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Improving mechanical properties of recycled polypropylene-based composites using Taguchi and ANOVA techniques

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Abstract

This study was aimed at optimising the composition of recycled plastic based composite for improving mechanical performance. The fractions of virgin polypropylene (vPP), talcum powder (talc) and maleic anhydride grafted polypropylene (MAPP) were selected as controllable factors. Taguchi L9 (3³) orthogonal array (OA) was applied as an experimental design tool. Compositions were prepared by extrusion and injection moulding. The performance was measured and expressed in signal to noise (S/N) ratio. The S/N ratios were investigated by the Taguchi coupled analysis of variance (ANOVA) to determine optimal condition. The effectiveness and efficiency of the proposed approach was demonstrated through confirmation tests.

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Keywords: Recycled polypropylene; Mechanical property; Taguchi method; Analysis of variation; Signal to noise ratio

1. Introduction

Under the concept of circular economy, it is more economic and environmental to use recycled plastics to replace virgin plastics, especially for materials of enormous consumption, such as polypropylene (PP) [2]. Also, extending the use of recycled materials could promote the recycling rate [2]. But, recycled PP (rPP) tend to have inferior performance during manufacturing process when compared with virgin materials [1,2], the applications of rPP are thereby limited [2,3,4]. With the addition of fillers, recycled polymer matrix composites (RPMCs) with desirable properties can be produced [2,3,4], and some of them are even better than virgin materials [2]. Talcum powder (talc) is one of the most used reinforcing fillers used for promoting the performance of rPP [2,5,6,7,8]. Talc not only enables to improve both the thermal and mechanical properties of rPP [2,5], but also facilitates the production by reducing and homogenizing the molding shrinkage [6]. Also, talc has a positive stabilising effect on the mechanical properties of PP composites [7,8]. The stabilising effect of talc addition is highly depended on the interfaces

between the talc and the PP matrix [9]. Further, there are some tradeoffs in rPP/talc composites, as tensile and flexural properties increased with addition of talc while impact properties deteriorated [2,8]. Thus, formula analysis, design and optimisation of rPP/talc composite are required to extend its application.

Nomenclature

AEV	Accumulated explained variation
ANOVA	Analysis of variance
DF	Degree of freedom
EV	Explained variation
FM	Flexural modulus
FS	Flexural strength
GRA	Grey relational analysis
HCA	Hierarchical clustering analysis
MAPP	Maleic anhydride grafted polypropylene
MS	Mean of square
MFI	Melt flow index
OA	Orthogonal array

PP	Polypropylene
PC	Principle component
PCA	Principle component analysis
PCE	Principal component estimate
RPMC	Recycled polymer matrix composite
rPP	Recycled polypropylene
RSM	Response surface methodology
SEM	Scanning electronic microscopy
S/N	Signal to noise
SS	Sum of square
SEBS	Styrene-ethylene-butylene-styrene
Talc	Talcum powder
TM	Tensile modulus
TS	Tensile strength
vPP	Virgin polypropylene

Formula design of polymeric composites has attracted an increasing attention during recent years, and several approaches have been developed to optimise the composition of PP composites for improving multiple properties [8,10,11,12,13,26]. Leu et al used experimental design to obtain the optimal levels of the selected components in an arranged sequence [10]. Homkhiew et al adapted a D-optimal mixture experimental design in modelling of mechanical characteristics of wood flour filled RPMCs [11]. Ayaz et al used Taguchi method combined with grey relational analysis (GRA) to determine the optimal composition of PP/LLDPE/TiO₂/SEBS composites for improving impact and flexural strength [12]. Taguchi optimisation approach was also adapted by Porras et al for improving tensile properties of biocomposites [26]. Ghasemi et al designed experiments according to response surface methodology (RSM) to optimise the fractions of talc, MAPP and exfoliated graphene nanoplatelets (xGnPs) [13]. In our previous work, hierarchical clustering analysis (HCA) and principal component estimate (PCE) was used to obtain cheapest rPP composites which met the technical requirements of automobile parts [8].

