Effect of Silane Coupling Agent and Compatibilizer on Properties of Short Rossells Fiber/Poly(propylene) Composites

Kasama Jarukumjorn, Wimonlak Sutapun, Yupaporn Ruksakulpiwat, Jongrak Kluengsamrong*

Summary: Rossells fiber reinforced polypropylene composites were prepared by melt mixing. The fiber content was 20 wt%. Octadecyltrimethoxysilane (OTMS) and maleic anhydride grafted polypropylene (MAPP) were used to improve the adhesion between poly(propylene) (PP) and the fiber. The mechanical, rheological, and morphological properties, and heat distortion temperature (HDT) of the composites were investigated. Tensile strength, impact strength, flexural strength and HDT of MAPP modified PP composites increased with an increase in MAPP content. However, no remarkable effect of MAPP content on the Young's modulus of the composites was found. OTMS resulted in small decreases of tensile strength and Young's modulus, and increase in impact strength. Scanning electron micrographs revealed that MAPP enhanced surface adhesion between the fiber surface and PP matrix.

Keywords: MAPP; OTMS; rossells fiber-PP composites

Introduction

The use of natural fibers to reinforce polymers has received great attention due to environmental concern and government regulations. Natural fibers have many advantages including low density, low cost, nonabrasive nature, and biodegradability. The main disadvantage of the natural fibers is their hydrophilic nature therefore; they are not well compatible with hydrophobic polymeric matrix. As a result, the mechanical properties of polyolefin composites may not be improved by natural fiber reinforcement. Silane coupling agents and maleic anhydride grafted polypropylene (MAPP) are widely used to improve interfacial adhesion between the natural fibers and non-polar thermoplastic matrix. Herrera-Franco and Aguilar-Vega[1] studied the effect of vinyltris (2-methoxyethoxy) silane on mechanical properties of Henequen-LDPE composites and found that the small increment in the properties was attributed to an improvement in the interfacial adhesion between the natural fiber and the polymer matrix. A significant enhancement of mechanical properties of kenaf-PP composites, especially notched impact strength after modification of the fiber with amino-ethyl amino-propyl triethoxysilane was reported by Karnani et al.[2] In addition, Cantero et al.[3] reported that vinyltrimethoxy silane (VTMO) treatment gave a slight improvement on flexural strength of flax fiber-PP composites. MAPP is a very effective compatibilizer to improve the interfacial adhesion between natural fibers and non-polar polymeric matrices.[4] Both chemical and physical interactions have been observed at the interfaces between MAPP and hydroxyl groups of natural fibers. While the PP chain of MAPP diffuses into the PP matrix to form the physical interaction (entanglement). Bos et al.[5] found that MAPP improved mechanical properties and fiber-matrix

School of Polymer Engineering, Institute of Engineering, Suranaree University of Technology, Nakorn Ratchasima, 30000, Thailand
E-mail: kasama@g.sut.ac.th

adhesion of short flax fiber-PP composites. An increase of mechanical properties of sisal-PP composites was observed when MAPP was used.[2,6] Addition of MAPP in flax-PP composites increased mechanical properties and decreased the water uptake rate.[7] Rana et al.[8] found that maleated polypropylene (MAPP) significantly improved the mechanical properties of jute fiber-PP composites.

This study examined the effect of fiber treatment with octadecyltrimethoxysilane (OTMS) and the addition of MAPP, as a compatibilizer, on the mechanical, rheological and morphological properties, and HDT of rossells-PP composites.

Experimental Part

Materials

A commercial grade of isotactic PP (700J) was supplied by Thai Polypropylene Co., Ltd. Maleic anhydride grafted PP (Fusabond® P MZ 109D, DuPont) was used as a compatibilizer and octadecyltrimethoxysilane (OTMS, Aldrich) as a coupling agent. Rossells fibers were obtained from NEP Realty and Industry Public Co., Ltd. Tensile strength and tensile modulus of the fiber were 473.4 ± 120.2 MPa and 40.6 ± 9.9 GPa, respectively.

