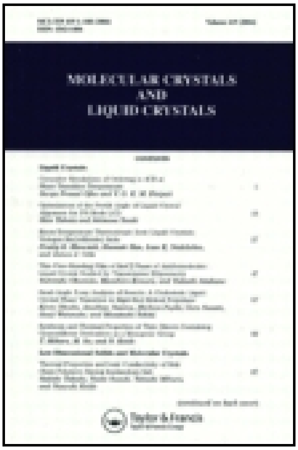

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Rheological Behaviour of Injection Moulded Oil Palm Empty Fruit Bunch Fibre – Polypropylene Composites: Effects of Electron Beam Processing Versus Maleated Polypropylene

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In this study, oil palm empty fruit bunch fibres (EFB) reinforced polypropylene (PP) composites were prepared by two different methods i.e. physical treatment of the composites using electron beam irradiation and chemical treatment by adding coupling agent such as maleated PP. The composites pellets were then subjected to injection moulding for preparation of test pieces for physical, mechanical and rheology properties study. Haake Rheometer RS 150 was used to study the changes in viscosities of the composites at different shear rates. The melt flows of the composites were also measured to complement and confirm the rheology behaviour of the composites. Upon irradiation, the viscosity of PP decreased due to chain scission and the sudden drop of viscosity at high shear rate was not significant. The presence of EFB fibre in the PP matrix was obviously interrupted the flow ability of PP that cause viscosity to increase. Upon irradiation of EFB/PP, the viscosity of the composites decreases. This indicate that no crosslinking occur between EFB and PP during melt mixing although PP and EFB active radicals were present in the composites. On the other hand, the additional malaeted PP increases interfacial adhesion that result in enhance resistance to flow, thus significantly increased in the viscosity of the composite. The addition of 6% maleated PP exhibited significant drop in viscosity of the composites. These results were further confirmed by the melt flow measurement of irradiated EFB/PP with reactive additives and maleated PP. The rheological behaviour of the composite is important parameters in plastic processing particularly in relation to the shear rates for injection moulding and extrusion processes.

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Keywords: crosslinking; electron beam; empty fruit bunch fibre; maleated polypropylene; rheology

INTRODUCTION

Natural plant fibres have been used in the past as a reinforcing material for different types of matrices [1–4]. In recent years, attention has been paid to their use as a reinforcing material for thermoplastics. In particular, the automotive industries have shown interest in the advantages that this type of fibre-reinforced system can provide [2]. The advantages of biofibres over traditional fibre reinforcements, such as glass fibres, are: low cost, low density (good specific properties), reduced wear in processing equipment, high toughness, biodegradability and "ecological friendliness" (since they can be produced from renewable resources) [5].

Maleated polypropylene (MAPP) as a compatibilizer for the natural plant fibers-thermoplastics, specifically PP composite, improved most of the properties of the plastic composites [6–9]. In the conventional processing, the MAPP act as compatibilizer whereby, the covalent bonding occurs only between the maleic functional group and the hydroxyl group of cellulose wall fibres. The long chain of polypropylene of the MAPP is entangled with the PP molecules chain [10]. The longer chain of MAPP, the more entanglement with the PP chain will be take place and this make stronger bind and therefore need better strength to align the molecule chains till slippage.

Radiation treatment was also found to enhance the properties of wood fiber-thermoplastic composites [7,11,12]. The long live polypropylene radicals induced by ionizing radiation and the subsequent formation of polypropylene peroxides radicals have been skill fully exploited by Czvikovszky to be used in the reactive extrusion with irradiated wood fibres to produce wood fibre reinforced polypropylene composites [11,13].

In this study attempt is made to study the rheological behaviour of the EFB/PP composites under treatment of MAPP and radiation processing. The study of flow, rheology of the composite gives in detail perceptive on the processing of the composites and the interface among two or more elements inside the composites.

MATERIALS AND METHODS

The oil palm empty fruit bunch (EFB) fibres used was processed into 0.05–0.15 mm sizes. These fibres usually have moisture content in the range of 1% ~ 1.5%. The polypropylene (PP) used was Titan Pro-fax 6331 (homopolymer) with specific density and melt index of 0.9 g/cm^3 and 14 g/10 min, respectively. A ratio of 50/50 of EFB fibres to polypropylene pellets at were melt-blended using a Thermo Haake kinetic mixer, at 180°C.

The hot mixed composite of EFB/PP were cut into pieces and subsequently undergo injection moulding for the preparation of test piece samples. In this study, two types of maleic anhydride polypropylene (MAPP) that have difference Melt Flow Index (MFI), 10.6 g/10 min (type A) and 5.9 g/10 min (type B) were used as compatibilizers at difference weight percentages, 2%, 4% and 6%.