The literature review shows research on formula design for rPP composites is limited [8,11]. Adapting rPP into manufacturing could increase the recycling rate of plastic waste [4,14], and the composition optimisation can be thereby considered to be the first crucial step [8]. Taguchi method and analysis of variance (ANOVA) were applied for this purpose. Taguchi method coupled with ANOVA has been extensively applied in optimising manufacturing procedures of plastic products [15,16,17,18,19,20,21], only limited applications were found in formula design [11,26]. In this study, vPP, talc and MAPP were selected as the controllable factors, and experimental design was based on a Taguchi L9 (3³) OA. The aim of using Taguchi OA is to reduce the number of initial composition trials without sacrificing comparability of controllable factors [22]. Compared with other formula optimisation approaches [8,10,12,13], the trials used in Taguchi method are minimal [11,26]. The tested mechanical properties were expressed in form of signal to noise (S/N) ratios, and the effect of each component was analysed. Due to the single response controlling capacity of Taguchi method [12] and [19], the tested mechanical properties were transformed to a series of scores using principal component

analysis (PCA). The optimal formula was hence obtained via analysing PCA scores and was later verified through confirmation tests.

2. Experimental

2.1. Materials

The rPP pellets used in this study was purchased from Tianqiang Recycling Co., Ltd., Shanghai, China, and have a density of 0.91 g cm⁻³ and a melt flow index (MFI, at 230 °C under a load of 2.16 kg according to ISO1133-1:2011) of 2.47 g/10 min. An injection grade of PP homo-polymer with trade mark of H1500, produced by LG Chem Co., Ltd., Republic of Korea, was used as vPP in the matrix resin. The density and MFI are 0.9 g cm⁻³ and 10.97 g/10 min, respectively. The coupling agent - maleic anhydride grafted polypropylene (MAPP) used in this study is bought from Nanjing Deba Chemical Co.,Ltd, has a density of 0.9 g cm⁻³ and a MFI of 88.67 g/10 min. Talc used in this study is purchased from Ningbo Haike, a local supplier, has an average particle size of 12.5 µm and a density of 2.75 g cm⁻³.

2.2. Design of Compositions

The 3 selected components - vPP (wt.%), talc (wt.%) and MAPP (wt.%) and their levels are shown in Table 1. The full factorial design requires 3³=27 possible combinations of tests to evaluate the effects of the selected components, while only a total of 9 experiments required in a Taguchi L9 (3³) OA, as shown in Table 2. A significant reduction in the number of trials has been achieved when compared with previously proposed formula optimisation methods [10,11,13].

Table 1. Selected components (vPP, talc and MAPP) and their levels.

Components	Level 1	Level 2	Level 3
vPP (wt.%)	20	40	60
talc (wt.%)	5	10	20
MAPP (wt.%)	0	2.5	5

Table 2. Designed compositions based upon Taguchi L9 (3³) OA.

Trial No	vPP (wt.%)	talc (wt.%)	MAPP (wt.%)
1	20	5	0
2	20	10	2.5
3	20	20	5
4	40	5	2.5
5	40	10	5
6	40	20	0
7	60	5	5
8	60	10	0
9	60	20	2.5

2.3. Sample Preparation

After drying in an oven at 75°C for 12 h and mixing in a high-speed mixer for 3 min, all materials were compounded by a KRSHJ-20 co-rotating twin-screw extruder, whose screw

diameter is 20 mm and L/D=44, with processing temperatures were increased from 170°C to 190°C. The screw rotating speed was set at 150 rpm. The pellets were dried at 75°C for 12 h before being injection moulded into ISO specimens using a Haitian MA1200/370 injection moulding machine in a temperature profile of 190°C-195°C-200°C-200°C-200°C.

2.4. Tests

In this study, the tested mechanical properties were tensile modulus (TM), tensile strength (TS), flexural modulus (FM) and flexural strength (FS). Tensile tests were performed on an Gotech TCS-2000NE universal testing machine at a crosshead speed of 50 mm min⁻¹ following ISO527-2:2012. Flexural tests were performed on an Gotech TCS-2000NE universal testing machine at a crosshead speed of 2 mm min⁻¹ and with a span of 64 mm following ISO178:2010. Average values of 10 tested specimens were reported. The fracture surface of tested specimens was scanned using a Zeiss Sigma scanning electron microscope at an acceleration voltage of 3.00 kV. The targeted surface was coated with gold prior to the SEM analysis.