Fiber Preparation

Rossells fibers were washed by water, dried in an oven at 60 °C, and cut into a short fiber. The average length of the fiber was 2.65 mm and the aspect ratio of the fiber was 33.68. After that the short fibers were boiled with a methanol and benzene mixture (1:1) to remove waxes and low molecular weight species and then cleaned with 2 wt% NaOH to eliminate hemicellulose. These fibers were called cleaned fibers. In order to modify rossells fibers with OTMS, the cleaned fibers were immersed in 2 wt% solution of OTMS (pH 3.5) for 3 hrs. The OTMS treated fibers were subsequently washed and dried in an oven at 60 °C overnight.[9]

Composite Preparation

The rossells-PP composites were prepared using an internal mixer (Hakke Rheomix Polylab). The rotor speed was 50 rpm and the mixing temperature was 170 °C. The fiber content used was 20 wt%. The total mixing time was 13 min. The MAPP contents were 1, 2, 4, 6, 8, and 10 phr. The test specimens were molded by an injection molding (Chuan Lih Fa, CLF 80T). Composition of composites and their designation are shown in Table 1.

Characterization

Tensile properties of the composites were tested according to ASTM D638 using an Instron universal testing machine (model 5565) with a load cell of 5 kN and a crosshead speed of 10 mm/min. Izod impact tests were performed according to ASTM D256. Flexural properties were measured in accordance to ASTM D5943 with a crosshead speed of 15 mm/min and a span length of 56 mm. Melt flow index (MFI) was measured at 180 °C and 2.16 kg load. Heat

Table 1.
Composition and designation of materials used.

Designation	PP (wt%)	Unmodified rossells fiber (wt%)	OTMS treated rossells fiber (wt%)	MAPP (phr)
PP	100	–	–	–
Unmodified	80	20	–	–
OTMS	80	–	20	–
MAPP_1 phr	80	20	–	1
MAPP_2 phr	80	20	–	2
MAPP_4 phr	80	20	–	4
MAPP_6 phr	80	20	–	6
MAPP_8 phr	80	20	–	8
MAPP_10 phr	80	20	–	10

distortion temperature (HDT) was determined following ASTM D648. Surface morphology was examined using scanning electron microscope (SEM JEOL model JSM6400) at 10 keV. For the morphology investigation, the specimens were fractured in liquid nitrogen.

Results and Discussion

Mechanical Properties

Tensile strength and Young's modulus of OTMS treated rossells-PP composite were lower than that of unmodified rossells-PP composite as shown in Figure 1 and 2. This might be because the long and flexible octadecyl group of OTMS enhanced flexibility at interfacial layer between the fiber and matrix. The tensile strain of the composites, as shown in Figure 2, was increased as well when the fibers were treated with OTMS. However, Young's modulus of the OTMS-treated rossells-PP composites was lower than that of the MAPP modified PP composites. In the case of MAPP, the tensile strength of the composites increased significantly with increasing MAPP content up to 2 phr. Young's modulus of MAPP modified rossells-PP composites was not significantly different from that of the unmodified composite. Impact strength of the composites tended to increase with MAPP contents. The small reduction in tensile strain of the composites with addition of MAPP, as shown in Figure 2, indicated that tensile strain cannot be achieved by the improvement of the fiber-matrix interaction. Flexural properties of the composites are shown in Figure 3. Flexural strength and modulus also increased with an increase of MAPP contents. The enhancement in the mechanical properties of the MAPP modified composites indicated that the compatibility between PP and rossells fibers was improved. MAPP enhanced the adhesion between non-polar PP and polar rossells fibers resulting in an improvement in the mechanical properties. The formation of covalent linkages between maleic anhydride and hydroxyl groups of cellulose was reported by Hedenberg and Gatenholm [10] through IR and ESCA analysis.