The radiation processing treatment require both PP and EFB to be exposed under the electron beam irradiation from 3.0 MeV and 90 kW accelerator, EPS3000 NHV, Japan at 10 kGy. Two types reactive additives (RA) were applied, describe as, hexanediol diacrylate or 1,6-hexadiol diacrylate (HDDA) and trimethylol propane triacrylate (TMPTA) at 1% and 2% weight ratio. The melt flow index test was carried out according to ASTM D 1238–95 using Toyoseiki melt indexer model SO1 at temperature 190°C and 2.16 kg load.

RESULTS AND DISCUSSION

Effects of MAPP on Rheology of EFB-PP Composites

The addition of MAPP coupling agent in the EFB/PP composites is expected to increase the interfacial adhesion between the fibers and polymer chain. The maleic anhydride group of the MAPP will react with the hydroxyl groups of the fibers to form covalent bond as shown in Figure 1 whereas the polypropylene molecular chain of the MAPP will undergo physical entanglement with the polypropylene matrix.

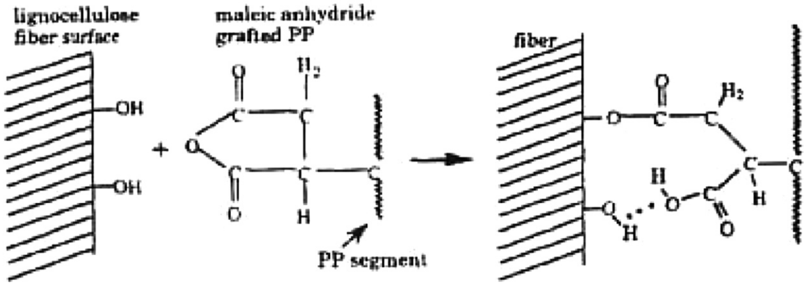

FIGURE 1 Reaction mechanism of MAPP with the cellulose surface and to the PP segments. Source [10].

With this arrangement, the composite is expected to have high viscosity and would require high shear stress for the alignment of the molecules [14].

Figures 2(a) and (b) demonstrate the effects of shear rates on the viscosity of the composites at 2%, 4% and 6% MAPP concentrations

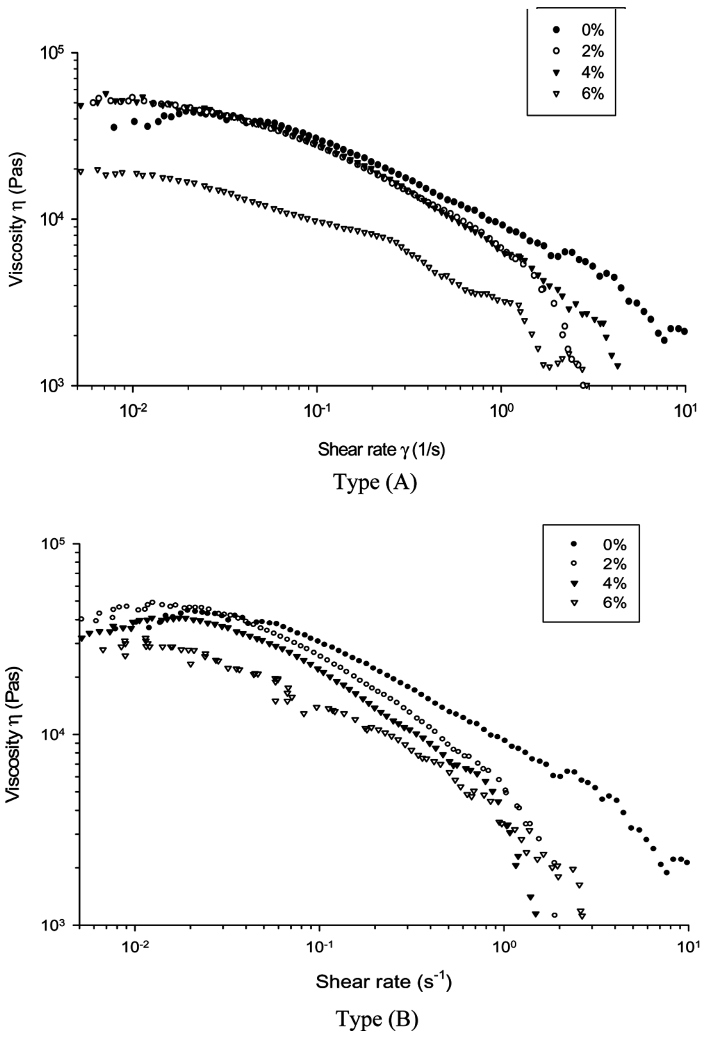

FIGURE 2 Effect of shear rate on viscosity of EFB/PP composites prepared from different MAPP (a) MFI, 10.6 g/10min (Type A) (b) MFI, 5.9 g/10min (Type B).