3. Results and Discussion

3.1. Analysis of S/N Ratios

In this study, since higher mechanical performance was expected, the S/N ratios of tested mechanical properties were thereby calculated using 'the higher the better' equation which shown as follow [19] and [23]:

$$S / N = -10 \log_{10} \left(\frac{1}{n} \sum_{i=1}^n \frac{1}{y_i^2} \right) \quad (1)$$

in which y_i is the i th experiment at the test, n is the total number of trials in the test, and s is the standard deviation of y_i . In this study, $n=10$. The experimental results of the tested mechanical properties and their S/N ratios are shown in Table 3, and the main effects of the selected components are plotted in Fig.1 to Fig.4. Since the highest S/N ratio always yields the optimum quality with minimum variance [22,23], the optimum level of controllable factor can be chosen via comparing the values of S/N ratios [12,18,19,20].

Table 3. Designed compositions based upon Taguchi L9 (3³) OA.

Trial No	TM	S/N of TM	TS	S/N of TS	FM	S/N of FM	FS	S/N of FS
1	1460	63.37	26.29	28.39	1575	63.95	36.43	31.23
2	1720	64.68	26.14	28.35	1707	64.63	37.43	31.46
3	1991	65.96	25.43	28.09	2041	66.19	38.56	31.72
4	1583	63.99	28.62	29.13	1644	64.32	38.83	31.78
5	1658	64.39	28.49	29.09	1808	65.14	40.03	32.05
6	1883	65.54	25.52	28.14	2000	66.02	38.80	31.78
7	1607	64.12	30.91	29.80	1694	64.57	41.12	32.28
8	1653	64.36	29.09	29.28	1783	65.02	40.38	32.12
9	2161	66.67	28.69	29.15	2130	66.56	42.97	32.66

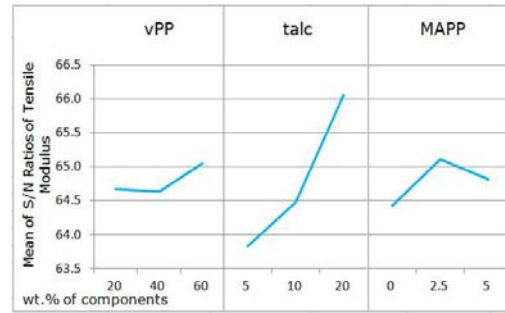


Fig. 1. Plot of S/N ratios against the selected components for TM.

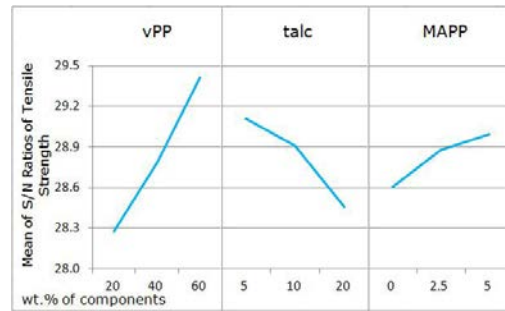


Fig. 2. Plot of S/N ratios against the selected components for TS.

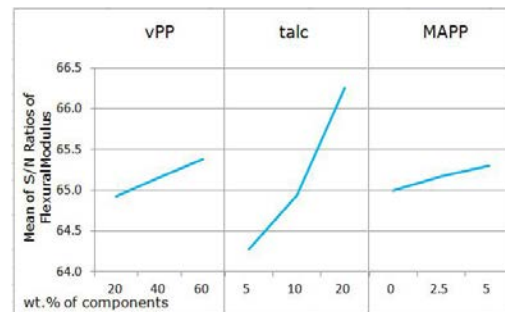


Fig. 3. Plot of S/N ratios against the selected components for FM.

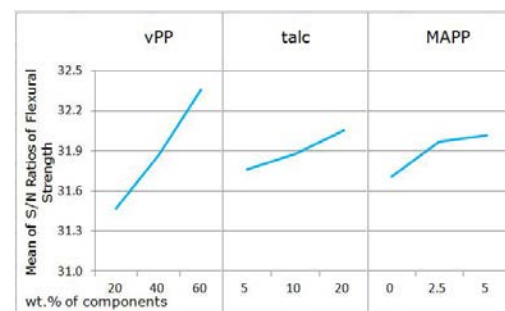


Fig. 4. Plot of S/N ratios against the selected components for FS.

