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PERFORMANCE OF SHORT NOIL HEMP FIBRE POLYPROPYLENE COMPOSITES

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ABSTRACT

In this study, short noil hemp fibre composite with varying fibre content were fabricated using intermixer and injection moulding machines. The maleic anhydride grafted polypropylene (MAPP) and maleic anhydride grafted Poly (ethylene octane) (MAPOE) were used as coupling agents for modifying the matrices. Tensile, dynamic mechanical and microstructural characterizations were performed by tensile strength testing, scanning electron microscope (SEM) and X-ray microtomography. The composites revealed better temperature resistance at higher fibre content. However, the increase in storage modulus and tensile strength was negligible in composites reinforced with more than 40 wt.% hemp fibres; due to the agglomeration of the fibres. The results also indicated that the tensile strength of the noil hemp fibre reinforced composites without a coupling agent was below the tensile strength of pure polypropylene. However, the addition of coupling agents improved the tensile strength of the noil hemp fibre reinforced polypropylene composites by increasing the reinforcing effects of the fibres due to the enhanced fibre/matrix interfacial adhesion. This was also confirmed by dynamic mechanical analysis. X-ray microtomography study concluded that coupling agent addition can also reduce the fibre breakage due to the better fibre dispersion in the polypropylene.

KEYWORDS

Polymer-matrix composites (PMCs), fibre/matrix bond, tensile strength, injection moulding.

INTRODUCTION

Hemp fibre has shown a promising specific tensile strength to be used as reinforcement in composites used in automotive, construction and packaging applications. Hemp is a sustainable resource due to its biodegradability and renewability (Roulson et al., 2006,Shubhra et al., 2010,Ku et al., 2011). Injection moulding can be used to produce high quality complex parts, which is evident from the mass of plastic parts with complex shapes used in the construction, furniture, appliance and automotive industries. Short fibres are normally used in injection moulding. Noil hemp fibres, which are by-products of hemp fibres used in the textile industry, are short fibres, and hence more economical than chopped scutched long hemp fibres to be used in short natural fibre composite. Additionally, noil fibres are highly degummed, more thermally resistant due to their lower pectin content and also 80% cheaper than scutched long fibres(Yan et al., 2013). Therefore, this project aimed to study the influence of fibre content and coupling agent addition on performance of short noil hemp fibre polypropylene composites.

MATERIALS AND METHOD

Noil hemp fibres were supplied by China-Hemp Industrial Investments and Holding Co. Ltd. A M800E polypropylene (Sinopec Shanghai petrochemical Co. Ltd., China) with melt flow index of 8.0 ± 1.5 g/10 min was used as the matrix. MAPP with MA content of 1.0 wt.% (Bondyram1001, Polyram Ram-On Industries, Israel) was used as the compatibilizer to the resin mix.

The composites were fabricated with varying compositions (as shown in Table 1). The coupling agent was mixed with the PP at 170–180˚C in the intermixer (XH-409, Dongguan City Xihua Testing Machine Co. Ltd., China) for 15 min before the addition of the hemp fibres. The oven-dried noil hemp fibres were mixed in the intermixer at 180˚C for another 15 min. The mixture was then rolled and crushed into granules with the size of 5-7 mm using a roll mixer (XH-401B, Dongguan City Xihua Testing Machine Co. Ltd., China) and a crusher (RPC-180, Shanghai Runpin Industry & Trade Co. Ltd., China). The compounded and dried granules were placed in an injection press (MJ 55, Chen Hsong Group, China) and the tensile specimens are injection moulded.

The tensile specimens were evaluated using a multifunctional tensile machine (AG-2000A, Shimadzu Corp., Japan) according to the ASTM D638-91 standard at the specified loading rate of 10 mm/min. The composite specimens were tested to failure and, the average value of 10 tensile-tested specimens was reported for each sample. Fracture surfaces of tensile test specimens were examined by scanning electron microscope (SEM) (JSM-646-LV, Shimadzu Corp., Japan)), operated at 20 kV, to investigate the effects of coupling agent on fibre–matrix interface adhesion.