Rheological Properties

Melt flow index (MFI) of PP, unmodified and modified Rossells-PP composites was

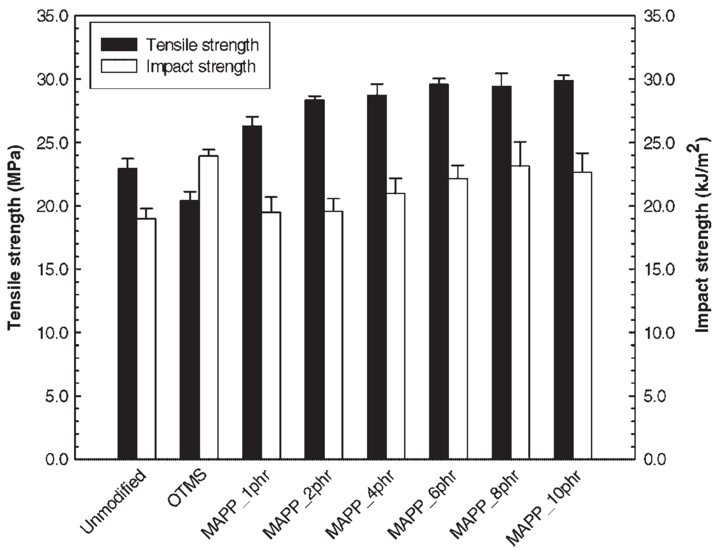

Figure 1.
Effect of OTMS treated rossells fibers and the addition of compatibilizer on tensile and impact strength of rossells-PP composites.

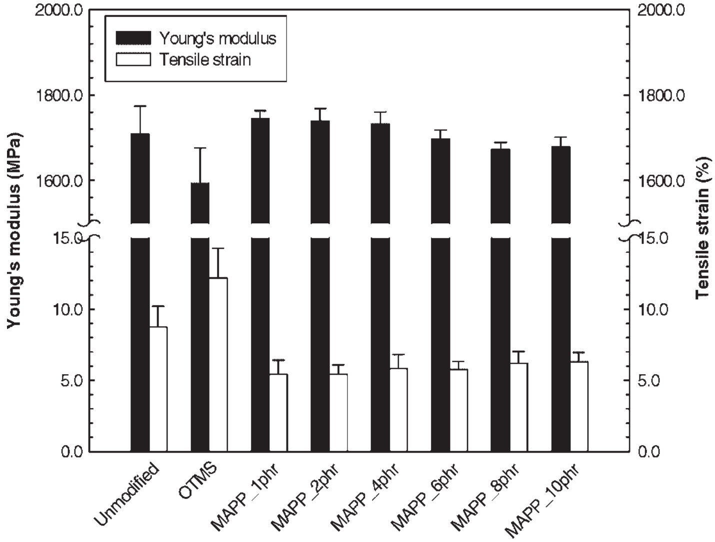

Figure 2.
Effect of OTMS treated rossells fibers and the addition of compatibilizer on Young's modulus and tensile strain of rossells-PP composites.

shown in Table 2. MFI decreased when the fibers were added. MFI of MAPP modified rossells-PP composites was not remarkable different from unmodified and OTMS treated rossells-PP composite. Schemenauer et al.[11] had reported that no significant effect of MAPP on viscosity of jute fibers-PP composites as well.

Heat Distortion Temperature

HDT of PP and PP composites were shown in Table 3. HDT of PP increased from 74.5 °C up to 128 °C when rossells fiber was added. As expected, the presence of the fibers substantially improved the HDT of PP. Whereas HDT of OTMS treated rossells-PP composite was about 6 °C lower

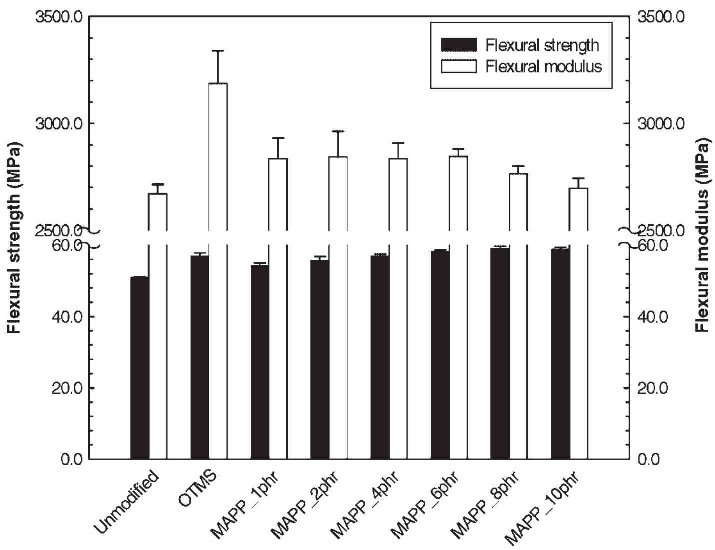

Figure 3.
Effect of OTMS treated rossells fibers and the addition of compatibilizer on flexural strength and modulus of rossells-PP composites.