Fig. 1 and Fig. 3 show that the talc was the most influential factor affecting TM and FM, as improvements of 29.79% and

25.57 % have been achieved for TM and FM when talc content was increased from 5 wt.% to 20 wt.%. The enhancement is compatible with our previous work [2], which could attribute to the increased content of rigid talc particles [7] and the higher crystallinity content formed with the increasing talc content [6]. Fig. 5 shows a good dispersion of talc particles within PP matrix which supports this assertion. Fig. 2 and Fig. 4 show talc has a negative effect on TS and FS, while the vPP is the most significant factor in improving TS and FS values followed by the MAPP. These findings are consistent with the results provided by Gu [24].

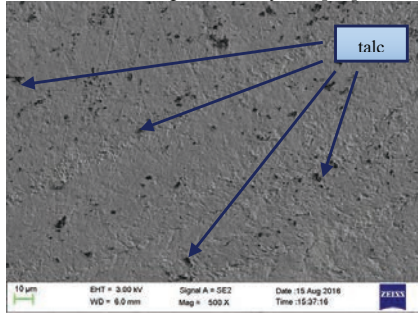


Fig. 5. SEM image of the specimen with vPP=20 wt.%, talc=5 wt.% and MAPP=0 wt.% at the non-deformed state.

The interactions between the selected controllable factors of tested mechanical properties were investigated via the interaction plots shown in Fig.6 to Fig.9. The average S/N ratios of the mechanical properties are shown in the vertical axis in the figures, while the variation of the components are shown in the horizontal axis.

The interactions between talc and MAPP of TM are insignificant since parallel lines are observed in Fig.6 (a). The interactions between vPP and MAPP of TM are most significant, which followed by vPP and talc. In the vPP × MAPP interactions of TM (Fig.6 (b)), the S/N ratios of 0 wt.% MAPP are initially increased with increasing vPP content and finally decreased, while the S/N ratios of 2.5 wt.% MAPP show a completely opposite pattern, decreased initially and later increased with vPP content. The S/N ratios of 5 wt.% MAPP is decreasing with vPP content. In the vPP × talc interactions of TM (Fig.6 (c)), the S/N ratios of 5 wt.% talc and 10 wt.% talc show little change with vPP content, while the S/N ratios of 20 wt.% talc are initially decreased and eventually increased with vPP content. For the S/N ratios of TS, the vPP × talc and vPP × MAPP interactions are insignificant since parallel lines shown in Fig.7 (b) and (c), except for 0 wt.% MAPP in vPP × MAPP which is initially increased and then eventually decreased with vPP content. The interactions between talc and MAPP of TS (Fig.7 (a)) are most significant.

The vPP × MAPP interactions of FM are as the same as that of TM, which are most significant as shown in Fig.6 (b) and Fig.8 (b). In the vPP × talc interactions of FM (Fig.8 (c)), the S/N ratios of 5 wt.% talc are increasing with vPP content, and the S/N ratios of 10 wt.% talc are initially increased and eventually decreased with vPP content while the S/N ratios of 20 wt.% talc have shown an opposite behaviour. For talc ×

MAPP, their interactions are most insignificant as all the S/N ratios are increased with talc content. The generic pattern of interactions between the selected components of FS is similar to the interactions of TS. The vPP × talc and vPP × MAPP interactions were quite insignificant as shown in Fig.9 (b) and (c). The talc × MAPP interactions (Fig.9 (a)) are most significant.

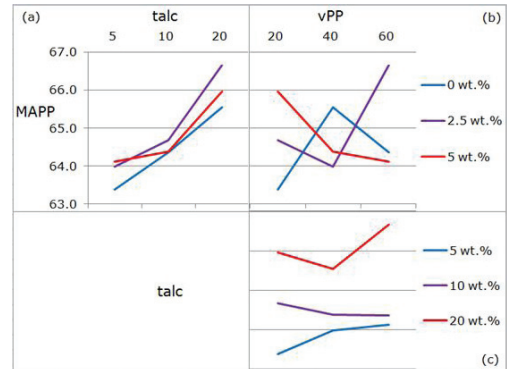


Fig. 6. Interaction plots of (a) talc × MAPP, (b) vPP × MAPP, (c) vPP × talc, for TM.