Table 1. Composition of fabricated composite samples

Samples	Hemp (wt.%)	PP (wt.%)	MAPP (wt.%)	MAPOE (wt.%)
PP	0	100	0	0
10H	10	90	0	0
20H	20	80	0	0
30H	30	70	0	0
40H	40	60	0	0
50H	50	50	0	0
60H	60	40	0	0
30H2.5MAPP	30	67.5	2.5	0
40H2.5PP-MH	40	57.5	2.5	0
30H5MAPP	30	65	5	0
40H5MAPP	40	55	5	0
30H2.5MAPOE	30	67.5	0	2.5
40H2.5MAPOE	40	57.5	0	2.5
30H5MAPOE	30	65	0	5
40H5MAPOE	40	55	0	5

RESULTS AND DISCUSSIONS

The tensile strength of the composites as a function of fibre weight content is illustrated in Figure 1. Referring to the literature (Ku et al., 2011), it was expected to observe an initial increase from pure PP then a subsequent decline in tensile strength with an increase in fibre content. Although higher fibre content is generally preferred for achieving better mechanical properties of short fibre reinforced polymer composites (Ahmad et al., 2006), Figure 1 indicates that increase in fibre content resulted in a decrease in tensile strength of all composites without a compatibilizer. The tensile strength was reduced from 31 MPa for the pure PP to 27 MPa for the composite with 10 wt.% hemp fibre (10H sample). Tensile strengths of 20H, 30H, 40H anf 50H samples were, on average, 28 MPa and also below the tensile strength of pure PP. This phenomenon is symptomatic of poor interfacial adhesion between the hemp fibres and the polypropylene matrices as can be seen in the SEM micrographs of fractured surfaces without a coupling agent (Figure 2) reveals a number of holes and fibres with clean and smooth surfaces which are evidence of fibre pull-out. Also, gaps can be observed between the fibres and the matrix as a result of fibre deboning. Debonded/pulled out fibres implies poor interfacial bonding between the fibres and the matrix

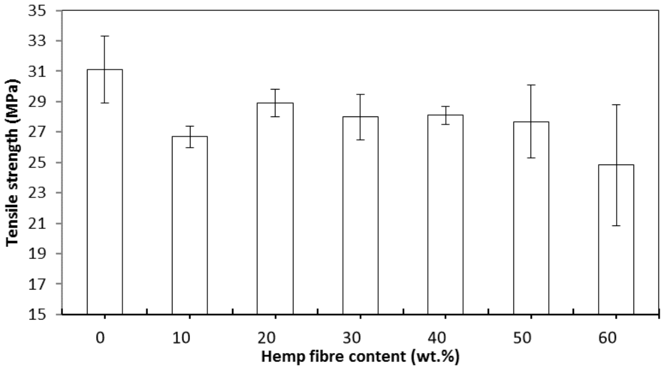

Figure 1. The influence of the fibre weight content on tensile strength of the noil hemp fibre reinforced polypropylene composites

Referring to Figure 1, it also can be noted that the tensile strengths of 30H, 40H, 50H and 60H composite samples are even lower than that of the 20H sample. It implies that the reinforcing effect of the fibres declined at higher fibre content. In order to take full advantage of the reinforcing effect of the fibres, the fibres' length must be greater than the critical value. Higher level of fibre breakage occurs during the processing of higher fibre content composites. Thus, at higher fibre content, the majority of the fibres are shorter than the critical length and the greater proportion of the total fibre length is not completely loaded.

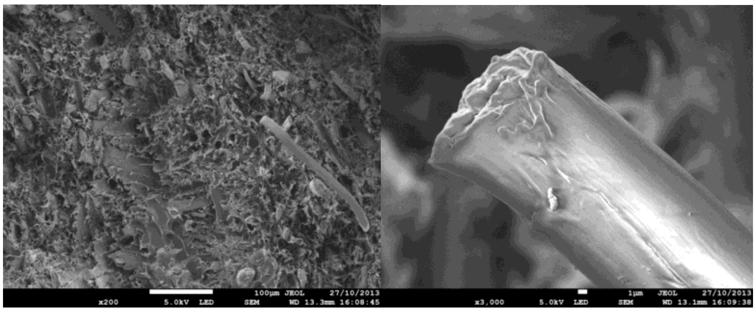

Figure 2. SEM micrographs of fractured surface for the 60 wt.% hemp fibre polypropylene composite without a coupling agent