of different MFI, For MAPP type A, the viscosity of the composite is not influence by the presence of MAPP at low shear rate. However, above $0.05\,s^{-1}$ shear rate, the MAPP treated composite viscosity started to reduce lower than the composite without MAPP. The viscosity continues to reduce at high shear rate. Similar behaviours were observed when using MAPP type B. However, the composite viscosity reduction even faster, and started at $<0.02\,s^{-1}$ shear rate. This observation indicates that MAPP is acted like lubricant although fibers are bound with MAPP through covalent bond between maleic and hydroxyl groups of fibers. The PP backbone of the MAPP may entangle with PP matrix but apparently they are easy to slip under stress. At 6% concentration of MAPP, the viscosity of the composite is much lower than without MAPP. This is obviously revealed that the PP backbone of the MAPP is entangle among them self and not with PP of the matrix. The fibres seem to be coated by MAPP. Although this will improve the compatibility between fibres and PP matrix but there are not enough physical entanglement. It has been suggested that the amount of MA grafted and the molecular weight of the MAPP determine the efficiency of the additives as compatibilizer [15,16]. Figure 3 shows the MFI of EFB/PP composites prepared from different type of MAPP at various concentrations. It can be seen that the

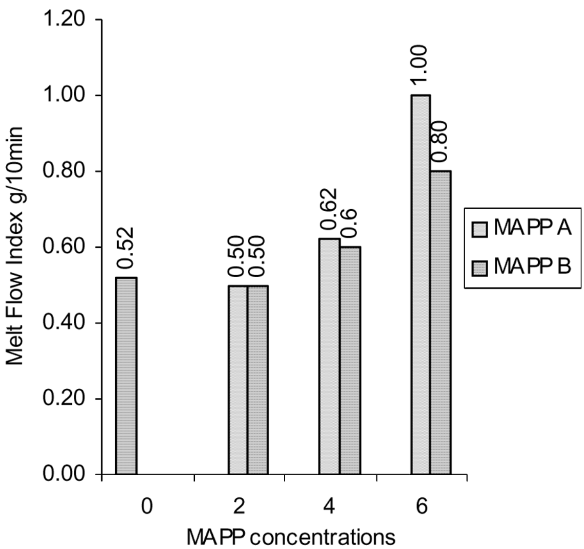

FIGURE 3 MFI of EFB/PP composites prepared from different type of MAPP at various concentrations.

MFI of 6% of MAPP type A slightly increase in MFI thus convince the lubricant effects of MAPP of higher MFI.

Effects of Electron Beam Irradiation on EFB/PP Composites

The viscosity of unirradiated and irradiated EFB/PP as a function of shear rate is shown in. At a lower molecular weight of PP, the viscosity drop at a steady rate as the shear rate increases (cf. Fig. 4). However, for unirradiated EFB/PP, the shear thinning region is very clear at high shear rate, above 10^{-1} s.

Figure 5 implies that along the shear rate investigated, the additional RA increased the viscosity of the composites. However the trend decreased when increased the RA levels. It reveals also that with the presence of tri functional monomer additive (TMPTA) the viscosity of the composite increases ten fold. This is due to the bonding radicals occur between PP radicals and TMPTA and also EFB fibre and TMPTA. With such network function restrict the flow of the composite and hence increase the viscosity. However di functional monomer additive HDDA seems not effectively significantly increase the viscosity of the irradiated composites. However, to some extend HDDA facilitates to partially cross link PP radicals and EFB that maintain the viscosity of the composite at the same level as untreated irradiated composites.