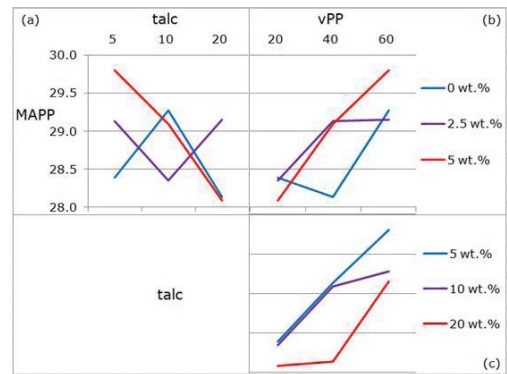


Fig. 7. Interaction plots of (a) talc × MAPP, (b) vPP × MAPP, (c) vPP × talc, for TS.

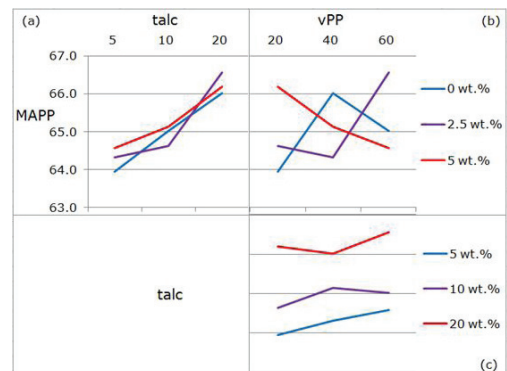


Fig. 8. Interaction plots of (a) talc × MAPP, (b) vPP × MAPP, (c) vPP × talc, for FM.

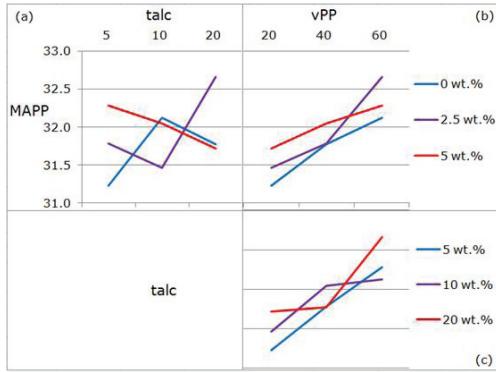


Fig. 9. Interaction plots of (a) talc × MAPP, (b) vPP × MAPP, (c) vPP × talc, for FS.

ANOVA was applied to estimate the error variance of the components' effects and to obtain their contributions. The results of ANOVA for the S/N ratios of tested mechanical properties are given in Table 4. The ANOVA results confirm the previous findings, as talc is the most influential factor in controlling TM and FM with contributions of 88.43% and 92.51% respectively, while vPP is the most influential factor in controlling TS and FS with contributions of 67.63% and 78.50% respectively. The influences of error on all tested mechanical properties are negligible since their contributions are less than 3%.

Table 4. Contributions for S/N ratios of tested mechanical properties obtained by ANOVA.

Source	DF	Contribution (%) of TM	Contribution (%) of TS	Contribution (%) of FM	Contribution (%) of FS
vPP	2	3.55	67.63	4.87	78.50
talc	2	88.43	23.13	92.51	8.52
MAPP	2	7.99	8.51	2.14	10.92
Error	2	0.03	0.73	0.47	2.07
Total	8				

3.2. Reduction of Dimensionality

PCA can reduce the dimensionality of a data set consisting of a number of interrelated variables while retaining as much as possible of the variation present in the original data set [25]. Principal components (PC) are selected and ordered according to their explained variation (EV) [2,25]. In this study, PCA is carried out in a generic correlation manner based on the method used in our previous work [2,19]. Compared with GRA method which used in previous literature [12], employing PCA for reducing dimensionality can preserve most of the information when avoid the impacts of related indicators. The first two PCs were selected since their accumulated explained variation (AEV) was over 99%. The complete descending ranking of PCA scores are shown in Table 5.