The tensile strength of the composite with 60 wt.% hemp content is the lowest among all samples. The lower tensile strength of the composite with 60 wt.% hemp content occurred not only due to the low interfacial adhesion but also due to the voids can be observed in Figure 3. Figure 3 shows some typical voids observed in the microstructure of the composite. Since fibre addition increases the viscosity of the mixture, the voids are more likely to happen as the mixture with 60 wt.% hemp fibre is too viscous to effectively fill the mould.

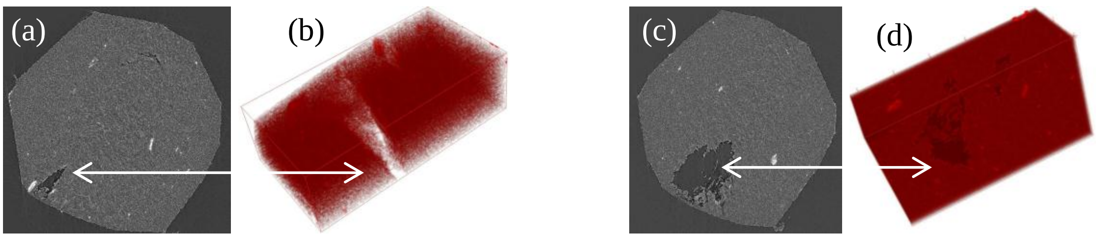

Figure 3. (a&c): Typical recontracted x-ray microtomography images of the composite with 60wt.% hemp fibre which shows voids, (b&d): 3D views of associated voids in the composites

When a coupling agent was added, the tensile strength of the composites improved. Figure 4 illustrates the influence of MAPP and MAPOE on the tensile composites with 30 wt.% and 40 wt.% noil hemp fibre. Referring to Figure 4, it can be seen that for the composite with 30 wt.% hemp fibre, there was very little difference between incorporation of 2.5 wt.% and 5 wt.% MAPP. While, for the composite with 40 wt.% hemp fibre, the addition of 5 wt.% MAPP caused the greatest improvement among the composites due to the higher availability of the fibre surface area. This figure indicates that addition of 5 wt.% MAPP into 40 wt.% hemp fibre improved both tensile strength (approx. 40%) and young's modulus (approx. 15%) of the composites in comparison with uncoupled ones. Also, it can be seen that the addition of MAPP had a stronger impact to enhance the tensile strength than MAPOE. This conclusion can be confirmed by a comparison between SEM micrographs illustrated in Figure 5 and Figure 6. Although MAPOE addition partially improved the interfacial adhesion, the evidence of pull-out can be observed yet in Figure 5. On the other hand, there was almost no evidence of fibre pull-out when MAPP was added. The micro-fibrils marked in Figure 6 indicate that composite failure occurred mainly due to the fracture of the cellulose chains.

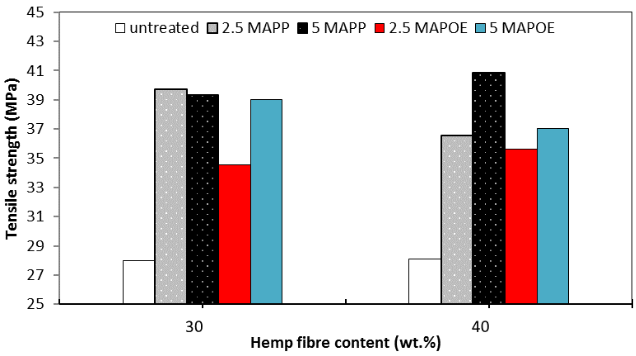

Figure 4. The influence of coupling agent type and their content on tensile strength of the noil hemp fibre reinforced polypropylene composites

Figure 5. SEM micrographs of fractured surface for the 40 wt.% hemp fibre polypropylene composite with 5 wt.% MAPOE coupling agent

