ACA1300-30uc camera with a yellow filter (75 mm×75 mm No.12, Kodak Wratten gelatin filter, excitation wavelength: 510–530 nm) was used to image the surface of the samples stained with BY40. Using a camera instead of ToF SIMS and a bespoke stepper motor and stepper motor driver, a similar set up was created to create an image of the surface of the samples. The sample was rotated in increments of 1.9° and a photograph was taken using the camera after each rotation. Software written in LabView was used to extract image strips from the top of the surface of the casing. These strips were stitched together to form a complete image of the surface of the sample. To avoid any distortion of the strip captured, the camera was mounted vertically with the lens facing the surface of the sample. A fluorescent light source, with a wavelength of 405 nm, was used to illuminate the surface. To ensure the light was normally incident on the surface a beam splitter was used. A bright field image was taken using ambient light and no filters.

3.5 Assessing the deformation of a pattern applied to a cylinder

To determine if there was an issue with the cylindrical nature of the casing and how that would alter a fingerprint being deposited on the surface, a control test was conducted using a small piece of foam, ink, a machined cylinder with the same dimensions as a 12mm Mark Webley round, and a piece of paper.

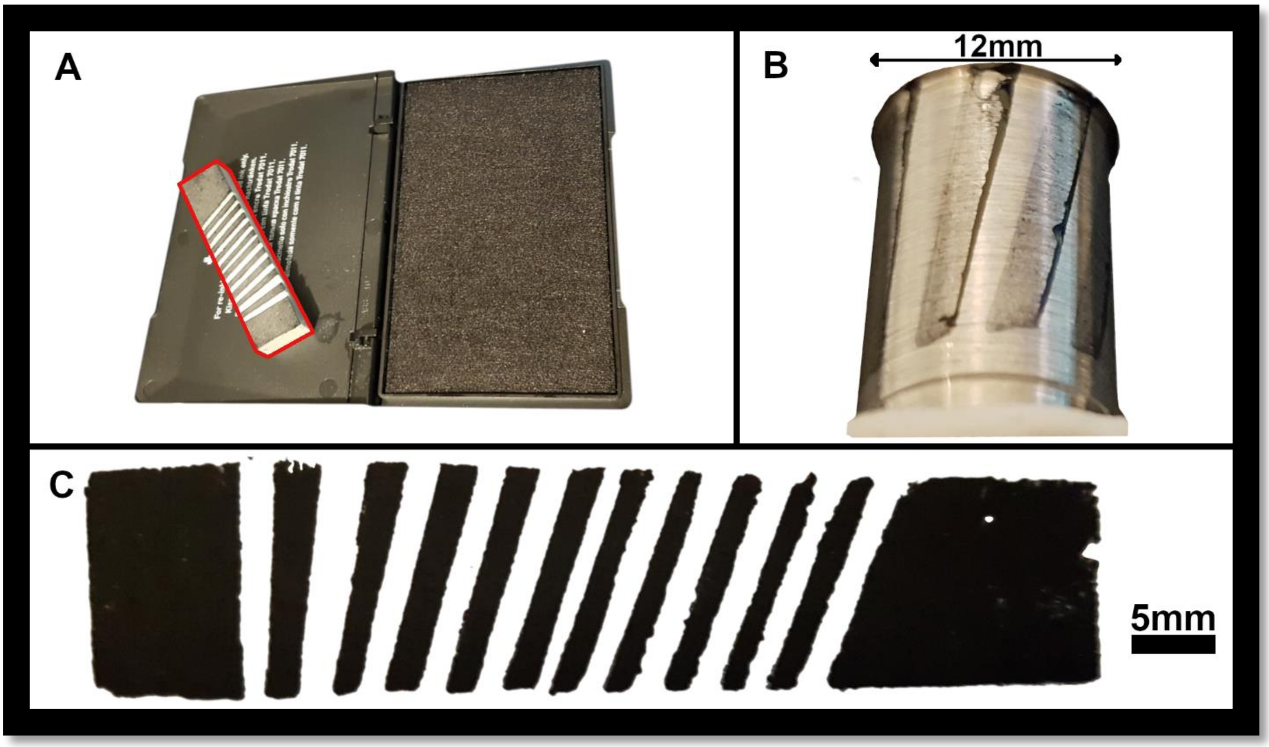

Figure 3.3: In photo A, the carved foam block, highlighted in red, can be seen resting on an ink pad. This ink pad coated the areas of positive relief on the foam and was also used in the depletion series. B shows the marked surface of the steel dummy round with the ink clearly part of the pattern see on the foam block. Scan C shows the ink print caused by the foam block when pressed onto ordinary white, printer paper. This image is compared with the developed ToF SIMS image of photo B.

The foam block (highlighted in red), A in figure 3.3, had grooves cut from it so there would be a geometric pattern on the surface where ink or steel would be in contact. The foam block was cleaned of dust and debris by using a damp paper towel and then left to dry completely. It was left for 1 hour in ambient conditions to do this. The block was pressed onto the surface of the ink pad, A in figure 3.3, for 5 seconds to allow ink to transfer to the surface of the foam. This foam was then placed onto a level surface and the steel replica round was slowly rolled across the surface of the foam. This action transferred the pattern from the foam onto the surface of the ideal casing. The result of the roll can be seen in on the steel replica round, B in figure 3.3. The areas of ink that are more opaque are due to the surface tension of the ink. As the casing rolled across the foam the ink would transfer and as the areas in contact would lift from the foam the ink would pull from the foam and these pools of ink would form at the edges of the pattern.

After the ink pattern had been transferred to the casing, a depletion series was conducted for the foam block. Several ink prints were deposited on ordinary, white printing paper, one after each other with the prints becoming gradually less detailed as the ink began to deplete. Using the same method as before, the foam was pressed onto the ink pad and then pressed onto the white paper with 5 second gaps between each press onto the paper. The clearest candidate, C in figure 3.3, as the edges are mostly defined, and the blocks are solid and opaque.

The machined, ideal casing was handled in the same way as the real round with precautions taken not to contaminate the surface with any unwanted materials. All cleaning, preparation, processing, and analysis were identical except the ion profiles. When using ToF SIMS, the same parameters were set, and the same stitching algorithm was used during the process of the images. Instead of using ion profiles that would show eccrine secretions, ions that indicate the presence of ink were used so the pattern could be seen in the scans.

3.6 Post analysis processing and stitching program development

Two hundred and nine sets of data were produced from scanning approximately 110% of the surface of the sample. Theses sets would need to be converted into images and then these images would need to be "stitched" together to form a complete image of the surface of the casing. Both processes were performed by a program written in python using the visual studio application.

The scans from the ToF SIMS came in an ascii format with 3 distinct variables. The "X" and "Y" coordinates and the intensity, I, for a chosen ion that was being examined. Using these three variables an image could be created for each scan. During development, the intensity was scaled to a value between 0 and 255 resulting in a spectrum of black to white across the X-Y plane depending on the intensity of the ion at a given point. Later, a colour scale of black to red to orange to yellow was added. This was added to create contrast between the background and the ridges. The program used grayscale to compare boundaries for the stitching part of the process.

