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The Effect of Aminopropyltrimethoxysilane (Aps-Silane) on Polypropylene(PP)/Recycled Acrylonitrile Butadiene Rubber(NBRr)/Palm Pressed Fiber (PPF) Composite



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ARTICLE INFO	ABSTRACT
Article history: Received 3 April 2018 Received in revised form 3 August 2018 Accepted 20 September 2018 Available online 26 September 2018	Polypropylene (PP)/ recycled acrylonitrile butadiene rubber (NBRr)/ palm pressed fibre (PPF) are composites were fabricated and studied. Waste filler PPF of 63 - 300 μ m was utilized in this work. The thermal mixing technique is used in this research. All samples were prepared on a heated two roll mill at 180 °C for 9 minutes. Prepared samples were moulded in a hot press machine at 180 °C and cut into dumbell for testing. PPF filler loading were varied from 0 to 30 w/w for investigation. Coupling agent γ -APS was also evaluated on this composites. The samples were analyzed for mechanical and morphological properties. Tensile properties of PP/NBRr/PPF composites was found to be decreased with increasing PPF filler loading. The treatment of γ -APS on PPF filler showed better mechanical properties of the composites.The adhesion of PPF with PP/NBRr matrices were found to be improved. This may due to esterification bonding of the filler with PP in the presence of γ -APS. This finding is well supported by the morphological studies of SEM micrographs. Dispersion of filler onto PP/NBRr matrices were also found to be enhanced with less pull outs.
<i>Keywords:</i> Polypropylene, NBRr, palm pressed fibre, aminopropyltrimethoxysilane	Copyright © 2018 PENERBIT AKADEMIA BARU - All rights reserved

1. Introduction

Natural fibers can be utilized as fillers or reinforcement in a polymer composite. The usage of natural fibers has gained a lot of attention in recent years. Many researches used on the reinforcing materials on application of natural fillers like cellulose fibers, sago, rubber wood powder, sisal, sugarcane, short silk fibre, oil palm, banana fibers, cotton stalk, empty fruit bunch, jute fibre, rice husk ash, hemp, sisal, jute, and kenaf [1-5]. In comparison with man-made fibers such as glass fibers, natural fibers have more benefits due to its biodegradable, recyclable, renewable, cheaper in cost,

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lower in density, properties of specific tensile which are equivalent, lower in energy usage and possesses reduced health threat [6-9]. Hence natural fillers serves as a better substitute in comparison with other man-made fibres substitute [10-12].

In this research, the fabrication of Polypropylene (PP), Recycled Acrylonitrile Butadiene Rubber (NBRr) and Palm Pressed Fibre (PPF) composite was experimented based on mechanical properties. Lignocellulose materials such as PPF filler are hydrophilic and polar caused by the hydroxyl groups that are present in the cellulose. The NBRr matrix is also found a polar substance, however inherit large chemical energy differences [13,14]. Hence, these three components exhibit different physical and chemical characteristics among them due to high incompatibility. Consequently it leads to incompatibility or immiscibility of the filler and matrices. Large chemical energy differences from the non-polar matrix with polar fillers are also partially responsible for these findings. The main aim of this study is to reduce the incompatibility of these materials and to fabricate miscible composites with the help of appropriate coupling agent such as γ -APS.

2. Material and Methods

Materials used for the preparation of PP/NBRr/PPF composites are as in Table 1. Palm pressed fibre powder was grounded using pulverizing machine (Rong Tsong Precision Technology Co. Product Id: RT-34) with the speed of 2850 rpm, sieved at $63 - 150 \mu m$ in particle size. Next, the proposed materials was dried at 110 °C for 24 hours in an oven to produce palm pressed fiber powder of homogeneous fractions.

