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Investigation on the effect of micro-fillers on the strength reinforcement of polypropylene

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ABSTRACT

This paper describes on the effect the ratio of fillers (Na10MB3A and CaCO₃) have on the tensile strength of polypropylene (PP). These fillers have been added to PP in various ratios and mixed evenly before injecting. Experimental specimens are then made by the injection process under the same conditions including mold temperature of 60° C, filled pressure of 200MPa, and injecting speed 110cm³/s using injection process. It was discovered that the tensile strength of PP increases gradually when the ratio of fillers increases. The results revealed that the tensile strength reaches its maximum value with a certain ratio of fillers, and thereafter the strength decreases as the ratio of fillers is increased. This research also indicated that the tensile strength of PP can rise by approximately 13.5% with the addition of Na10MB3A at a ratio of 3%, and increase to 21.46% with the addition of a 4% ratio of CaCO₃. They are expected that experimental samples are the usefully promising materials for automotive components. In the future work, this study will further carry out an investigation into glass filler -reinforced PP and determine the fatigue life limit of the fillers-reinforced PP material. Copyright © 2014 VBRI press.

Keywords: Reinforced polypropylene (PP); Na10MB3A; CaCO₃; tensile strength.



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Introduction

During the past few decades, the polymer industry technique has been paid a considerable attention in researching and manufacturing for automotive fields. In particularly, reinforced polymer materials using a variety of methods to increase their strength are prospective in the future. In this research, added fillers were used to reinforce polypropylene (PP) strength. They were mixed into polymers in varying ratios and injected to produce the various experimental specimens. The injection mold (Fig. 1 and 2) made experimental specimens to ISO-527 standard. The tensile process was carried out at the gate of the injection molding where materials exhibit their lowest tensile strength due to the meeting of two opposing flows. Regarding the literature review, many reports and scientific researches have written about a number of various approaches to reinforce the strength of PP material. Investigation of the effect of annealing on the mechanical behaviors of PP film was performed. As a result, the tensile strength of PP is improved. The toughness of this material could be increased by making the crystals more perfect [1]. Moreover, one experiment was conducted on carbon-fiber PP using thermal gravimetric analysis and dynamic mechanical analysis to investigate the effect of fiber length on the thermal stability and degradation of PP composite. The results showed that an increase in fiber length increases thermal stability and develops the damping behavior of PP [2].

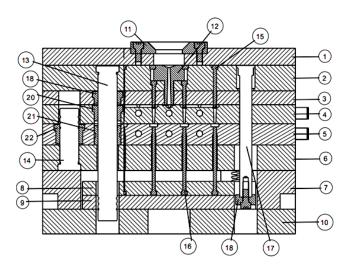


Fig. 1. Injection mold used to produce experimental specimens.

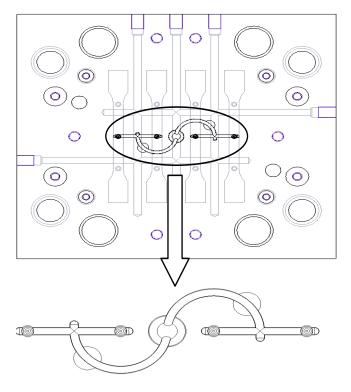


Fig. 2. General structure of channel and product arrangement.

Moreover, other study investigated reusing nonmetal waste from discarded printed circuit boards as the reinforcing fillers in PP composites. The results found that the mechanical behaviors and thermal properties of PP were enhanced, while also providing a significant way to recycle waste materials [3]. And then a report on the use of exfoliated graphics as fillers in polymers was performed [4]. In addition, the use of mineral fillers (ultrafine kaolin) in PP was carried out as an advanced technique for enhancing the mechanical properties of PP material. The results confirmed that both the impact strength and mechanical behaviors of PP were considerably improved [5]. Besides,