Each scan was converted from grayscale to binary format, where each pixel was determined to either be white or black by comparing it to a grey value of 50, where pure white is 255 and absolute black is 0, meaning if the intensity was lower than 50, it was converted to be black, and equal or above was converted to be white. This conversion made it much easier for the boundary comparison as it used a Boolean subtraction method to determine where the overlap between scans were.

To determine where the exact overlap occurred. The program systematically compared the scans starting with a very small overlap with the overlap increasing after each iteration. The two binary images were subtracted from each other leaving many remaining white pixels if the overlap was not a match. The result of total pixels left was stored in an array and the result with the smallest number of pixels remaining would be the most accurate value for the length that the two scans overlapped. If the overlap were perfect and the regions matched exactly, the subtraction would leave zero pixels.

The regions for comparison spanned 10 to 30 pixels wide as the rotation of the casing each time must result in the overlap in this zone. The width of overlap would generally be found to be between 10 and 20 pixels. If there was no error in the rotation and the casing did not slip at all as the motor rotated, it then the overlap would be exactly 10 pixels each time. The reality is that there was an issue with the friction between the base of the casing and the plate mounted to the rotating axis of the motor.

This process was tested using images of regular geometric patterns. These images were sliced into many strips that had varying amounts of overlap between them. These strips were then put through the program and it was able to find the correct orientation each time for each region of overlap. This is proven by the stitched image of the fabricated slices being identical to the original image. Furthermore, the comparison between the stitched scans of the 12mm Mark Webley round and a scan of the ink version of the fingerprint shows the success of the stitching program and its accuracy.

3.7 Ink depletion series

A comparison was needed for the scans taken by ToF SIMS. A depletion series of ink prints were deposited by the same subject by using an ink pad and ordinary white, printer paper.

Figure 3.4: This image shows 5 prints as part of a depletion series done by the subject. Print 3 was chosen to be compared with the developed images by ToF SIMS. The scale bar in frame 3 is 1cm long.

The subject coated their thumb by using an ink pad. The fingerprint was then applied to the paper by pressing the thumb to the paper. The process of depositing prints onto paper was repeated a further 4 times without refreshing the ink on the surface of the thumb. This created a depletion series of prints on the paper as the amount of ink lessened each time. These were the prints used as a reference point that would be compared with the scans generated by the ToF SIMS tests and analysis.

Print number 3 was used for comparison purposes as it was a clear, full print with no excess ink filling in the gaps between the ridges in the fingerprint.

4. Results and Discussion

4.1 Results and images yielded by using ToF SIMS

Out of hundreds of unique ions that were detected on the surface of the casing, 10 stood out as showing relatively high levels of intensity meaning there was a significant amount of these substances on the surface. It can be deduced that the relative abundance of these ions was due to the contact between the skin of the finger and the surface of the casing. It is unlikely that these ions in their abundance would be due to contamination. By looking at the regions of the surface where the fingerprint was not present, it is shown that the casing is free from significant numbers of these ions. Due to the distribution of the ions seen in figures 4.1 and 4.2, it can be asserted that the pattern was caused by friction ride detail. There were other ions that showed some pattern to a much lesser degree and so just the clearest 10 were included in this section.

Three sets of data for each ion have been recorded and presented in figures 4.1 and 4.2. The first set was gathered by using ToF SIMS 1 day after the print had been deposited on the casing. The same process was repeated 1 week after the deposition and then finally 7 months. There is little difference between the scans over the course of 7 months showcasing ToF SIMS non-destructive capabilities as well as indicating that the prints persist over this length of time when kept within a controlled environment at ambient temperature. More quantitative analysis would be needed to confirm that there is no degradation over time as relative values are used in the images. The intensity of each ion was preserved as well as the ability to generate an image with a pattern resembling a fingerprint. The print generated by the $C_3H_8N^+$ ion is used for the comparison with the ink prints taken from the subject during the depletion series conducted.

As the surface of the round was thoroughly cleaned with acid, methanol, and water, any contaminants were removed. As the round was kept in controlled conditions free from contamination, it is reasonable to suggest that the only ions that were present during the scanning of the surface were those provided by the sweat of the finger. The only other ions that are able to be seen are that of the composition of the substrate. This was apparent when examining the areas free from any fingermarks.

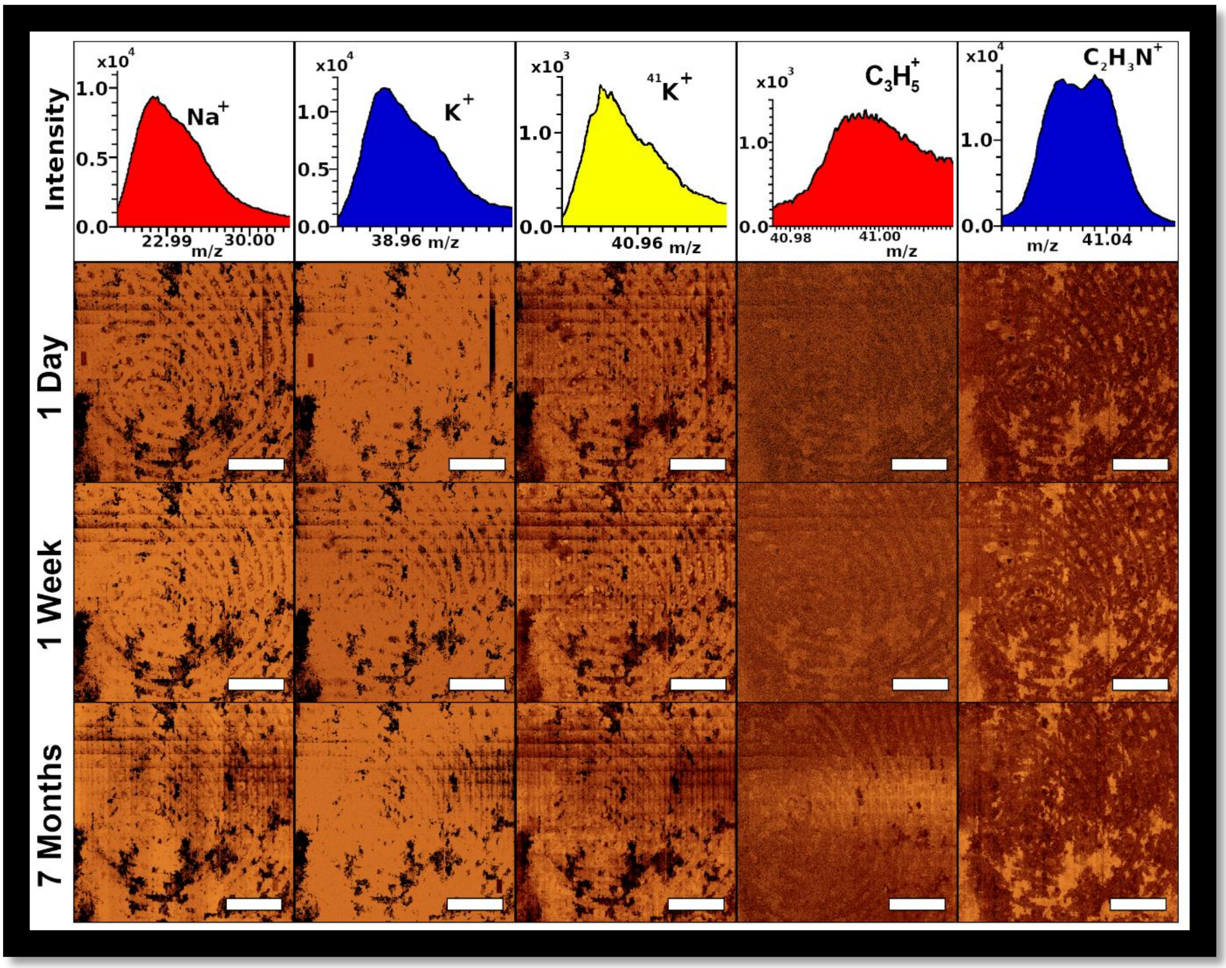

Figure 4.1: ToF SIMS images of 5 different ions present on the surface of the round. The intensity of the ion is shown above the latent prints along with the mass to charge ratio of the ion. The developed prints for the 1 day, 1 week and 7 months tests have been included next to each other to better see how time affects the quality of the latent print on the substrate.

