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The Effects of Coupling Agents on the Mechanical and Thermal Properties of Eucalyptus Flour/HDPE Composite

Siripan Metanawin[1,a], Phiromrat Pengrung[1] and Tanapak Metanawin[2]

[1] *Department of Textile Engineering, Faculty of Engineering, Rajamangala University of Technology Thanyaburi, PathumThani, 12110, Thailand*

[2] *Department of Materials and Production Technology Engineering, Faculty of Engineering, King Mongkut's University of Technology North Bangkok, Bangkok, 10800, Thailand*

Abstract. The aim of this research was to study the effects of the coupling agents, Fusabond[TM] E-528 (polyethylene-grafted maleic anhydride; PE-g-MA, MA) and Amino Silane (Si), on the thermal properties, and mechanical properties of Eucalyptus flour-HDPE composite. Variation of the Eucalyptus flour contents in the HDPE resulted in properties of the composite. With increasing in the contents of Eucalyptus flour in polymer matrix, the mechanical properties of the HDPE composite decreased in EU-MA series samples while they were gradually decreased in EU-Si series samples. SEM micrographs showed the fracture surface of the HDPE/Eucalyptus composite at different ratios of Eucalyptus flour. SEM micrograpgh exhibited the dispersion of EU flour in polymer matrix. The samples of both coupling agents showed an increase in interfacial adhesion, observed for the considerable decreased of gaps between the matrix and the dispersed phase. However, the EU-MA sample appeared to be more uniformly than the EU-Si sample.

1 Introduction

Much attention has been focused on the new materials in the use of biofibers as fillers and/or reinforcers elements in polymeric matrix which has been concerning on global ecological. Furthermore, the specific properties of this natural product such as low cost, light weight, renewable character, high specific strength and high modulus, reactive surface and the possibility to generate energy, without residue after burning at the end of their life-cycle, motivate their association with organic polymers to elaborate composite materials [1-4]. For the above reason, much work has been done in studying and developing general thermoplastic/natural fibers composites, especially wood plastic composites (WPCs). WPCs have successfully proven their applications in various fields such as lumber, decking and railing, window profiles, wall studs, door frames, furniture, pallets, fencing, docks, siding, architectural profiles, boat hulls, and automotive components [4, 5].

However, it is well known that different surface properties between the fiber and the matrix, i.e. the matrix is highly polar and hydrophilic while the filler is non-polar and relatively hydrophobic, the compatibility and adhesion between matrix and filler are poor. Therefore, the coupling agents are much interested in order to improve the fibers /polymer compatibility and their interfacial adhesion [4, 6, 7]. It is known that coupling agents such as silane and olefin grafted maleic anhydride are an effective way to improve its dispersibility in polymer matrix as well, and hence ameliorate the polymer matrix, thus enhancing the properties of the resulting composites [6, 7].

The high density polyethylene (HDPE) is one of the most widely used in automotive, electric, and consumer applications and household applications. Beside, HDPE also enhances the comprehensive properties of semicrystalline engineering thermoplastic including a high rate of crystallization, good solvent resistance, thermal stability, and excellent processing properties and hence finds its applications in despite its exquisite properties; HDPE is very often modified with other polymers and particulate fillers [5].

Recently, much interest in new materials that can demonstrate WPCs products which developed polymeric matrix compatibility with wood flour for specific properties [2, 5]. To improve the compatibility between hydrophobic (non-polar) thermoplastics and hydrophilic (polar) wood flours, the preparation of polymer composites with wood flour requires good compatibility in the interface between the polymer matrix and the wood flour. In this research we designed a wood plastic composite (WPCs) for household application such as furniture. The Eucalyptus wood residues (EWR) were used as a filled from these post-consumer materials in order to reduce the environmental impact and add value to these materials. Two type of coupling agents, Fusabond[TM] E-528 and Silane were used in this process in order to enhance compatibility and

[a] Corresponding author: siripan.m@en.rmutt.ac.th

adhesion between wood-flours and thermoplastic matrices.

2 Experiments

2.1 Materials

High density polyethylene (HDPE H5818J) was purchased from SCG Plastic Co., Ltd. Fusabond^{TM} E-528 (polyethylene-grafted maleic anhydride, PE-g-MA, MA) were supplied by Creative Polymer Co., Ltd. Amino Silane (Si) and Chiguard R326 (UV absorber) were purchased from Chitec Technology Co., Ltd. Eucalyptus fiber was from Bangkok Thailand. All samples were used as received. All other chemicals were used as supplied by the companies.