Table 2.
Melt flow index of PP and rossells-PP composites.

Materials	MFI (g/10 min.)
PP	4.97 ± 0.01
Unmodified	2.57 ± 0.01
OTMS	2.89 ± 0.05
MAPP_1phr	2.37 ± 0.04
MAPP_2phr	2.37 ± 0.02
MAPP_4phr	2.19 ± 0.03
MAPP_6phr	2.19 ± 0.04
MAPP_8phr	2.10 ± 0.05
MAPP_10phr	2.03 ± 0.07

Table 3.
Heat distortion temperature of PP and rossells-PP composites.

Materials	HDT (°C)
PP	74.5 ± 2.12
Unmodified	128.0 ± 1.00
OTMS	122.3 ± 1.00
MAPP_1phr	132.5 ± 1.46
MAPP_2phr	135.3 ± 3.32
MAPP_4phr	134.4 ± 2.85
MAPP_6phr	133.0 ± 2.93
MAPP_8phr	133.7 ± 3.42
MAPP_10phr	132.8 ± 3.50

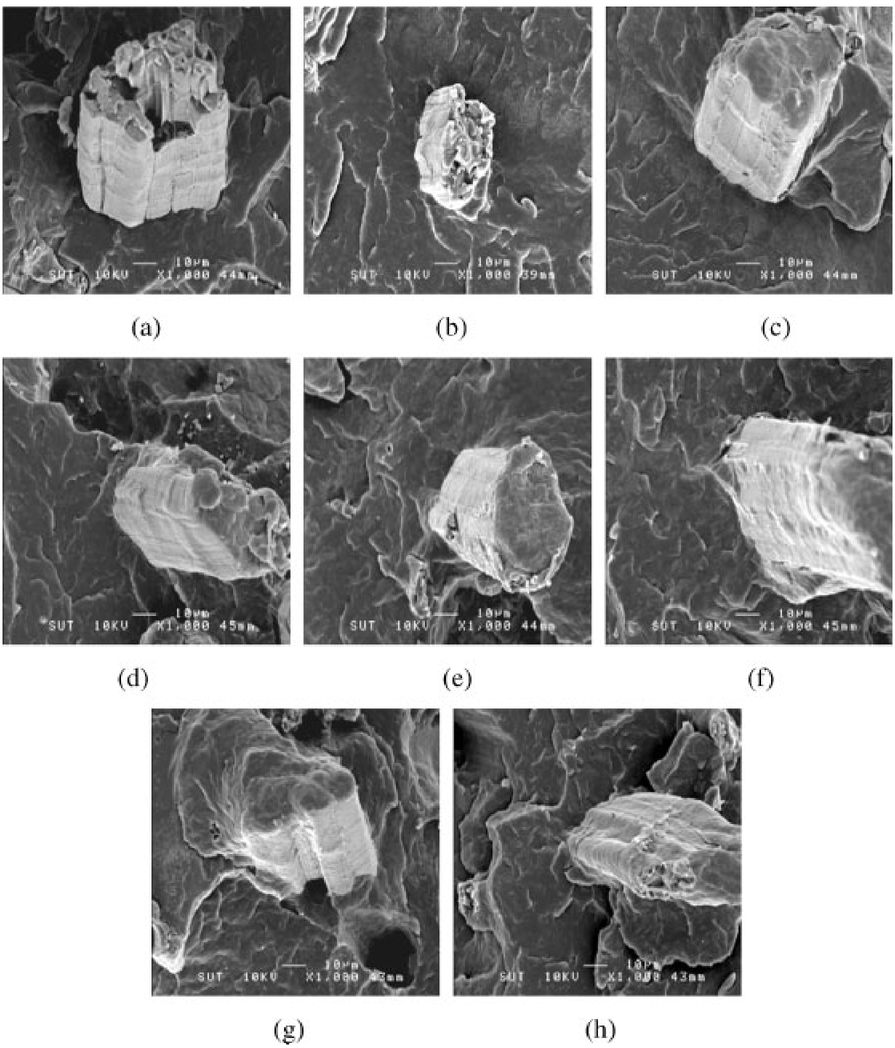

Figure 4.
Surface morphology of PP composites; (a) Unmodified, (b) OTMS, (c) MAPP_1phr, (d) MAPP_2phr, (e) MAPP_4phr, (f) MAPP_6phr, (g) MAPP_8phr and (h) MAPP_10phr.

than that of the unmodified rossells-PP composite. This might be attributed to the presence of OTMS. For the MAPP modified rossells-PP composites, HDT was higher than that of the unmodified composite. However, adding more compatibilizer from 1 phr up to 10 phr, no remarkable difference on the HDT of the composites was observed.

Morphological Properties

Fracture surface of the rossells-PP composites as shown in Figure 4 (a)–(h) revealed that addition of the compatibilizer enhanced the surface adhesion between fiber and PP. Since the gap between the fiber surface and PP matrix was reduced and the fiber surface was more coated with the PP matrix. However, better surface adhesion was found in MAPP modified composites compared to that of OTMS treated fiber-PP composite. This was well corresponding to the mechanical properties previously.

Conclusions