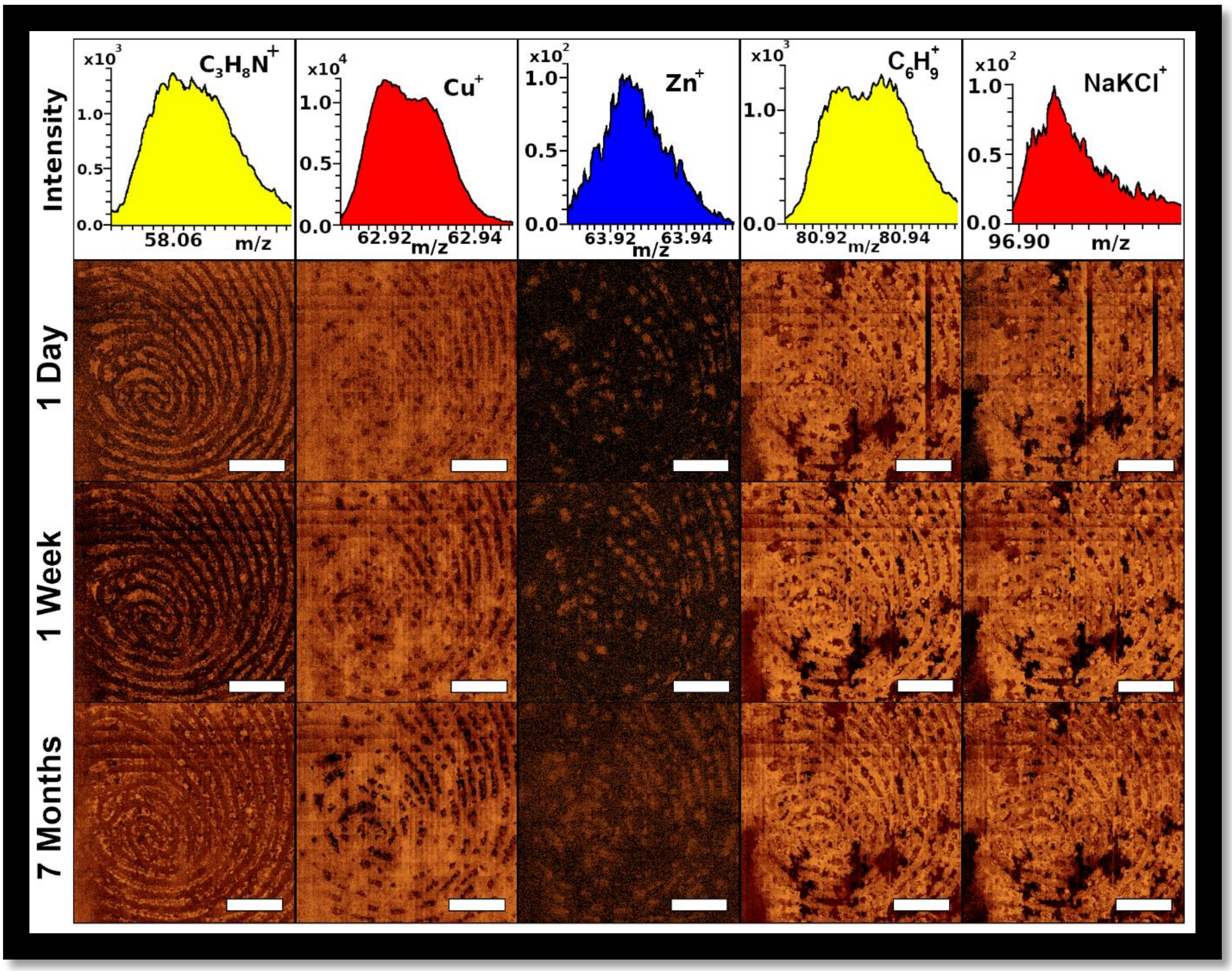

Figure 4.2: ToF SIMS images of 5 further ions present on the surface of the round. The intensity of the ion is shown above the latent prints along with the mass to charge ratio of the ion. The developed prints for the 1 day, 1 week and 7 months tests have been included next to each other to better see how time affects the quality of the latent print on the substrate.