2.2 Preparation of Eucalyptus wood-flour/HDPE

Wood fibers were grinded using grinding machine. The Eucalyptus flours (EU) with particle size about 35µm was obtained from sieving method. The HDPE pellets and EU were dried at 80°C overnight prior used. The formulation of composites materials were presented in Table1. The 500g of HDPE pellets was mixed with 5wt%-20%wt EU fine powder containing 1wt% of coupling agent and 0.5wt% of ChiguardR326. The glycerol 3 drop was added into the mixture. The EU was blended with HDPE using Twin Screw Extruder. The extruder barrel-temperatures zones were set at 190°C. The screw speed was 60 rpm. The obtained EU/HDPE compound composites were cut into pellet size.

2.3 Preparation of EU/HDPE slit.

The EU/HDPE composite pellets were mixed using ThermoHakePolyDrive (single screw extruder). The barrel-temperatures zones were operated 190°C. The screw speed was 40 rpm. The melting composite passes through the slit die (slit = 0.3 mm x 4 mm.) was first draw at 10m./min. and follow by recrystallized at 80°C and then second draw speed at 20m./min. The EU/HDPE slit was obtained.

Table 1. Formulation of composites materials.

Specimen ID	Materials	Filler	Coupling agent	HDPE (%w/w)	Eucalyptus (%w/w)	Coupling agent (%w/w)
EU-0	HDPE	-	-	100	-	-
EU-MA 5	HDPE	EU	PE-g-MA	95	5	1
EU-MA 10	HDPE	EU	PE-g-MA	90	10	1
EU-MA 20	HDPE	EU	PE-g-MA	80	20	1
EU-Si 5	HDPE	EU	Silane	95	5	1
EU-Si 10	HDPE	EU	Silane	90	10	1
EU-Si 20	HDPE	EU	Silane	80	20	1

2.4 Characterization

The morphology of the specimens were observed using scanning electron microscope model JSM-5410LV from Jeol, Japan. The cross-sectional fracture were obtained by breaking the specimens after freezing in liquid nitrogen. All of the samples were coated with gold before observed.

The tensile test of the slit composites were performed on Instron 5569 universal testing machine according to ASTM D3822. The speed of the cross head was 30 mm min^{-1}.

Thermal properties of the polymer compounds were investigated using differential scanning calorimetry model DSC 200 F3 from Netzsch, Germany. The scan were performed from 30°C to 300°C with heating rate and cooling rate 10°C/min. under nitrogen atmosphere.

3 Results and Discussions

3.1 EU/HDPE polymer compound

The incorporation of fillers into thermoplastics is another method widely used to enhance certain properties. The degree of property enhancement depends on the filler particle size and shape, the content of filler, the surface treatment promoting interaction between the polymer matrix and filler and most importantly the filler's origin. One of the disadvantages of using natural fibers is that they are incompatible with hydrophobic thermoplastics. Therefore, two type of coupling agents, polyethylene-grafted maleic anhydride (PE-g-MA) and silane were used in this process in order to enhance compatibility and adhesion between wood-flours and thermoplastic matrices. PE-g-MA has been widely used at low concentrations as a compatibilizing agent and adhesion promoter for bio-filler filled polyethylene composites. The maleic anhydride (MA) functional group which grafts onto the PE backbone acts as a chemical link between the hydrophobic matrix polymer and the hydrophilic surface of natural fillers. Silane is another effective coupling agent, which improved the toughness and impact strength of the composites through enhanced bonding at the wood–matrix interface. The polymer composite compound were prepared by mixing HDPE pellets and Eucalyptus flours (EU) 5wt%-20wt% in the present of PE-g-MA (MA) or Silane (Si). Then the EU/HDPE compound composites were cut into pellet size. The EU/HDPE slit were obtained from single screw extruder. The rough surfaces of the slit were observed with respected to the content of EU.