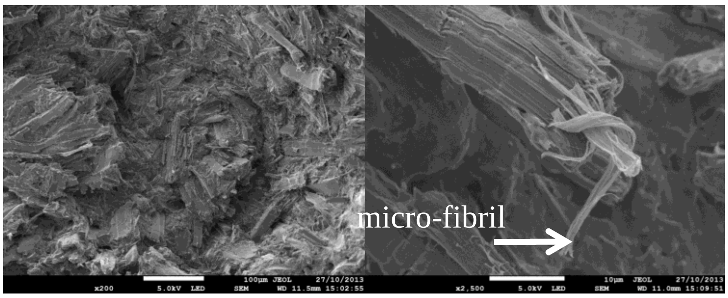

Figure 6. SEM micrographs of fractured surface for the 30 wt.% hemp fibre polypropylene composite with 2.5 wt.% MAPP coupling agent

Also, the effects of compatibilizer (MAPP) addition on dispersion of fibres in the composites can be observed in Figure 7 (40 wt.% hemp fibres). It can be observed that the addition of 5 wt.% MAPP resulted in better dispersion of fibres. The better fibre dispersion is attributed to the fact that addition of coupling agents to the matrix improves the wettability of the matrix by reducing the contact angle which ultimately results in more homogenous structure and accordingly higher tensile properties.

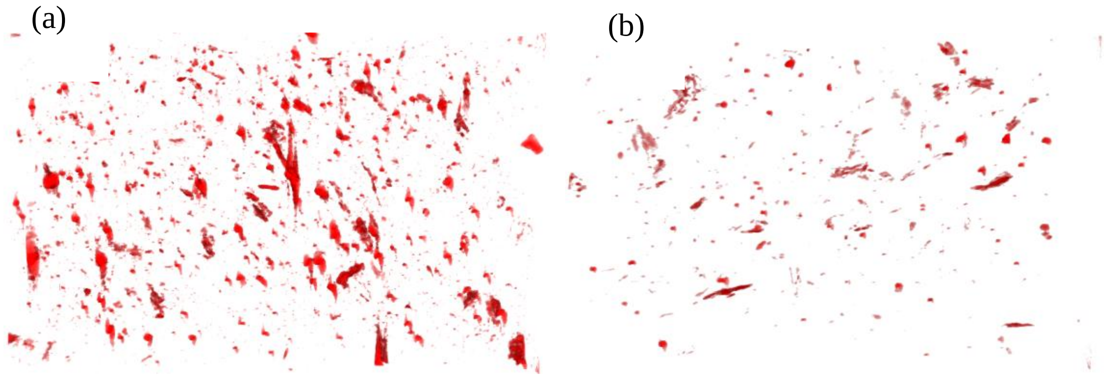

Figure 7. Noil hemp Fibre agglomerations in polypropylene matrix: (a) 40H and (b) 40H5MAPP

Figure 8 illustrates the effects of coupling agent addition on storage modulus values of 30 wt.% noil hemp fibre polypropylene composites. It can be seen that the storage modulus values of composites were greatly improved by the addition of the coupling agents into the PP matrix, which result from the enhancement of interfacial adhesion between the fibre and polypropylene matrix. Maleic anhydride coupling agents improve the bonding between fibre and matrix by chemically bonding to available OH groups on the fibre surface, and then adhering to the matrix through molecular chain entanglement. It is interesting that the greatest improvements in E′ were achieved again by the addition of 2.5 wt.% of either coupling agents and, higher coupling agent content declined E′ values of the composite. Table also confirms that 30 wt.% noil hemp composite coupled with 2.5 wt.% MAPP has the lowest effectiveness coefficient C (highest reinforcing effect) in the investigated temperature range.

As far as the bonding between the fibre and the matrix is concerned, tan δ is a better indicator, than storage modulus, as its value is not dependent on sample geometry (Tajvidi et al., 2010). Thus, in order to evaluate the effects of coupling agent on the composites, their effects on the polypropylene matrix and the matrix/fibre interphase was studied separately. In this regard, tan δ_S and tan δ_{in} of the coupled composites were calculated and then plotted in Figure 9. Referring to 9a, decrease in tan δ_s of the composites implies that the polypropylene matrix became more elastic as coupling agents were added.