4.2 Results and images yielded by using conventional methods

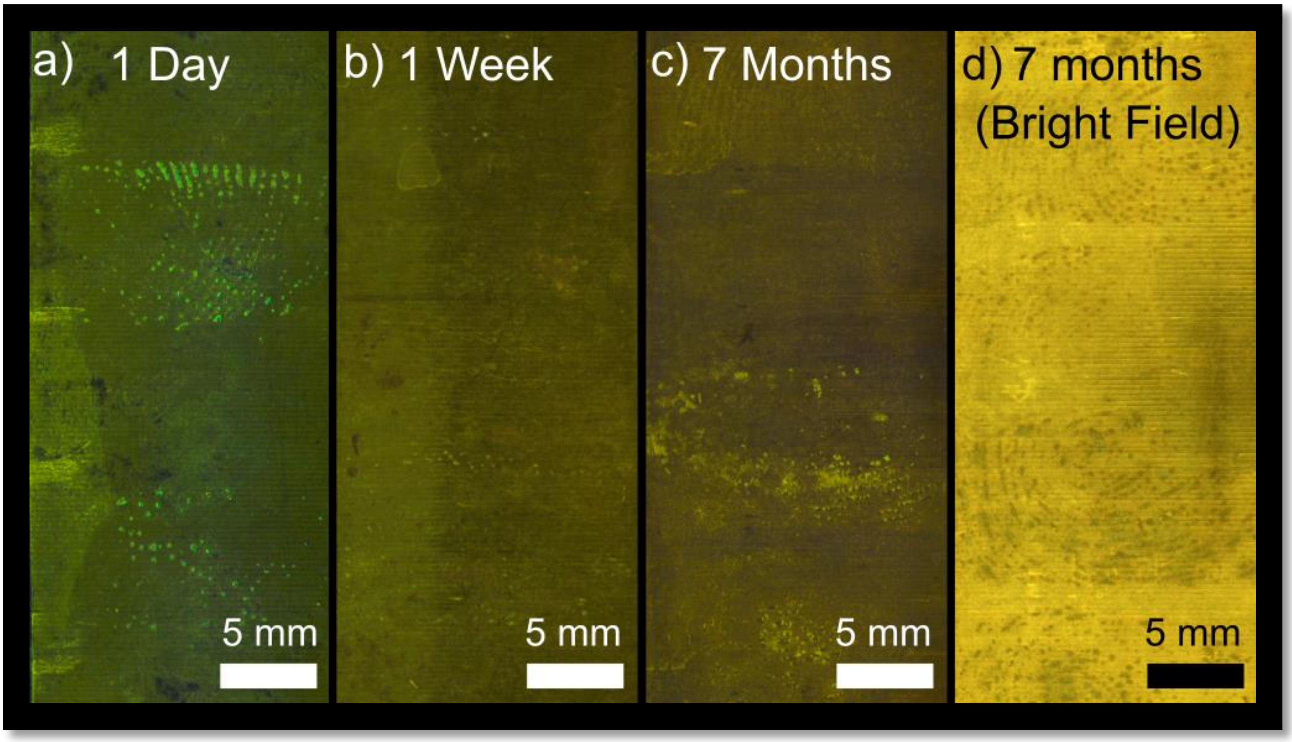

Figure 4.3: Images for the 3 separate rounds when subjected to conventional developing methods. These rounds were subjected to a superglue fuming and coated with a BY40 stain to enhance the contrast of the developed mark.

Figure 4.3 showcases the level of quality to be expected from conventional methods when dealing with the brass casing from a round. The ridge detail can be seen to some degree in the 1-day old mark, with no level 2 minutiae being visible. The prints cannot be observed after 1-week and the same is true after 7-months with no visible minutiae.

The prints using a BY40 stain would be classed as a partial print as ridge detail can be seen over roughly 50% of the mark. However, it is incredibly difficult to see any minutiae within this image. As the ridges across the print are partial and broken, the areas where bifurcations and terminations of ridges would be present cannot be identified as the lines of the ridges are not visible. This has resulted in a mark that cannot be compared easily with either the mark from ToF SIMS or ink.

4.3 Comparing ToF SIMS, Ink and Optical images

The subjects' fingermark and fingerprint can be seen by using 2 different methods, Tof SIMS, A in figure 4.4, and ink, B in figure 4.4. Each of these methods have yielded prints of different quality with the ink print being the standard that the other will be measured against.

The Ink print shows a great level of detail and would be akin to the prints that would be taken if law enforcement were to request finger marks from a subject. Level 2 details such as bifurcations and ridge endings on the finger can be easily identified and highlighted in an overlay in figure 4.5. The same is true of the ToF SIMS image. The level of detail is high and, at a glance, is superior to the ink print. The bifurcations and ridge endings of the friction ridge detail are very clear and can be easily identified. There are even some level 3 details (not labelled and identified) emerging showing the quality of this image, but they will not be used for comparison as that level of detail is not present in the ink print.

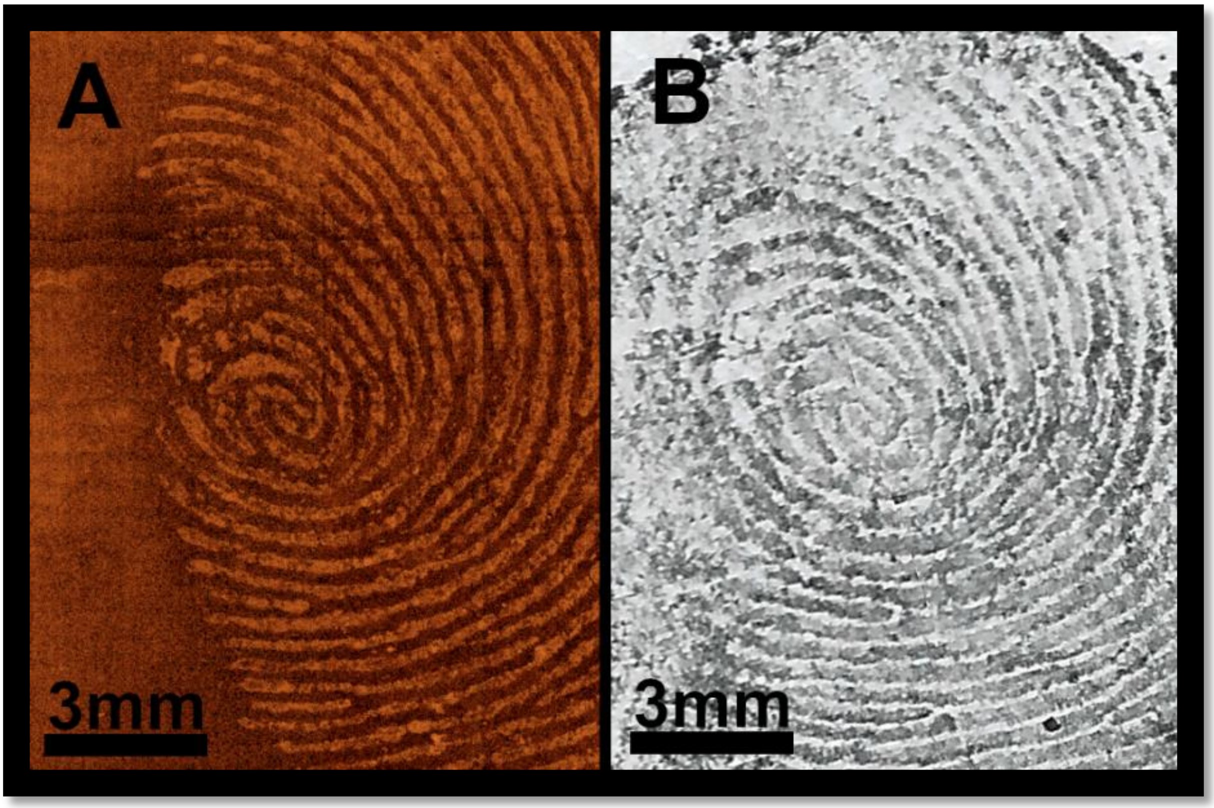

Figure 4.4: In this figure, image A shows the clearest developed fingermark generated by using ToF SIMS. Image B shows ink thumb print 3 from the depletion series conducted by the subject.