Table 5. PCA scores for the S/N ratios of the rPP/talc composites.

Rank	Score	Trial No
1	1.2538	9
2	0.8232	7
3	0.7361	8
4	0.6888	5
5	0.6243	3
6	0.6006	6
7	0.4840	4
8	0.3129	2
9	0.0588	1

Based upon Table 5, the main effects of the selected components on the PCA scores are plotted in Fig. 10. Since a larger PCA score indicates a better overall mechanical performance, the optimal conditions for the selected controllable factors were hereby obtained: 60 wt.% vPP, 20 wt.% talc and 5 wt.% MAPP. This is compatible with previous research, as the content of rPP should be limited for yielding a better mechanical performance [18,19].

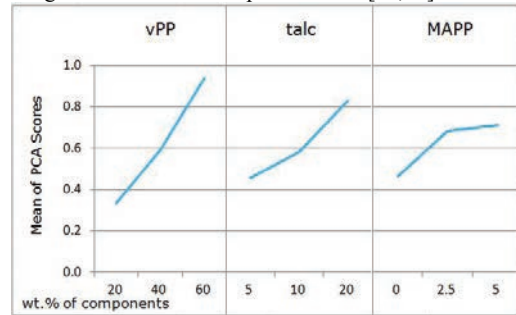


Fig. 10. Plot of S/N ratios against the selected components for PCA scores.

Table 6. ANOVA results for PCA scores.

Source	DF	SS	MS	F	p>F	Contribution (%)
vPP	2	0.55	0.28	46.91	0.02	62.31
talc	2	0.21	0.11	18.11	0.05	24.06
MAPP	2	0.11	0.05	9.27	0.10	12.30
Error	2	0.01	0.01			1.33
Total	8					

3.3. Confirmation Tests

A set of confirmation tests was carried out to verify the obtained optimal conditions. The rPP based composite with 60 wt.% vPP, 20 wt.% talc and 5 wt.% MAPP was processed and tested in the same manner as previous composites. An average of 10 specimens were tested for each property, and the experimental results and their S/N ratios were shown in Table 7. As shown in Table 7, tensile and flexural properties were enhanced at the optimal conditions when compared to the average performance of the initial designed composites. TM was the property with the most significant improvement of 42.24%, followed by 19.28% of FM and 11.14% of FS. Thus, all tested properties show an observable improvement,

and the overall mechanical performance was optimised by the proposed approach. Although the optimal formula only contains 15 wt.% rPP, the blend ratio is higher than the current usage of recycled materials in manufacturing sector (e.g. less than 1% in automobile industry) [27].

Table 7. Experimental results and S/N ratios of the optimal composite.

Property	Experimental	Δ * of Experimental	S/N Ratio	Δ * of S/N
TM	2484	42.24	67.90	4.81
TS	28.89	4.33	29.21	1.35
FM	2171	19.28	66.73	2.41
FS	43.78	11.14	32.83	2.91

* Δ is the variation expressed in % with respect to the average values presented in Table 3.

4. Conclusion

In this study, Taguchi method coupled with ANOVA were employed to optimise the formula of rPP/talc composites for improving mechanical performance. Three components, vPP, talc and MAPP were selected as the controllable factors, and a Taguchi L9 (3³) OA was used for experimental design as number of initial trials was minimised. From the S/N ratio analysis, it was found that talc is the most influential factor in controlling TM and FM, while vPP has the most significant impact on TS and FS. The interactions between components were also investigated, as vPP \times MAPP has most observable influences on TM and FM, while talc \times MAPP is the most influential interaction for TS and FS. The impacts of the three components were verified by ANOVA results. The multi-dimensional data was reduced to a single array of scores via PCA, and the optimal composition was hence obtained by analysing the PCA scores. The effectiveness of the Taguchi-ANOVA optimising approach has been successfully proven by confirmation tests on the composite of the optimal formula. Compared with current industrial practice, this approach extends the use of recycled plastics, and hence improves the sustainability of plastic manufacturing sector.

Acknowledgements

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