Material	Description	Source				
Polypropylene (PP)	Code: 331 MFI: 14g/10 min at 230 °C	Titan Pro Polymers (M)				
	Density: 0.9g/cm ³	Sdn.Bhd. Johor, Malaysia				
Recycled Acrylonitrile Butadiene	Content: 33% of Acrylonitrile Density:	Juara One Resources Sdn. Bhd.				
Rubber (NBRr)	1.015 g/cm ³	Penang, Malaysia				
Palm Pressed Fiber (PPF)	Cellulose: 35%	Solid Orient Holdings Sdn.Bhd.				
	Hemicellulose: 25%	Kedah				
	Lignin: 20%					
	Ash: 17%					
	Density:1.4702g/cm ³					
	Size: 63-150 μm					
Silane Coupling Agent	γ-aminopropyltrimethoxysilane (APS)	Alfa Aesar (M) Sdn Bhd				

Table 1

Materials specification and description

Treatment reaction was carried out in a mixture of water/ethanol (40/60 volume). Firstly, a 3g of y-aminopropyltrimethoxysilane was produced into 1000ml of the mixture water/ethanol and allowed to stand for 1 hour. The pH of the solution was maintained at 4 point with the addition of acetic acid. Then, 10 g of palm pressed fiber powder was added into the solutions and it was continuously stirred for 1.5 hours. The filler was filtered, dried in the air and then in the oven at 80 °C for 24 hours. Polypropylene (PP) was mixed with recycled Acrylonitrile Rubber (NBRr) and palm pressed fiber (PPF) at various loading (0, 5, 10, 15, 20, 25, 30) w%. Palm pressed fiber powder was dried at 110 °C for 24 hours in an oven before mixing. A constant amount 70 g of PP and 30 g of NBRr was used. Table 2 shows the formulation of PP/NBRr/PPF composites.



Composition					PP/	NBRr/P	PF com	nosites					
Materials	\$1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13
PP	70	70	70	70	70	70	70	70	70	70	70	70	70
NBRr	30	30	30	30	30	30	30	30	30	30	30	30	30
Pure PPF γ-APS Treated PPF	- -	5 -	10 -	15 -	20 -	25 -	30 -	- 5	- 10	- 15	- 20	- 25	- 30

Table 2 Formulation for PP/NBRr/PPF composite

PP/NBRr/PPF composites were mixed using a heated two roll mill at 180 °C with the rotor speed of 15 rpm. In the mixing process, PP was heated at 180 °C. After the 2 minute, NBRr was added and mixed to make PP/NBRr matrices became homogeneous. PPF was added to the matrix on the 5th minute, the mixing was continues and intense for homogenous mixing between the filler and the matrices. The compound was mixed on the heated two roll mill was removed at 9th minute and was kept in a desiccator for 24 hours to remove the moisture. The mixed compounds were compressed in electric heated hydraulic press model GT 7014 A at 180° C. Pre-heating until 6 minute and compression in 4 minutes at 1000 psi. A cooling duration of 2 minutes was also carried out. The 1 mm thickness sheet was cut into dumbbell shapes using Wallace die cutter S6/1/6.A.

Mechanical tests was conducted using ASTM D412 using Instron tensile machine model No 3366. A dumbbell specimens in 1 mm thickness were utilized for tensile test. Tensile strength, elongation at break and Young's modulus were measured at a cross head speed of 5 mm/min. Five specimens were used for each test and the average data with corresponding standard deviation was reported. Besides, a scanning Electron Microscopy (SEM) micrographs were taken at various magnifications for fracture surface. The samples were coated with a thin layer of gold to avoid electrical charging during examinations.

3. Results and Discussion

Tensile strength of the PP/NBRr/PPF composites was steady decreases with the increasing amount of PPF filler. This may due to poor stress transfer from the filler to the matrix in higher filler loading. Figure 1 (a) displays the tensile strength of the PP/NBRr/PPF composites with and without γ -APS treatment on the PPF filler. In the presence of γ -APS treated filler, higher tensile properties were recorded. The even distribution of PPF filler and better bonding with PP/NBRr matrices may due to the esterification bonding in the composite. This findings is similar with Tajvidi *et al.*, [15] that the natural fiber polypropylene composites whereby stress-strain of the composite is highly independent to the filler matrix interface.