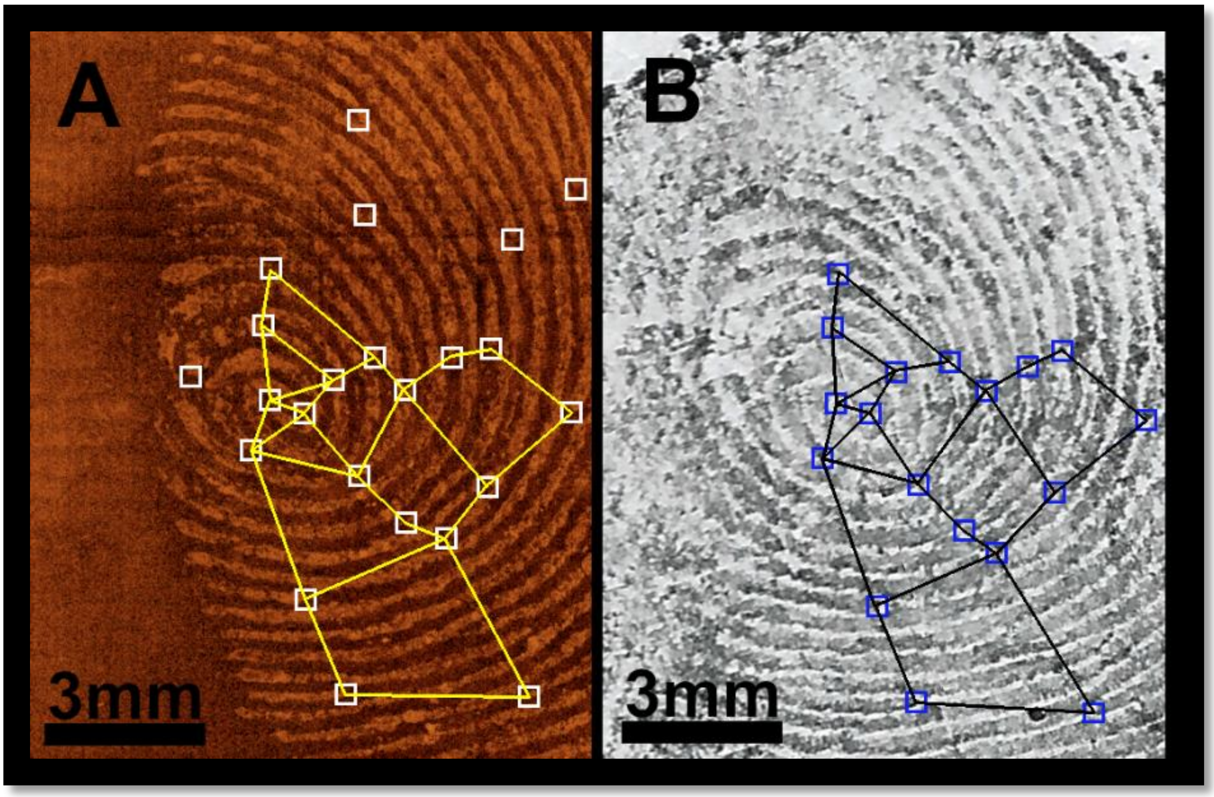

Figure 4.5: For both images of the finger marks, minutiae are identified and then connected via bridges to aid distance comparison and to showcase the similarity between the two patterns.

For each of the images obtained by different methods, the level 2 minutiae were identified by eye. Two types of minutiae, bifurcations, and ridge endings were looked for by the inspector within the mark. These were identified by using a small square of a contrasting colour. The centre of the square was placed directly over the inside corner of the bifurcation or the tip of the ridge ending. The bridging links were placed according to the location of the identification squares. Another contrasting colour was chosen, and the bridge was set to link the centre of two neighbouring squares. This was done to create a unique shape with dimensions relating to the geometry of the finger mark. It is these profiles that will be used to compare the mark from using ToF SIMS and the ink print. By overlaying the different profiles and using the scale bars present on the images, the difference in distance between the same two minutiae can be quantified. This can be seen in figure 4.6.

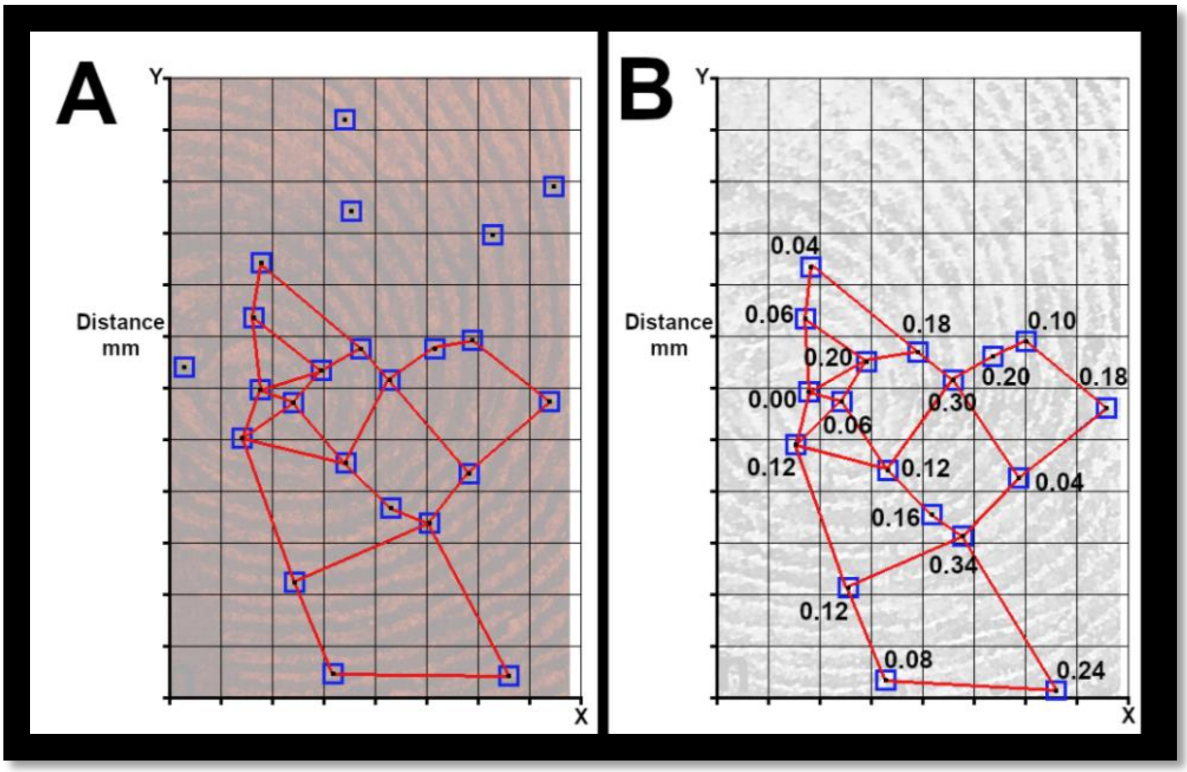

Figure 4.6: With a 1mm grid plotted over the minutiae profiles, the difference in distance between each pair of minutiae have been measured with the range of variation between 0.00mm and 0.34mm. With the first image from ToF SIMS used as a base line, the points on the ink print are given a value denoting the difference in distance for the corresponding minutiae.

Using the inner most ridge ending at the core of the loop on the fingermark as a base, the difference in distance from their counterparts in the other images were calculated. This can be seen, as the reference point had a difference of 0 mm and the others had non-zero values showing variation between the ToF SIMS profile and the profile of the ink image. Where there are larger distances registered, it shows that the difference between the two images for those minutiae is greater. The values next to each identifier are recorded in mm. In Figure 4.6 it is shown that the difference in location between two corresponding minutiae has a maximum value of 0.34 mm, a mean value of 0.14 mm. Furthermore, only 3 of the 18 points compared had a value over 0.20 mm meaning 83% of the minutiae locations differed by 0.20mm or less. Considering the width of a fingertip is in the region of 10 mm, the difference in position of the minutiae is small in comparison.

