THE EFFECT OF COUPLING AGENT ON THERMAL PROPERTIES AND MORPHOLOGY OF PAPER SLUDGE FILLED POLYPROPYLENE (PP)/ETHYLENE PROPYLENE DIENE TERPOLYMER (EPDM) COMPOSITES

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Abstract: The paper sludge was used as a filler in PP/EPDM composites and 3-aminopropyl triethoxysilane (3-APE) was used as a coupling agent in this study. The effects of filler loading and 3-APE on the thermal properties and morphology of composites were investigated. It was found that incorporation of a silane coupling agent (3-APE) increased the crystallinity of PP/EPDM/PS composites. The scanning electron microscopy (SEM) study of the tensile fracture surface of the composites indicates that the presence of 3-APE increased the interfacial interaction between paper sludge and PP/EPDM matrix.

Keywords: coupling agent, paper sludge, polypropylene, ethylene propylene diene terpolymer, composites.

Introduction

The disposal of sludge from the production of pulp and paper is a difficult environmental problem bacause most sludge is landfill. Although some sludge is mixed with hog fuel and incenerated, some is composted and sold, and some is applied to forest lands, these are not common practices [1].

On the other hand, waste wood fibres separated from paper sludge, beside conventional inorganic reinforcing filler such as clay, talc, calcium carbonate, and so forth, also have the feasibility of being effective filler in manufacturing thermoplastic polymer composites. The use of reinforcing fillers for the reduction of material cost and improvement of composites performance is constanly increasing in the area of the thermoplastic polymer composites. These benefits, in comparison with typical inorganic filler, include its low hardness and the resulting minimal abrasion of processing equipment, relatively low composites density, and low production cost on the unit volume basis.

In our previous work, we have reported the use of paper sludge as a filler in thermoplastic elastomer composites [2-5]. The main problem of preparation of paper sludge-thermoplastic elastomer composites is the incompatibility of hydrophilic paper sludge and hydrophobic thermoplastic elastomer matrix, which yield composites of poor properties. Coupling agents or compatibilizer has been used to improve dispersion, adhesion and compatibility for system containing a hydrophilic filler and hydrophobic matrix.

In this study investigates the effect of a coupling agent, 3-Aminopropyltriethoxysilane (3-APE) on thermal properties and morphology of paper sludge filled polypropylene/ethylene propylene diene terpolymer composites.

Experimental

Materials

Polypropylene homopolymer used in this study was of injection molding grade, from Titan PP polymers (M) Sdn Bhd, Johor, Malaysia (code 6331) with MFI value of 14.0 g/10 min at 230 ^{0}C. Ethylene propylene diene terpolymer grade Mitsui EPT 3072E with tertiary component ENB and 74 Mooney viscosity ($ML_{1+4}100^0C$) was obtained from Luxchem Trading Sdn Bhd., Selangor, Malaysia. Paper sludge (PS), a waste product from paper mills process, was obtained from Nibong Tebal Paper Mill Sdn Bhd, Penang, Malaysia. Paper sludge was dried in a vacuum oven at 80 ^{0}C for 24 hour to make it free from moisture and then grinded to become powder. An Endecotts sieve was used to obtain an average filler sizes of 63 µm (density, 2.2 g/cm^3). A coupling agent, 3-aminopropyltriethoxysilane (3-APE) was supplied by Bumi Sains, Selangor, Malaysia. The formulation of PP/EPDM/PS composites with and without coupling agent used in this study is shown in Table 1.

Table 1. Formulation of PP/ EPDM/PS composites with different filler loading and 3-aminopropyl triethoxysilane (3-APE)

Materials	**Composites 1**	**Composites 2**
Polypropylene (PP) (wt %)	50	50
Ethylene propylene diene terpolymer (EPDM) (wt %)	50	50
Paper sludge (PS) (wt %)	0, 15, 30, 45, 60	0, 15, 30, 45, 60
3-APE (wt %)	-	3

Mixing Procedure

Composites were prepared in a Haake Reomix PolyDrive R 600/610. Mixing was done at 180 ^{0}C and 50 rpm. EPDM was first charged to start the melt mixing. After 3 min filler and coupling agent were added followed by PP at the fifth minutes. Mixing was continued for another 5 min. At the end of 10 min, the composites were taken out and sheeted through a laboratory mill at 2.0 mm nip setting. Sample of composites were compression molded in an electrically heated hydraulic press. Hot-press procedures involved preheating at 180 ^{0}C for 6 min followed by compressing for 4 min at the same temperature and subsequent cooling under pressure for 4 min.

Differential scanning calorimetry

Thermal analysis measurements of selected systems were performed using a Perkin Elmer DSC-7 analyzer. Samples of about 10 – 25 mg were heated from 20 to 220 ^{0}C using a nitrogen air flow of 50 ml/min and the heating rate of 20 ^{0}C/min. The melting and crystallization behavior of selected composites were also performed using a Perkin Elmer DSC-7.