Figure 1 (b) shows the effect of PPF filler loading on elongation at the break of PP/NBRr/PPF composite. Both untreated and treated γ -APS composites exhibits decreasing trend with the increasing filler loading. This may due to the brittle behaviour of PPF filler in the composite besides poor mixing of the compound during processing. The agglomeration may also result due to strong hydrogen bond between the fillers at higher filler loading. This finding was supported by Ragunathan *et al.*, [16] in PP/NBRr/RHP composite. The elongation at break of γ -APS treated was found to be further reduces. This may due to better enhanced matrix-filler bonding in the presence of γ -APS. The γ -APS have improved the adhesion of the PPF filler with the PP/ NBRr matrices. This may due to



restricted mobility of polymer chain due to entanglement of the anhydride group with the PP plane [16]. According to Razavi-Nouri *et al.*, [17], it has been reported that the composite has undergone stiffening process which lead to higher rigidity.

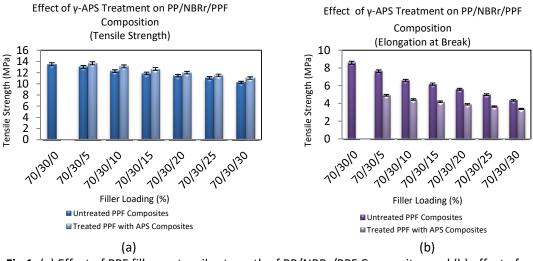


Fig.1. (a) Effect of PPF filler on tensile strength of PP/NBRr/PPF Composites and (b) effect of PPF filler on elongation at break of PP/NBRr/PPF Composites

The effect of PPF filler loading on Young's modulus of PP/NBRr/PPF composite was shown in Figure 2. Both untreated and treated γ -APS composites exhibits increasing trend with the increasing filler loading. The treatment of γ -APS on the composite shows even higher Young's modulus than the untreated composite. These findings are agreed with the elongation of break decrease trend in Figure 1 (b). The enhanced adhesion and dispersion of filler in the presence of esterification bond may be responsible for this condition. According to the Ismail and Mega [18], usage of γ -APS as a coupling agent provides the composite with good compatibility in hydrophobic matrices of PP/NBRr.

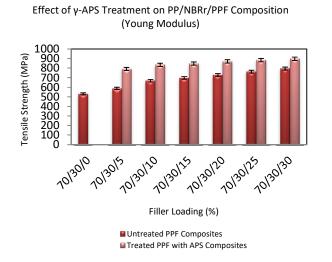
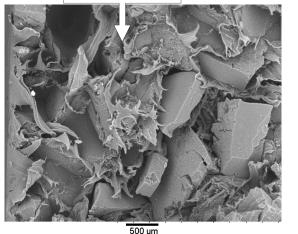


Fig. 2. Effect of PPF filler Young's Modulus of PP/NBRr/PPF Composites

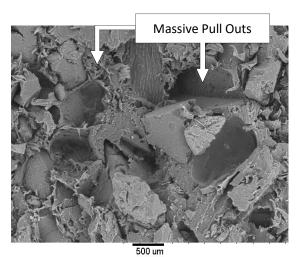


Figure 3 and Figure 4 shows the SEM micrograph of the untreated (PP/NBRr/PPF) composites and treated (PP/NBRr/PPF) composites with γ -APS. Based on the SEM observation, Figure 3 (a-c) shows that the untreated PP/NBRr/PPF composites exhibits massive pulls out and PPF detachment sites. This is due to poor bonding of PPF filler and PP/NBRr matrices. According to the Naderi *et al.*, [19], it is because of the polymers incompatibility which is the differences in polarity of the matrices and filler. However, in Figure 4 (a-c), with γ -APS treated PP/NBRr/PPF composites shows the better adhesion and stronger bonding with the PPF filler. Thus, there are less pull outs and visible good attachment of PP/NBRr matrices and PPF filler. The coupling agents, γ -APS proved to improved the bonding of the PPF filler and PP/NBRr matrices.