4.4 Results and images of a rigid pattern applied to a cylinder

To determine where inaccuracies between the ToF SIMS image and ink image stem from, the control test using a carved piece of foam and an ideal version of a 12mm Mark Webley round was analysed. Using a similar technique, ToF SIMS was used to scan the surface of the fake round and an appropriate ion was selected that showcased the pattern that was deposited onto the case using the foam. The side-by-side comparison between the ink print and the ToF SIMS image can be seen in figure 4.7.

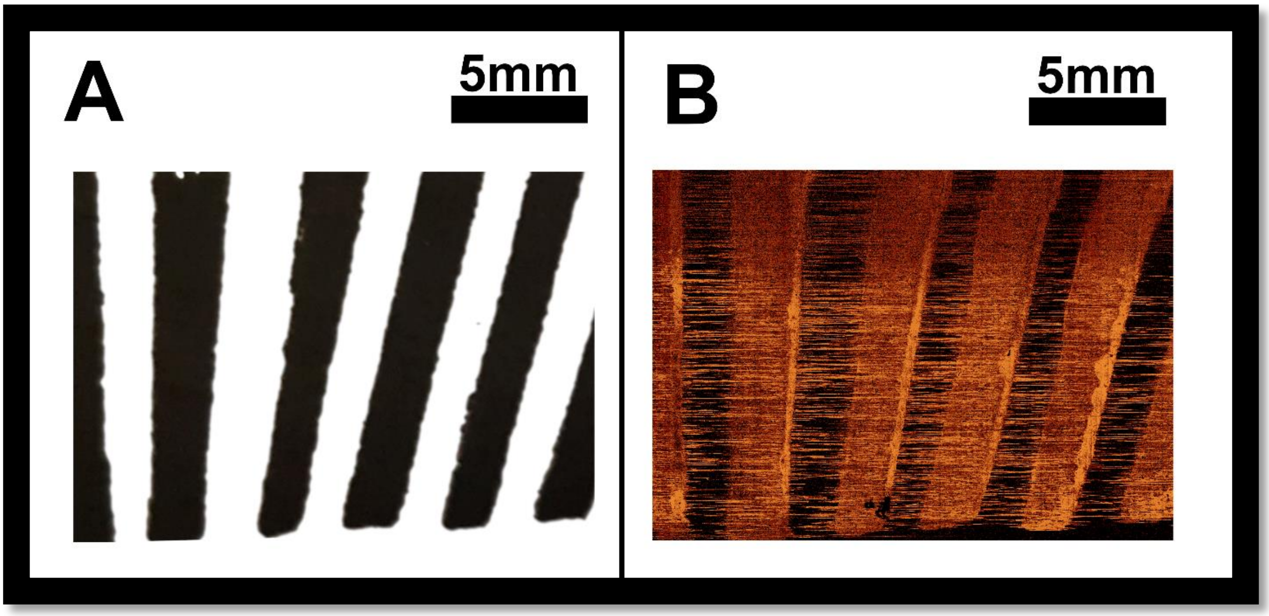

Figure 4.7: Scan A shows a close up of a region of the ink print left by the carved foam when pressed on white paper. Image B is the same region developed by ToF SIMS.

The two images both show the overall pattern of the carved foam. Unfortunately, the sharp detail of the edges has been lost in both the ink print and the ToF SIMS scan. While the foam is waterproof, there were clearly areas on the edges where excess ink built up and subsequently was transferred to the paper when the ink print was created. Although the grooves in the foam were cut very cleanly with a sharp scalpel blade, there may have been unseen remnants of foam left on the edges that contributed to the rough edge that can be seen in the ink print image.

While the edges of the areas of intensity are cleaner than that of the ink print, there was an issue with the ink “bleeding” across the surface of the steel round. While the round was smooth in the same way a real bullet would be, the surface of the casing had a brushed effect after it was polished and refined. This brushed effect left very small grooves in the surface of the fake round that the ink travelled through after it was transferred via the foam. Visually this effect could not be seen, but all the ions from the ToF SIMS scan that showed a complete print also showed this bleeding across the surface of the case. Both these issues relating to the edge of the positive areas of the pattern make it hard to draw a comparison between this control test and the fingerprint analysis.

The overall vector of the pattern edges of the ink print and ToF SIMS image can be used for comparison. Doing this will still reveal any differences between the two images as well as identify similarities.

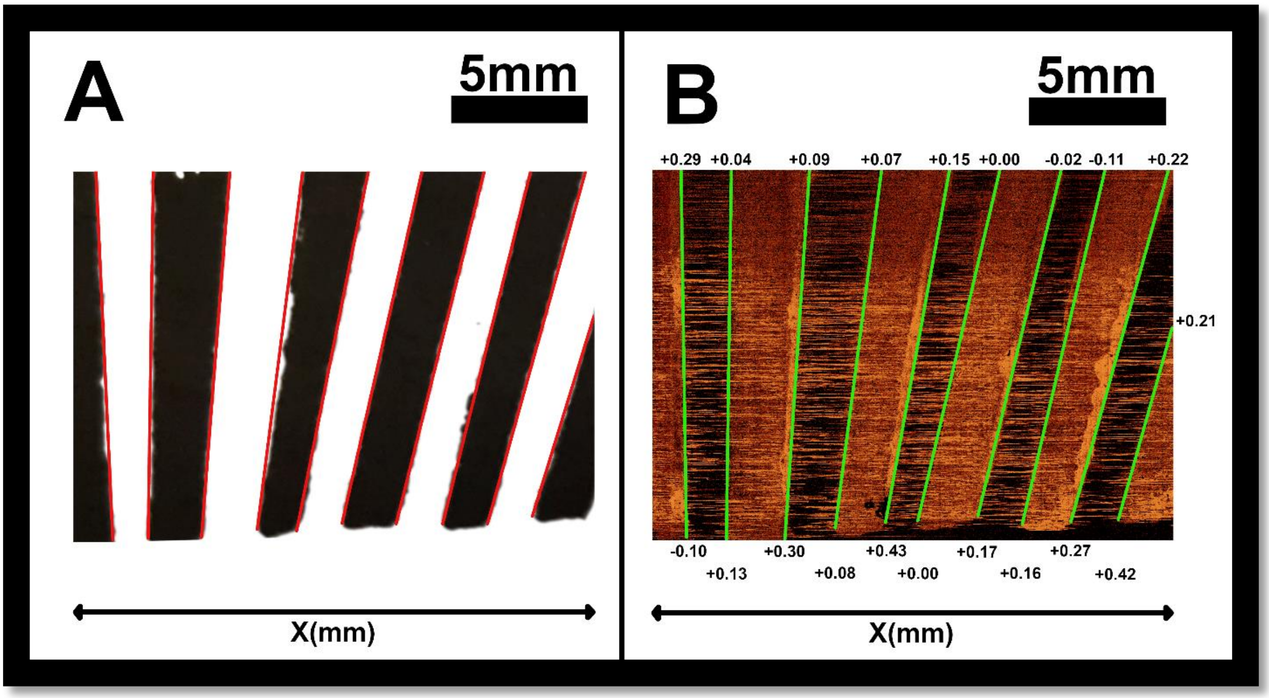

Figure 4.8: For each pattern, the edges of the positive areas have been framed with straight lines. The top and bottom of these lines have been compared with their counterparts to determine the difference in distance relative to the border of the right side of the 4th strip.