Morphology Study

Studies on the morphology of the tensile fracture surface of the composites were carried out using a scanning electron microscope (SEM), model Leica Cambridge S-360. The fracture ends of specimens were mounted on aluminium stubs and sputter coated with a thin layer of gold to avoid electrostatic charging during examination.

Results and Discussion

Figure 1 shows the DSC curve of PP/EPDM/PS composites with and without 3-APE at different filler loading. Table 2 indicates the value of melting temperature(Tm), heat fusion of composites ($\triangle H_{f\ (com)}$), crystallinity of composites(X_{com}) and crystallinity of PP (X_{pp}) for PP/EPDM/PS composites with and without 3-APE. The results show that the percentage of crystallinity of both composites changes with filler loading. It can be seen that the value of $\triangle H_{f\ (com)}$ and X_{com} decrease with increasing filler loading. This is due to the decreasing of PP content at higher filler loading. The addition of filler results in an increase in $X_{pp.}$ This observation was due to the nucleating ability of fillers for the crystallization of PP. From Table 4, at similar filler loading PP/EPDM/ PS composites with 3-APE exhibit higher X_{com} and X_{pp} than PP/EPDM/PS without 3-APE. This might be due to the better nucleation effect of paper sludge with the presence of 3-APE. However, the presence 3-APE has reduced the melting temperature of composites.

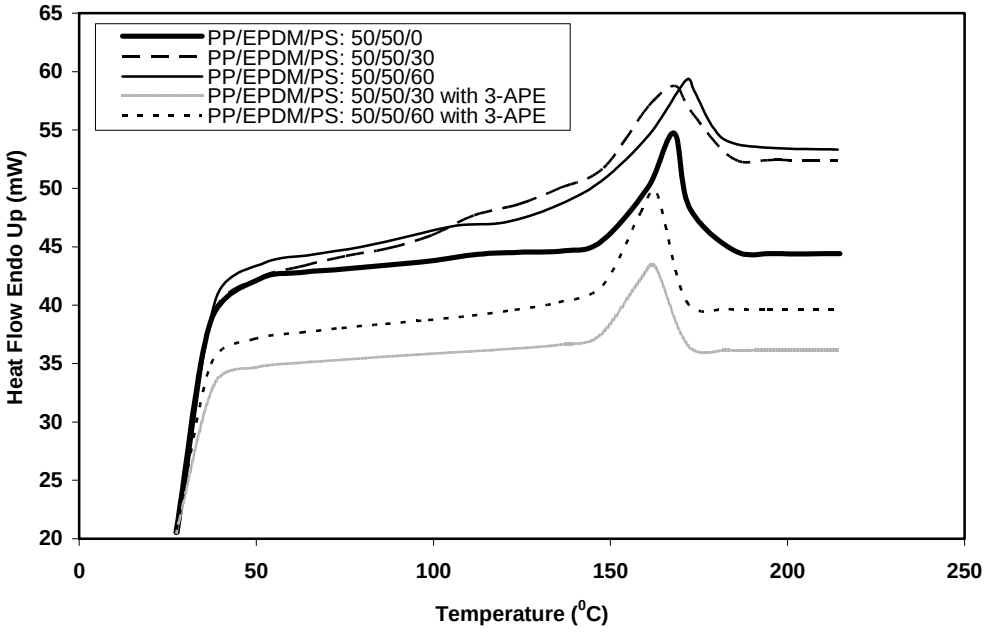

Figure 1. Comparison of differential scanning calorimetric curve of PP/EPDM/PS composites with and without 3-APE.

Table 4. The thermal parameter DSC of PP/EPDM and PP/EPDM/PS composites with and without 3-APE

Composites	Melting temperature T_m (^{0}C)	$H_{f\ (com)}$ J/g	X_{com} (% crystallinity)	X_{pp} (%)
PP/EPDM: 50/50	167.1	40.07	19.2	38.4
PP/EPDM/PS: 50/50/30	167.3	36.45	17.4	45.1
PP/EPDM/PS: 50/50/60	167.5	35.35	16.9	54.1
PP/EPDM/PS: 50/50/30 with 3-APE	161.4	40.50	19.3	50.1
PP/EPDM/PS: 50/50/60 with 3-APE	161.7	39.17	18.7	59.8

Figures 2 and 3 show the tensile fracture surface of paper sludge filled PP/EPDM composites at 30% and 60% weight of filler. It can be seen that surfaces exhibit poor dispersion, filler agglomerate pull out and unwetted paper sludge agglomerates in the PP/EPDM matrix. The polar characteristic of paper sludge is not capable of forming a good filler- matrix interaction with

non-polar PP/EPDM composites. However, in the presence of 3- APE, there is an evidence of improvement in interfacial bonding between the filler and PP/EPDM composites, as can be seen in Figs. 4 and 5. The both of Figures also show the fillers were coated with PP/EPDM matrix and better adhesion occurred between filler and PP/EPDM matrix.