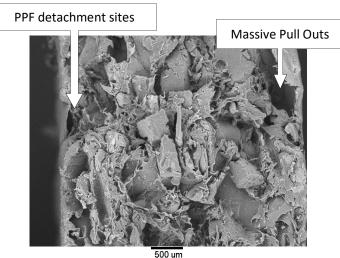
Massive Pull Outs



(a) 70/30/5



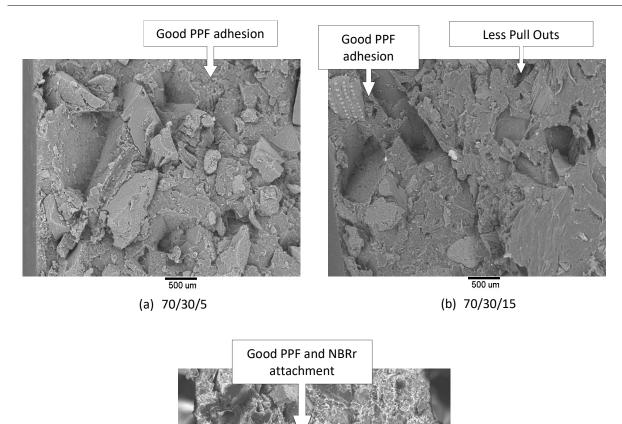
(b) 70/30/15



(c) 70/30/30

Fig. 3. SEM of tensile fracture surfaces of PP/NBRr/PPF composites and PP/NBRr/PPF treated with γ -APS composite at different PPF filler content





500 um (c) 70/30/30

Fig. 4. SEM of tensile fracture surfaces of PP/NBRr/PPF treated with γ -APS composite at different PPF filler content

4. Conclusion

The present study was designed to determine the effect of Aminopropyltrimethoxysilane (Aps Silane) On Polypropylene (PP)/Recycled Acrylonitrile Butadiene Rubber (NBRr)/Palm Pressed Fiber (PPF) Composite. Based on the experiment, a few important conclusions drawn from this work include

- i. By nature, PPF filler is hydrophilic. In addition to that, further studies were conducted on PPF filler which was SEM through morphological studies. From the SEM observation, the PPF filler does not have regular shape
- ii. Filler loading of 0-30 w% was utilized. As the filler loading increases, the tensile strength and elongation at break decreases. This is due to the increase in stiffness of the sample which can be supported with the finding with the increase in Young's modulus. In addition to that, the



possibility of agglomeration of the filler occurred with the increasing filler loading may due to the reason of the decrease in tensile strength.

iii. Incorporation of γ-APS as the coupling agent proves to improve the filler's dispersion in the matrix. This was clearly seen in tensile properties where the tensile strength, Young's modulus and elongation at break seem to be better. In addition to that, SEM observation supports the findings in tensile testing. PP/NBRr/PPF composites with the presence of γ-APS shows better adhesion, good attachment between the PPF filler and PP/NBRr matrices compared to the PP/NBRr/PPF composites that do not have the coupling agent. Besides that, the SEM for γ-APS treated composites showed less pull outs

Further research might explore a few others analysis in order to support the findings. More information result such as kinetic studies, thermal and water absorption on proposed materials would help us to establish a greater degree of properties on this matter.