The left and right edges of each positive region of the ink pattern was compared with their counterparts in the ToF SIMS image. From drawing lines of best fit along the edge of the positive regions, any distortion of the pattern can be analysed. This was done by matching the right edge of the 4th regions from both images and then comparing the other left and right edges with their counterparts. The numerical values denote the difference in distance in mm and whether the difference is positive or negative, meaning the edge has been distorted to the right or left respectively. Larger values show a greater level of distortion between the two prints.

When the edges are “cleaned” in this way, where the bulges and missing areas are ignored and they are treated as uniform, solid areas of positive relief, the patterns match well with minimal distortion. The maximum amount of distortion is 0.42mm with an average distortion for each edge of 0.17mm. 74% of the edges had a distortion of 0.22mm or less showing that these two scans are a very close match to one another with little distortion occurring when the print was transferred to the casing when rolling the round across the ink covered carved foam. This value of distortion is similar with that of the fingermark analysis. These are small variations considering the size of the objects depositing the patterns for measurement.

5. Conclusion

The capability of ToF SIMS for developing latent fingermarks on the surface of ammunition has been demonstrated. High-quality fingermark images were developed using this technique over the course of 7 months. It was clearly shown that this technique is not impeded by time as other methods might with the 7-month-old mark being of an equal quality with the 1-day-old mark. Furthermore, with the use of conventional techniques in the form of superglue fuming with a BY40 stain as a comparative method, ToF SIMS was shown to be far superior. Where the conventional method revealed partial level 1 detail for comparison with the ink print, ToF SIMS showed several level 2 details with some level 3 details being partially evident. The difference in the locations of the minutiae in the ToF SIMS image and the ink print were small and did not impede the matching of the corresponding minutiae. The resulting images were of a high quality and showed no evidence of deterioration over a 7-month period, which shows that the use of ToF SIMS as a method to develop latent fingermarks on unfired ammunition would be valuable as a forensic tool.

6. Improvements and future work

The main improvement that needs to be made in any future work surrounds the stage and mounting in the ToF SIMS chamber. As described previously, there was an issue with the round not always completing the desired rotation due to the lack of friction between the base of the round and the flat metal where the round was mounted. This part of the stage is in need of a redesign so that the rotation of the round will be reliable and consistent at the end of each scan.

A study into using this same method on latent fingermarks on ammunition still has more to offer by analysing fired rounds. ToF SIMS has proven itself to be a highly sensitive technique and so only some organic or inorganic materials deposited by the friction ridges of the skin need to survive the combustion of the round in order for ToF SIMS to be able to generate an image. Furthermore, ToF SIMS has a capability to depth profile so even if the secretions present in the fingermark are covered by combustion products that would obscure the mark, ToF SIMS can analyse several monolayers below the surface.

Bibliography

[1] N. Stripe, *Statistical Bulletin: Office for National Statistics, Crime in England and Wales, Year Ending September 2020*, Office for National Statistics [GB], (2020). https://www.ons.gov.uk/peoplepopulationandcommunity/crimeandjustice/bulletins/crimeinenglandandwales/yearendingseptember2020 [Accessed 28/03/2021]

[2] V. Sears, S. Bleay, H. Bandey, V. Bowman, *A methodology for finger mark research*, Sci. Justice 52 (3) (2012) 145–160.

[3] C. Champod, C. Lennard, P. Margot, M. Stoilovic, *Fingerprint and other ridge skin impressions*, CRC Press (2004).

[4] National Institute of Justice (U.S.), *The fingerprint sourcebook*, Washington, DC, U.S. Dept. of Justice, Office of Justice Programs, National Institute of Justice, (2011). http://purl.fdlp.gov/GPO/gpo18039 [Accessed 28/03/2021]

[5] M.M. Houck (Ed.), *Forensic Fingerprints*, Academic Press, 2016.

[6] S. Bunter, *How Long Can an Identifiable Fingerprint Persist on an Exterior Surface*, CSEye - The home of crime scene science articles, April, (2014).

[7] H. Lee, R. Ramotowski, R. Gaensslen (Eds.), *Advances in Fingerprint Technology*, CRC Press, (2001).

[8] G. Wightman, F. Emery, C. Austin, I. Andersson, L. Harcus, G. Arju, C. Steven, *The interaction of fingermark deposits on metal surfaces and potential ways for visualisation*, Forensic Science International 249 (2015) 241-254.

[9] T. Thandauthapani, A. Reeve, A. Long, I. Turner, J. Sharp, *Exposing latent fingermarks on problematic metal surfaces using time of flight secondary ion mass spectroscopy*, Sci. Justice 58 (6) (2018) 405-414.

[10] T. Thompson, S. Black (Eds.), *Forensic Human Identification: An Introduction* (1st ed.). CRC Press, (2006).

[11] Home Office, *National DNA Database Strategy Board Biennial Report 2018 – 2020* (2020) https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/913015/NDNAD_Strategy_Board_AR_2018-2020_print.pdf [Accessed 28/03/2021]

[12] N. Quinche, P. Margot, *Coulier, Paul-Jean (1824–1890): a precursor in the history of fingermark detection and their potential use for identifying their source (1863)*, Journal of Forensic Identification, 60 (2) (2010) 129-134.

[13] C. Girelli, B. Lobo, A. Cunha, J. Freitas, F. Emmerich, *Comparison of practical techniques to develop latent fingermarks on fired and unfired cartridge cases*, Forensic Science International, 250 (2015) 17–26.

[14] A. Dominick, K. Laing, *A comparison of six fingerprint enhancement techniques for the recovery of latent fingerprints from unfired cartridge cases*, Journal of Forensic Identification, 61 (2) (2011) 155-165.

[15] J. Hoover, *Fingerprint. Encyclopedia Britannic*a (2016) https://www-britannica-com.ezproxy.nottingham.ac.uk/topic/fingerprint [accessed 28/03/2021]

[16] P. Komarinski, *Automated Fingerprint Identification Systems (AFIS)* (1st ed.), Elsevier Academic Press (2005).

[17] G. Bumbrah, R. Sharma, O. Jasuja, *Emerging latent fingerprint technologies: a review*, Research and Reports in Forensic Medical Science, 6 (2016) 39-50.

[18] M. Bailey, M. Ismail, S. Bleay, N. Bright, M. Elad, Y. Cohen, B. Geller, D. Everson, C. Costa, R. Webb, J. Watts, M. Put, *Enhanced imaging of developed fingerprints using mass spectrometry imaging*, Analyst, 138 (21) (2013) 6246-6250.

[19] Fingerprint quality standards specialist group, *Fingerprint examination: terminology*, definitions and acronyms, Forensic Science Regulator (2020). https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/914266/126_FSR_fingerprint_terminology.pdf [Accessed 28/03/2021]

[20] I. Lewis, *Ever had your fingerprints taken? Meeting the challenges of 21st Century access control*, Biometric Technology Today, 2014 (5) 9-11

[21] R. Adhami, P. Meenen, *Fingerprinting for security*, IEEE Potentials, 20 (3) (2001) 33-38.

[22] L. Thomas, *The physics of fingerprints and their detection*, J. Phys. E: Sci. Instrum. 11 (1978) 722-731.