the creep behavior of PP was studied by adding wood flour fillers into PP material and then performing tests on residual tensile strength, creep strain and fractional deflection. The results revealed that increasing wood flour levels in composites decreased both tensile strength and creep strain [6]. Furthermore, the use of rice-husk flour and wood flour as fillers to reinforce PP was investigated. The results indicated that the tensile strength of PP produced by single screw extrusion was also found to be lower than that of PP produced using a twin screw extrusion process [7]. In particularly, reinforcing PP was studied by the addition of nanoparticles. The results showed that there was an improvement in toughness and strength using filler content [8]. The effect of added contents on the electrical and rheological properties of carbon nanotube-reinforced PP was considered. The results of experiments revealed that an increase in carbon nanotube content decreases resistivity and raises the viscosity of PP [9]. Regarding to scale fillers, the addition of nano-scale silica acting as reinforcing filler in PP was studied. The results determined that modified nanoparticles can produce more significant benefits to PP properties than conventional particulate fillers [10]. With regard to theoretical areas, the use of multifractal theory was investigated to analyze the relationships among the dispersion of CaCO₃ particles, morphology of fracture surfaces and PP tensile behavior. The results showed that multifractal theory can be effectively utilized to explain the relationship between structure and the mechanical behavior of PP-CaCO₃ composite materials [11]. In addition to the use of waste shellfish was researched and compared in shell-reinforced PP with conventional commercial calcium carbonate-reinforced polypropylene [12]. While more recently the mechanical and thermal properties of PP composites filled with modified shell waste were studied. It was noted that the introduction of modified powder could promote the heterogeneous nucleation of PP matrix [13]. In 2013, PP biocomposites reinforced with carbon nanotubes and hydroxyapatite nanorods for bone replacements were conducted [14]. And in 2014, a study on time, temperature and stress dependent behaviors of composites made from recycled polypropylene were taken into account [15]. The improvement of PP strength using various methods has been taken into account in the literature review, but most previous studies have not significantly investigated the influence of the ratio of added fillers on the mechanical properties of PP. The key objective of the present research is to investigate the effect various fillers on the strength of PP material. This paper studies and compares the effect of the ratio of fillers (Na10MB3A and CaCO₃) on the tensile strength of PP. The novelty of current study is the reinforced PP materials considered as novel materials to apply to automotive industry.

The remainder of this paper is divided into three sections. The following section presents a brief description of the experimental procedure. The next section provides the analysis of the results and the final section presents the concluding remarks. The schematic drawing of the injection mold used to produce experimental specimens is shown in **Fig. 1**. The components of the injection mold are described

in **Table 1**. The general structures of channel and product arrangement are illustrated in **Fig. 2**.

No.	Component name	No.	Component name
1	Top clamping plate	12	Ejector
2	Stationary plate	13	Guiding pin
3	Pulling plate	14	Guiding pin
4	Guiding channel plate	15	Pulling pin
5	Mold cavity plate	16	Ejection pin
6	Supporting plate	17	Retaining pin
7	Bearing	18	Washer
8	Bottom plate	19	Guiding ring
9	Holding plate	20	Guiding ring
10	Bottom clamping plate	21	Guiding ring
11	Locating ring	22	Guiding ring

Table 1. The components of the mold shown in Fig. 1.

Experimental

Material preparation

In this research, the fillers NaMB3A and CaCO₃ (purchased from VINAFAME Co., Ltd, Vietnam), respectively, were added to PP in various ratios and mixed evenly before injecting. Experimental specimens had a dimension of $160 \times 20 \times 4mm$ (**Fig. 3**). They were made by the injection process shown in **Fig. 1** under the same conditions (mold temperature of 60° C, filled pressure of 200MPa, and injecting speed 110 cm³/s) using the Shine Well injection machine (Shine Well Machinery Co., Ltd, Taiwan).

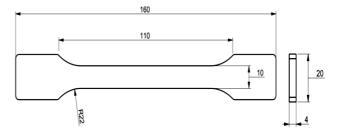


Fig. 3. Polypropylene specimen.

Mechanical tests

The experimental tensile strength testing was performed on the tensile test machine Instron 5566-CN2081 (Instron Company, Taiwan) with an ISO 527 standard as shown in **Fig. 4.** During the tensile process of the specimens, all databases were recorded on a computer linked to the tensile machine. The experimental conditions were as follows: Clamp length: 110mm, tensile speed: 50mm/min, environment humidity: 55%, room temperature: 25° C. In order to ensure a reliability of 95%, at least 16 specimens were tested using each of the filler ratios.

Results and discussion

After performing the experiments, a compressive-tensile schematic diagram for each of the experimental specimens was produced, as shown in **Fig. 5**. The figure shows that the strain-stress curves for different filler ratio samples are very similar. At the beginning stage, stress increases sharply as

the strain increases. After stress reaches a maximum value, tensile stress decreases gradually in a linear pattern. When deformation reaches a certain value, the stress decreases very sharply until the specimen is completely destroyed. The results of the 198 experiments obtained using Statgraphic software with a high reliability of $\gamma = 95\%$ are provided in **Table 2** and **3**.



Fig. 4. Tensile test machine Instron 5566-CN2081.

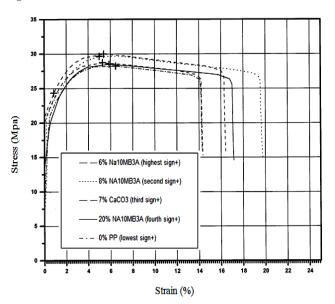


Fig. 5. Strain-stress curve diagram for PP filler-reinforced specimens.