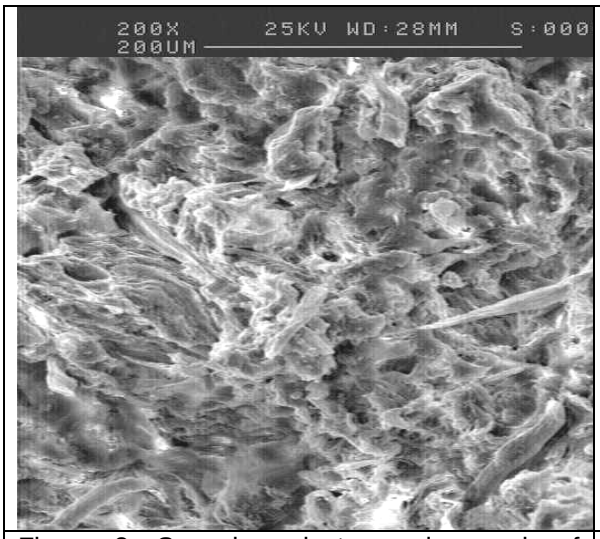

Figure 2. Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite (30 wt %) at magnification 200X.

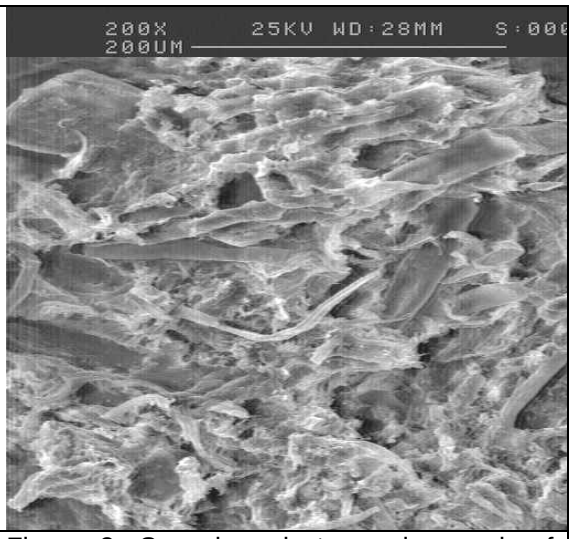

Figure 3. Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite (60 wt %) at magnification 200X.

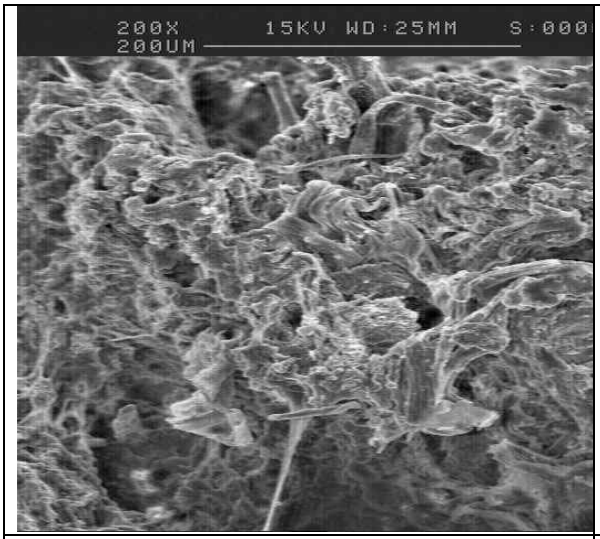

Figure 4. Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (30 wt %) at magnification 200X.

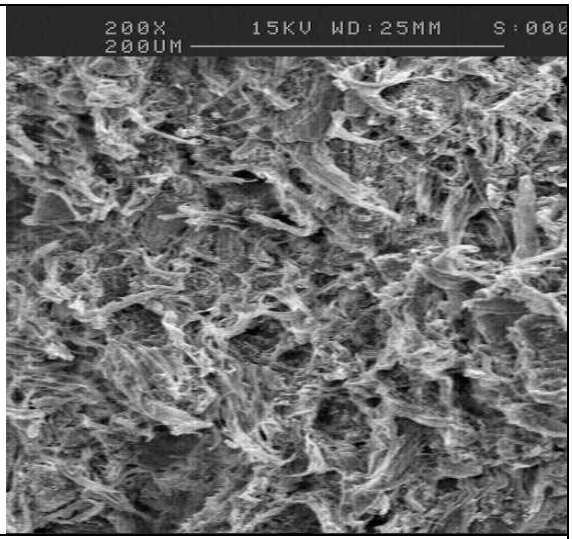

Figure 5. Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (60 wt %) at magnification 200X.

Conclusion

The result showed that the increasing of paper loading deacrease the crystallinity of PP/EPDM/PS composite. Thermal properties showed that the presence of 3-APE increased the crystallinity of PP/EPDM/PS composites. Scanning electron microscopy studies indicate that the interfacial adhesion between paper sludge and PP/EPDM matrix is improved with the presence of 3-APE.

Reference

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