[23] A. Jain, Y. Chen, M. Demirkus, *Pores and Ridges: High-Resolution Fingerprint Matching Using Level 3 Features*, IEEE Transactions on Pattern Analysis and Machine Intelligence, 29 (1) (2007) 15-27.

[24] C. Neumann, *Fingerprints at the crime-scene: Statistically certain, or probable?* Significance, 9(1) (2012) 21-25.

[25] R. Hicklin, *Anatomy of Friction Ridge Skin*, Encyclopedia of Biometrics, 14-19.

[26] K. Nandar Win, K. Li, J. Chen, P. Viger, K. Li, *Fingerprint classification and identification algorithms for criminal investigation: A survey*, Future Generation Computer Systems 110 (2020) 758-771.

[27] B. Scruton, B. Robins, B. Blott, *The deposition of fingerprint films,* Journal of Physics D: Applied Physics, 8 (6) (1975) 714.

[28] A. Girod, R. Ramotowski, C. Weyermann, *Composition of fingermark residue: a qualitative and quantitative review,* Forensic science international, 223(1-3) (2012) 10-24.

[29] S. Cadd, M. Islam, P. Manson, S. Bleay, *Fingerprint composition and aging: a literature review*, Science & Justice, 55 (4) (2015) 219-238.

[30] T. Thandauthapani, *The role of wetting effects on the development of latent fingermarks*, PhD thesis, University of Nottingham (2019)

[31] S. Li (Ed.), *Encyclopedia of Biometrics: I - Z., Volume 2*, Springer Science & Business Media (2009).

[32] T. Kent, *Section 2: Chemistry and Visualization (Sequential Treatment and Enhancement) Forensic Fingerprints*, Academic Press (2016)

[33] S. Bleay, R. Croxton, M. De Puit, *6. Optical detection and enhancement techniques. Fingerprint Development Techniques: Theory and Application*, John Wiley & Sons, (2018) 111-144.

[34] G. Sodhi, J. Kaur, *Powder method for detecting latent fingerprints: a review*, Forensic Science International, 120 (3) (2001) 172-176.

[35] C. Lennard, C. Champod, P. Margot, M. Stoilovic, *Chapter 4: Fingermark detection and Enhancement. Fingerprints and other ridge skin impressions*. CRC press (2004)

[36] S. Bramble, A. Cantu, R. Ramotowski, J. Brennan, *Deep red to near infrared (NIR) fluorescence on gentian violet-treated latent prints,* Journal of Forensic Identification, 50 (1) (2000) 33-49.

[37] S. Bleay, T. Kent, *The use of infra-red filters to remove background patterns in fingerprint imaging*, Fingerprint Whorld, 31(122) (2005) 225-238.

[38] G. Sodhi, J. Kaur, Po*wder method for detecting latent fingerprints: a review*. Forensic science international, 120 (3) (2001) 172-176.

[39] E. Menzel, *Fingerprint detection with lasers*, New York: M. Dekker, (1980) 45-78.

[40] M. Choi, T. Smoother, A. Martin, A. McDonagh, P. Maynard, C. Lennard, C. Roux, *Fluorescent TiO2 powders prepared using a new perylene diimide dye: Applications in latent fingermark detection*, Forensic Science International, 173(2-3) (2007) 154-160.

[41] B. Wilshire, *Advances in fingerprint detection*, Endeavour, 20 (1) (1996) 12- 15.

[42] R. Gaensslen, R. Ramotowski, H. Lee, *Chapter 4: Methods of Latent Fingerprint Development. Advances in fingerprint technology*. CRC press (2001).

[43] V. Bowman, *Manual of fingerprint development techniques* (2nd ed.) Sandridge: Home Office Police Scientific Development Branch (2004).

[44] P. Czekanski, M. Fasola, J. Allison, *A mechanistic model for the superglue fuming of latent fingerprints*, Journal of forensic sciences, 51 (6) (2006) 1323-1328.

[45] B. Chesher, J. Stone, W. Rowe, *Use of the Omniprint™ 1000 alternate light source to produce fluorescence in cyanoacrylatedeveloped latent fingerprints stained with biological stains and commercial fabric dyes*, Forensic science international, 57(2) (1992) 163-168.

[46] S. Wargacki, L. Lewis, M. Dadmun, *Understanding the Chemistry of the Development of Latent Fingerprints by Superglue Fuming*, Journal of Forensic Sciences, 52 (2007) 1057-1062.

[47] E. Brown, The Cyanoacrylate Fuming Method (2003). https://www.soinc.org/sites/default/files/uploaded_files/forensics/For_supergluing.pdf [Accessed 28/03/2021]

[48] K. Farrugia, J. Fraser, N. Calder, P. Deacon, *Pseudo-Operational Trials of Lumicyano Solution and Lumicyano Powder for the Detection of Latent Fingermarks on Various Substrates*, Journal of Forensic Identification, 64 (6) (2014)

[49] Scientific Working Group on Friction Ridge Analysis, Study and Technology (SWGFAST), *Standards for examining friction ridge impressions and resulting conclusions (latent/tenprint)*, version 1.0. (2011)

[50] A. Jain, Y. Chen, M. Demirkus, *Pores and ridges: High-resolution fingerprint matching using level 3 features,* IEEE Transactions on Pattern Analysis and Machine Intelligence, 29(1) (2007) 15-27.

[51] J. Fraser, R. Williams (eds.), Fingerprints: Handbook of Forensic Science ed.Routledge, Abingdon (2009)

[52] P. Kellman, J. Mnookin, G. Erlikhman, P. Garrigan, T. Ghose, E. Mettler, D. Charlton, I. Dror, *Forensic comparison and matching of fingerprints: using quantitative image measures for estimating error rates through understanding and predicting difficulty*, PloS one, 9 (5) (2014) e94617.

[53] NHS: How to wash your hands (2019) https://www.nhs.uk/live-well/healthy-body/best-way-to-wash-your-hands/ [Accessed 28/03/2021]

[54] A. Ramos, M. Vieira, *An efficient strategy to detect latent fingermarks on metallic surfaces*, Forensic Sci. Int. 217 (1) (2012) 196–203.

[55] Q. Wei, M. Zhang, B. Ogorevc, X. Zhang, *Recent advances in the chemical imaging of human fingermarks (a review)*, Analyst, 141 (22) (2016) 6172- 6189

[56] D. Touboul, P. Kollmer, E. Niehuis, A. Brunelle, O. Laprévote, *Journal of the American Society for Mass Spectrometry* 16 (2005)1608-1618.

[57] J. Vickerman (ed.), D. Briggs (ed.), *ToF-SIMS: Materials analysis by mass spectrometry* (2nd ed.), IM Pub. LLP, Chichester (2013)

[58] P. Green, I. Gilmore, M. Seah, *Journal of the American Society for Mass Spectrometry* 17 (2006) 514 - 523.

[59] B. Scruton et al, *The deposition of fingerprint films*, J. Phys. D: Appl. Phys. 8 (1975) 714

[60] H. Bandy, S. Bleay, V. Bowman, R.P Downham, V. Sears, *Fingermark visualisation manual,* (1st ed.), Home Office Centre for Applied Science and Technology (CAST), (2014)