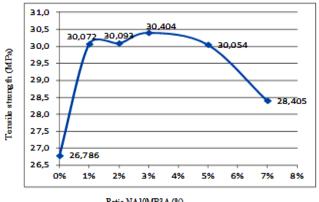
Table 2. The filler ratio of Na10MB3A (I) and tensile strength (II).

Ι	0%	2%	4%	6%	8%	20%
II	26.786	30.15	32.534	30.123	29.069	28.286

Table 3. The filler ratio of CaCO₃ (I) and tensile strength (II).

Ι	0%	1%	2%	3%	5%	7%
Π	26.786	30.072	30.093	30.404	30.054	28.405

From the results presented in **Table 2**, the relationship between the tensile strength of PP and the filler ratio (Na10MB3A) is described, as in **Fig. 6**. The figures reveal that the tensile strength of PP material increases rapidly when the ratio of the filler rises from 0% to 1%, the change in the tensile strength is only slight in the range from 1% to 5%. Tensile strength reaches a maximum value at the filler ratio of 3%. The maximum value increases approximately 13.5% compared to the tensile strength of the sample without fillers. When the ratio of the Na10MB3A filler increases to more than 5%, tensile strength decreases rapidly.



Ratio NA10MB3A (%)

Fig. 6. Schematic diagram of the relationship between the ratio of Na10MB3A (%) and the tensile strength of PP.

Processing the experimental databases provides the approximate quadratic polynomial experimental equation given in **Fig. 7**. This shows the relationship between the tensile strength and the ratio of added Na10MB3A as follows:

$$Y = -2402.4X^2 + 178.8X + 27.462 \tag{1}$$

where Y is the tensile strength (*MPa*), X is the ratio of NaMB3A (%).

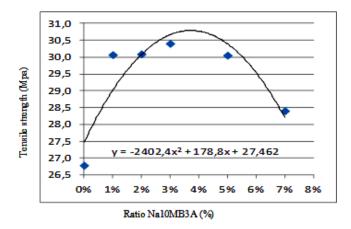


Fig. 7. The quadratic polynomial interpolation curve presents the relationship between the tensile strength PP (MPa) and the ratio of added Na10MB3A (%).

The relationship between tensile strength and the filler ratio of $CaCO_3$ is shown in **Fig. 8**. At the ratio ranges from

0% to 4%, the tensile strength of PP material increases rapidly. As the ratio of added CaCO₃ increases to between 4% and 8%, the tensile strength decreases rapidly. When the filler ratio ranges from 8% to 20%, the change in tensile strength is slight. Tensile strength reaches a maximum value and reinforces the PP by approximately 21.46% as the ratio of CaCO₃ reaches about 4%.

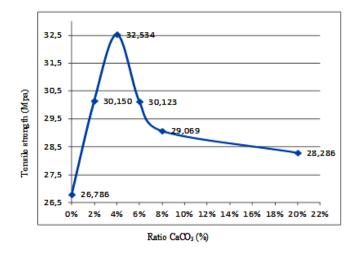
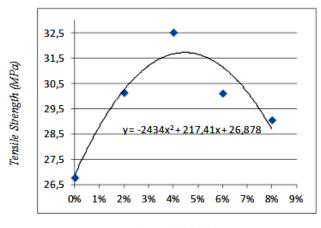


Fig. 8. Schematic diagram showing the relationship between the ratio of added CaCO₃ (%) and the tensile strength of PP (MPa).

The quadratic polynomial equation which follows describes the tensile strength of PP material and the ratio of CaCO₃ in the range of 0% to 8%, as shown in **Fig. 9**:

$$Y = -2434X^2 + 217.41X + 26.878 \tag{2}$$

where *Y* is the tensile strength (*MPa*), *X* is the ratio of $CaCO_3$ (%).

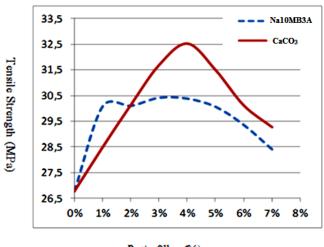


Ratio CaCO₃ (%)

Fig. 9. The quadratic polynomial interpolation curve gives the relationship between the tensile strength of PP (MPa) and the ratio of added $CaCO_3$ (%).

A compilation of **Fig. 6** and **8** with added fillers in the range of 0% to 7% is shown in **Fig. 10**. It shows that when the filler ratio ranges from 0% to 2% the tensile strength of PP material is reinforced more by Na10MB3A than by CaCO₃. On the contrary, when the ratio of added filler is

over 2%, the tensile strength of PP material is reinforced more by $CaCO_3$ than by Na10MB3A.