When the rossells fibers were modified with OTMS the Young's modulus and tensile strength were not much lower than that of the unmodified composite. However, the impact strength was higher than that of the unmodified composite. The tensile, flexural, and impact strength of the rossells-PP composites can be improved with the addition of MAPP due to the improvement of fiber-matrix interfacial adhesion. MAPP had no remarkable effect on Young's modulus of the composites. The results indicated that the composites based on addition of MAPP had superior mechanical properties compared to composite which the fibers were treated with OTMS.

Acknowledgements: The authors would like to thank Chemical Innovation, Co., Ltd. for providing Fusabond® MZ 109D and NEP Realty and Industry Public Co., Ltd., for donating rossells fibers. This research is financially supported by Suranaree University of Technology and National Metal and Materials Technology Center (MTEC), Thailand.

[1] P. J. Herrera-Franco, M. De. J. Aguilar-Vega, *J. Appl. Polym. Sci.* **1997**, *65*, 197.
[2] R. Karnani, M. Krishnan, R. Narayan, *Polym. Eng. Sci.* **1997**, *37*, 476.
[3] G. Cantero, A. Arbelaiz, R. Llano-Ponte, I. Mondragon, *Comp. Sci. Tech.* **2003**, *63*, 1247.
[4] T. T. L. Doan, S. L. Gao, E. Madar, *Comp. Sci Tech.* **2006**, *66*, 952.
[5] H. L. Bos, J. Mussig, M. J. A. Van den Oever, *Composites: Part A.* **2006**, *37*, 1591.
[6] K. L. Fung, R. K. Y. Li, S. C. Tjong, *J. Appl. Polym. Sci.* **2002**, *85*, 169.
[7] A. Arbelaiz, A. B. Fernandez, J. A. Ramos, A. Retegi, R. Llano-Ponte, I. Mondragon, *Comp. Sci. Tech.* **2005**, *65*, 1582.
[8] A. K. Rana, A. Mandal, B. C. Mitra, R. Jacobson, R. Rowell, A. N. Banerjee, *J. Appl. Polym. Sci.* **1998**, *69*, 329.
[9] W. Sutapun, J. Kluengsamrong, Y. Ruksakulpiwat, K. Jarukumjorn, N. Supakarn, P. Chumsamrong, *The 30th Congress on Science and Technology of Thailand*, **2004**, 175.
[10] P. Hedenberg, P. Gatenholm, *J. Appl. Polym. Sci.* **1995**, *56*, 641.
[11] J. J. Schemenauer, T. A. Osswald, A. R. Sanadi, D. F. Caulfield, *SPEANTEC.* **2000**, 2206.