References

- [1] Sgriccia, N., M. C. Hawley, and M. Misra. "Characterization of natural fiber surfaces and natural fiber composites." *Composites Part A: Applied Science and Manufacturing* 39, no. 10 (2008): 1632-1637.
- [2] Tajvidi, Mehdi, Robert H. Falk, and John C. Hermanson. "Effect of natural fibers on thermal and mechanical properties of natural fiber polypropylene composites studied by dynamic mechanical analysis." *Journal of Applied Polymer Science*101, no. 6 (2006): 4341-4349.
- [3] Satyanarayana, Kestur G., Gregorio GC Arizaga, and Fernando Wypych. "Biodegradable composites based on lignocellulosic fibers—An overview." *Progress in polymer science* 34, no. 9 (2009): 982-1021.
- [4] Pereira, Paulo Henrique Fernandes, Kelly Cristina C. Carvalho Benini, Cintia Yumi Watashi, Herman Jacobus Cornelis Voorwald, and Maria Odila Hilario Cioffi. "Characterization of high density polyethylene (HDPE) reinforced with banana peel fibers." *BioResources* 8, no. 2 (2013): 2351-2365.
- [5] M. Zainal, M. Z. Aihsan, W. A. Mustafa, and R. Santiagoo, "Experimental Study on Thermal and Tensile Properties on Polypropylene Maleic Anhydride as a Compatibilizer in Polypropylene / Sugarcane Bagasse Composite," J. Adv. Res. Fluid Mech. Therm. Sci., 43, no. 1 (2018): 141–148.
- [6] Bledzki, Andrzej K., V. E. Sperber, and Omar Faruk. *Natural and wood fibre reinforcement in polymers*. Vol. 13. iSmithers Rapra Publishing, 2002.
- [7] Mukhopadhyay, S., and Raúl Fangueiro. "Physical modification of natural fibers and thermoplastic films for composites—a review." *Journal of Thermoplastic Composite Materials* 22, no. 2 (2009): 135-162.
- [8] W. A. Mustafa, S. A. Saidi, M. Zainal, and R. Santiagoo, "Experimental Study of Composites Material Based on Thermal Analysis," *J. Adv. Res. Fluid Mech. Therm. Sci.*, 43, no. 1 (2018): 37–44.
- [9] W. A. Mustafa, S. A. Saidi, M. Zainal, and R. Santiagoo, "A Proposed Compatibilizer Materials on Banana Skin Powder (BSP) Composites Using Different Temperature," J. Adv. Res. Fluid Mech. Therm. Sci., 43, no. 1 (2018): 121–127.
- [10] Rozman, H. D., G. S. Tay, R. N. Kumar, A. Abusamah, H. Ismail, and ZA Mohd Ishak. "Polypropylene-oil palm empty fruit bunch-glass fibre hybrid composites: a preliminary study on the flexural and tensile properties." *European Polymer Journal* 37, no. 6 (2001): 1283-1291.
- [11] Gupta, G., A. Gupta, A. Dhanola, and A. Raturi. "Mechanical behavior of glass fiber polyester hybrid composite filled with natural fillers." In *IOP Conference Series: Materials Science and Engineering*, vol. 149, no. 1, p. 012091. IOP Publishing, 2016.
- [12] Bontempi, Elza. "Case Study of Raw Materials Substitution: Natural Fillers Substitution in Plastic Composites." In *Raw Materials Substitution Sustainability*, pp. 29-61. Springer, Cham, 2017.
- [13] Nah, Changwoon, Hyune Jung Ryu, Sang Hyub Han, John M. Rhee, and Myong-Hoon Lee. "Fracture behaviour of acrylonitrile–butadiene rubber/clay nanocomposite." *Polymer International* 50, no. 11 (2001): 1265-1268.
- [14] Noriman, N. Z., H. Ismail, and A. A. Rashid. "Characterization of styrene butadiene rubber/recycled acrylonitrilebutadiene rubber (SBR/NBRr) blends: the effects of epoxidized natural rubber (ENR-50) as a compatibilizer." *Polymer Testing* 29, no. 2 (2010): 200-208.
- [15] Tajvidi, Mehdi, Robert H. Falk, and John C. Hermanson. "Effect of natural fibers on thermal and mechanical properties of natural fiber polypropylene composites studied by dynamic mechanical analysis." *Journal of Applied Polymer Science*101, no. 6 (2006): 4341-4349.



- [16] Ragunathan, Santiagoo, Hanafi Ismail, and Hussin Kamarudin. "The Effect of Silane Treatment on Tensile Properties and Morphology of Polypropylene/Recycled Acrylonitrile Butadiene Rubber/Rice Husk Powder Composites." In *Key Engineering Materials*, vol. 471, pp. 472-477. Trans Tech Publications, 2011.
- [17] Razavi-Nouri, Mohammad, Fatemeh Jafarzadeh-Dogouri, Abdulrasoul Oromiehie, and Amir Ershad Langroudi. "Mechanical properties and water absorption behaviour of chopped rice husk filled polypropylene composites." *Iranian Polymer Journal* 15, no. 9 (2006): 757-766.
- [18] Ismail, H., and L. Mega. "The effects of a compatibilizer and a silane coupling agent on the mechanical properties of white rice husk ash filled polypropylene/natural rubber blend." *Polymer-Plastics Technology and Engineering* 40, no. 4 (2001): 463-478.
- [19] Naderi, Ghassem, M. Razavi Nouri, Mahmood Mehrabzadeh, and GH R. BAKHSHANDEH. "Studies on dynamic vulcanization of PP NBR thermoplastic elastomer blends." (1999): 37-42.