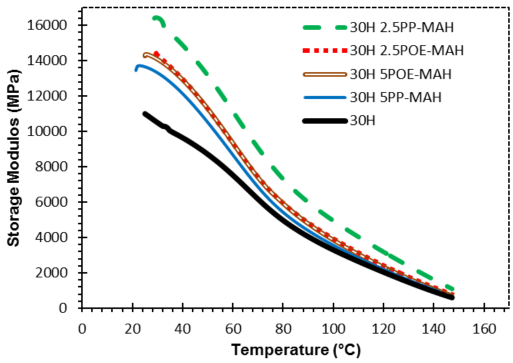

Figure 8. Influence of coupling agent addition on storage modulus values of of 30wt.%hemp sample

At constant fibre content, the damping term can be used to evaluate the interfacial properties between the fibre and the matrix. Figure 9b shows the effects of coupling agent addition on the interphase damping of the composite with 30 wt.% hemp fibre. General speaking, the addition of coupling agents significantly reduced the interphase damping (improvement in interfacial bonding).

It is worth noting that the effect of coupling agents appears to become more prominent at temperatures below the transition temperature than above it. Also, compared with MAPOE, MAPP coupling agent caused lower interphase damping ratio. This implies that bonding between the fibre and the matrix resulted from MAPP addition is considerably stronger than that resulted from MAPOE addition. This is in correlation of SEM graphs of figures 5 and 6.

On the other hand, at high temperatures (above transition temperature), the interphase damping ratios were almost at the same level for all coupled composites. It can also be seen that increase of coupling agent content from 2.5 to 5 wt.% MAPP reduced the δ_{in} of the composite, which then improved the interfacial bonding between the fibre and the matrix. This occurred due to higher probability of the MAPP groups to bond to OH groups on the fibre surface.

Table 2. Influence of coupling agent addition on effectiveness coefficient C of 30 wt.% noil hemp composite

Coupling agent	Content (wt.%)	C (90 ℃)	C (110 ℃)	C (130 ℃)	C (150 ℃)
MAPP	2.5	0.72	0.64	0.55	0.44
MAPP	5	0.79	0.74	0.66	0.55
MAPOE	2.5	0.77	0.72	0.64	0.53
MAPOE	5	0.78	0.73	0.68	0.58

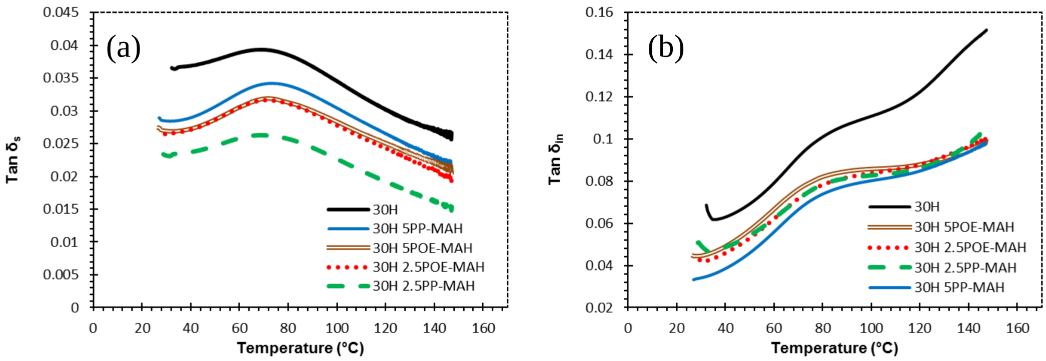

Figure 9. (a) System and (b) interphase damping ratio of coupled noil hemp fibre reinforced composites as function of temperature under DMA loading frequency of 1Hz

CONCLUSIONS

To conclude, poor interfacial adhesion between fibres and matrices is one of the main reasons caused poor tensile properties. Besides, the addition of short fibres to the matrix brought about a decline in composite strength as a result of higher fibre breakage (lower fibre length) occurring during processing with Furthermore, voids can happen due to the extremely high viscosity of the fibre/matrix blend at very high fibre content. 40wt.% hemp fibre polypropylene composite with 5 wt.% MAPP showed the highest tensile strength.

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