Ratio filler (%)

Fig. 10. Comparison of two curves in Figs. 6 & 8 in the range from 0% to 7%.

Conclusion

This research has presented the use of micro-fillers (Na10MB3A, CaCO₃) to reinforce polypropylene. The strength of PP material may reach a maximum value when a certain ratio of filler was added. When the filler ratio surpasses this value, the strength decreased significantly due to the material becoming brittle. This research revealed that the tensile strength of PP could rise by approximately 13.5% when Na10MB3A was added in the ratio of 3%, and increase by 21.46% when the ratio of added CaCO₃ was 4%. This paper described fracture mechanics at the crack surface of filler-reinforced polymer, including the debonding of polymer on the filler surface, the deformation process and the destruction of the polymer substrate. The filler-reinforced polymer process was based on the external impacting energy absorption process relative to stress concentration surrounding the filler, the debonding that happened between filler particles and the sliding stress ranges during the deformation process.

Achievements of this research are that the improved novel material PP is made, which has not studied before. It is hoped that the high strength material PP able to be utilized for the automotive components. In the future, this study will continuously investigate into reinforcement of the strength of PP using glass-filler and determine the fatigue limit of the fillers-reinforced PP material.

Reference

- Balas, D.F.; Maspoch, M.L.; Martinez, A.B.; Santana, O.O.; *Polymer*, 2001, 42, 1697-1705.
 DOI: <u>10.1016/S0032-3861(00)00487-0</u>
- Rezaei, F.; Yunus, R.; Ibrahim, N. A.; *Mater. Desig.* 2009, 30, 260. DOI: 10.1016/j.matdes.2008.05.005
- Zheng, Y.; Shen, Z.; Cai, C.; Ma, S.; Xing, Y.; J. Hazard. Mater. 2009, 163, 600.
 DOI: 10.1016/j.jhazmat.2008.07.008
- Debelak, B.; Lafdi, K.; *Carbon*, 2007, 45, 1727.
 DOI: 10.1016/j.carbon.2007.05.010

- Mareri, P.; Bastide, S.; Binda, N.; Crespy, A.; Composites Sci. Technol. 1998, 58, 747.
 DOI: 10.1016/S0266-3538(97)00156-5
- Lee, S.Y.; Yang, H.S.; Kim, H.J.; Jeong, C.S.; Lim, B.S.; Lee, J.N.; *Composite Structures*, 2004, 65, 459. DOI: 10.1016/j.compstruct.2003.12.007
- Yang, H.S.; Wolcott, M. P.; Kim, H.S.; Kim, S.; Kim, H.J; *Polym. Testing*, **2006**, 25, 668.
- **DOI:** <u>10.1016/j.polymertesting.2006.03.013</u>
 8. Wu, C.L.; Zhang, M.Q.; Rong, M.Z.; Friedrich, K.; *Composites Sci. Technol.*, **2002**, *62*, 1327.
- DOI: 10.1016/S0266-3538(02)00079-9
 Seo, M.K.; Park, S. J.; *Chem. Phys. Lett.* 2004, *395*, 44.
 DOI: 10.3144/expresspolymlett.2012.52
- Rong, M.Z.; Zhang, M.Q.; Zheng, Y.X.; Zeng, H.M.; Walter, R.; Friedrich, K.; *Polymer*, **2001**, *42*, 167.
 DOI: 10.1016/S0032-3861(00)00325-6
- Pérez, E.; Bernal, C.; Piacquadio, M.; *Appl. Surf. Sci.*, **2012**, 258, 8940.

DOI: <u>10.1016/j.apsusc.2012.05.124</u>

- Li, H.Y.; Tan, Y.Q.; Zhang, L.; Zhang, Y.X.; Song, Y.H.; Ye, Y.; Xia, M.S.; J. Hazard. Mater. 2012, 217, 256.
 DOI: <u>10.1016/j.jhazmat.2012.03.028</u>
- Yao, Z.T.; Chen, T.; Li, H.Y.; Xia, M.S.; Ye, Y.; Zheng, H.; J. Hazard. Mater. 2013, 262, 212.
- DOI: 10.1016/j.jhazmat.2013.08.062
 14. Liao, C.Z.; Li, K.; Wong, H.M.; Tong, W.Y.; Yeung, K.W.K.; Tjong, S.C.; *Mater. Sci. Eng. C*, 2013, *33*, 1380.
 DOI: 10.1016/j.msec.2012.12.039
- Homkhiew, C.; Ratanawilai, T.; Thongruang, W.; Construction Building Mater. 2014, 66, 98.
 DOI: 10.1016/j.conbuildmat.2014.05.048

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