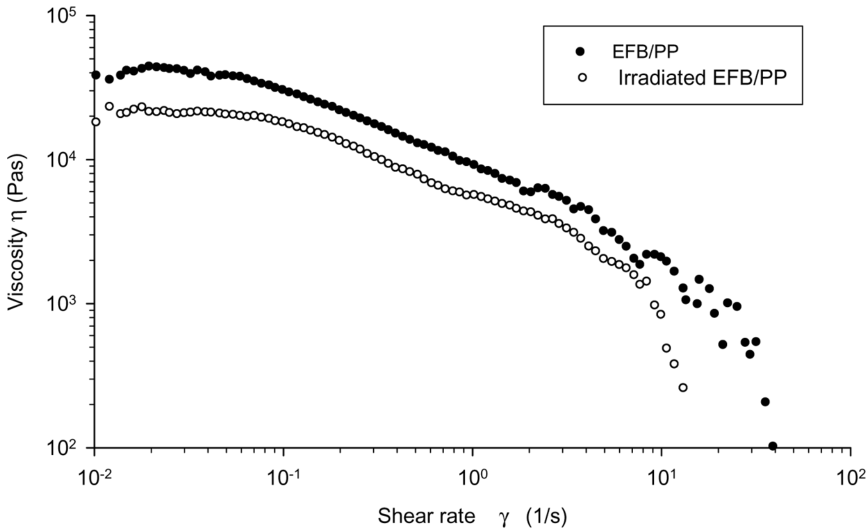

FIGURE 4 Viscosity curves of EFB/PP and irradiated EFB/PP as a function of shear rate.

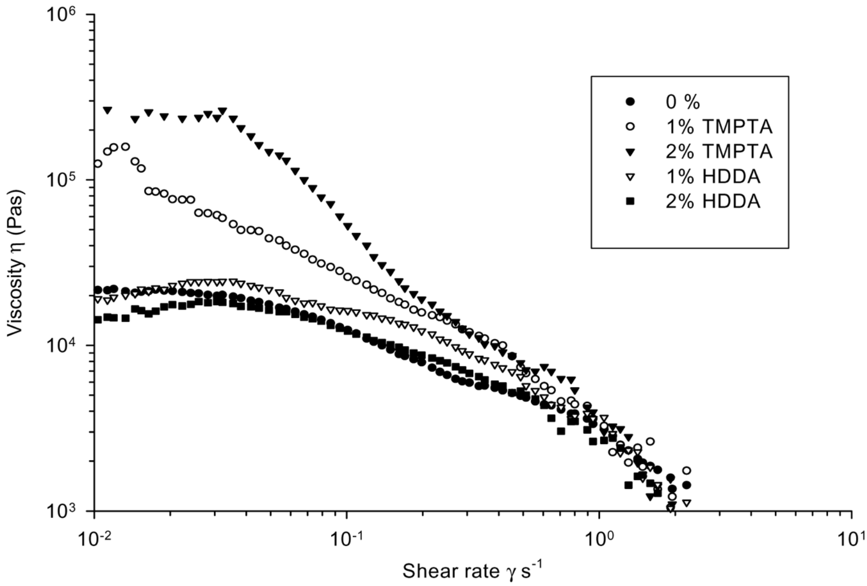

FIGURE 5 Viscosity curves of irradiated EFB/PP in the presence of RAs with function of shear rate.

Radiation induced degradation of polypropylene can be described as follows:

(a) $\sim CH_2$ ---------- $CH_2\sim$ ⁄VVVVV↘ -•CH-CH_2-

⁄VVVVV↘ $\sim \bullet CH_2 + \bullet CH_2 \sim \rightarrow \sim CH_3 + \sim CH_3$ (1)

(b) X• +~CH ---CH_2~ → ~C•---CH_2~ + HX

| |

~   ~

•O_2

| (2)

~C•---CH_2~ + O_2 → ~~C ---CH_2

l   l

~~   ~~

The free radicals of cellulose fibres are also produced upon exposure of EFB to electron beam irradiation. At the ambient temperature the original cellulose radical can be transformed into peroxy radicals.

$$\text{Cell}\bullet + O_2 \rightarrow \text{Cell-OO}\bullet \tag{3}$$

which can lead further oxidation of cellulose via chain reaction

$$\begin{aligned}&\text{Cell-OO}\bullet + \text{H-Cell} \rightarrow \text{Cell-OOH} + \text{Cell}\bullet\\&\text{Cell}\bullet + O_2 \rightarrow \text{Cell-OO}\bullet\end{aligned} \tag{4}$$

CONCLUSION

Rheology, plays a vital roll in developing industrial processes in the manufacture of those made of plastic which rely on injection moulding, extrusion, blow moulding and rotational moulding. The rheological behaviours of EFB/PP composites measured by the viscosity and shear stress have contributed some insight of the flow behaviour of natural fibre composites. The additional MAPP coupling agent in these studies increases the interfacial adhesion. However improvement on adhesion slightly increase the resistance to flow of the composite, thus increase the viscosity. The viscosity curve was altered depending to the melt flow index of the MAPP and concentrations level. 2% of both types of MAPP produced slightly increase in viscosity but 6% from similar additives dropped down the viscosity far lower than untreated EFB/PP. The interaction between fibres and polypropylene can only be achieved when both of them are subjected to electron beam irradiation in the presence of trifunctional acrylate. The resultant composite shows high viscosity at low shear rate.

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