3.2 Thermal properties

The melting temperature (Tm), crystalline temperature (Tc) and %crystallinity of the HDPE/Eucalyptus fibers were explored using Differential Scanning Calorimetry. It was found that the melting temperature of the composites were obtained from heating cycle while the crystalline temperatures were obtained from cooling cycle. There were no significant difference of the melting temperature and crystalline temperature when increase the amount of Eucalyptus fibers from 0wt% to 20wt% as shown in Table 2. Melting temperature of the HDPE and their composites was around 131.2°C-133.6°C. The %crystallinity was slightly increased when increased the content of EU flour 5wt% in both series samples. However, %crystallinity slightly decreased at high content of EU flour 20wt%. It was indicated that high content of EU flour effect the %crystalline [5]. In addition, there was no significant effect of coupling agent on the melting temperature and crystalline temperature of composite.

Table 2. Thermal properties of HDPE and EU/HDPE composite with various content of EU from 5wt%, 10wt% and 20wt% in the present of PE-g-MA or Silane.

Sample name	Tm (°C)	Tc (°C)	%crystallinity
EU-0	131.2	115.4	56.86
EU-MA 5	131.3	115.2	59.70
EU-MA 10	131.6	115.1	57.11
EU-MA 20	131.3	115.0	57.63
EU-Si 5	131.8	116.2	63.58
EU-Si 10	133.6	115.6	65.14
EU-Si 20	131.5	115.6	56.52

3.3 Scanning electron microscope (SEM)

Figure 1 show the fracture surface of the composites slit with both of coupling agents at content of EU 10wt%. It was well known that the coupling agent could be effect on the interfacial properties of the composites [7]. The samples in which PE-g-MA and silane were used as coupling agents showed an improving in interfacial adhesion, observed for the considerable decreasing of gaps between the matrix and the dispersed phase as shown in Figure 1. This SEM image of EU-MA10 showed less holes and void spaces at the fracture surface, which implies that there was a good filler–matrix interaction between MA and the matrix. On the other hand, SEM image of EU-Si10 showed holes and void spaces at the fracture surface, which denotes that there was a poor filler–matrix interaction between silane and the matrix. This indicates that the PE-g-MA coupling agent was better than Si coupling agent. The presence of voids in the EU-Si series, as shown in Figure 1, may create stress concentration points, which in turn reduce the strength of the samples [6]. In addition, this would affect the mechanical and thermal performance of the substrate where the flours will potentially be used [6].

EU-MA10

EU-Si 10

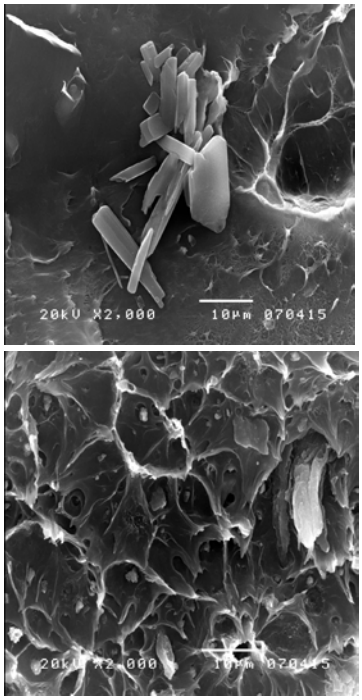

Figure 1. Fracture surface SEM images (2000x) of EU/MA and EU-Si10 composite with EU 10wt% in the present of PE-g-MA and Silane.

3.4 Tensile Test

The mechanical properties of HDPE and EU/HDPE composite with various content of EU from 5wt%, 10wt% and 20wt% in the present of PE-g-MA and Silane were examined using Instron 5569 universal testing machine. The tensile strength and elongation of the composite were demonstrated in Figure 2 and Figure 3, respectively. The results showed that EU-MA5 had higher tensile strength more than EU-Si5. Moreover, tensile strength of the composite dramatically decreased when increased EU to 10wt% but it was steady with increasing EU 20wt%. Furthermore, tensile strength of EU-Si composite steadily decreased with increasing from 5wt% to 20wt% of EU in polymer composite. Regarding on SEM micrograph of EU-Si sample, it was noticed that the presence of voids of the EU-Si sample, as shown in Figure 1, may create stress concentration points, which in turn reduce the strength of the samples. Therefore, the mechanical properties of the EU-Si samples were poor than the EU-MA samples (Figure 2-3). It concluded that PE-g-MA coupling agent offer the better coupling agent over Silane coupling at higher content of EU flour.

The %elongation was presented in Figure3. It was found that %elongation was decreased with contents of EU in the composite. Moreover, %elongation of EU-MA samples slightly higher than EU-Si samples at all ratio. EU-MA with EU 5%wt gave higher elongation. It was concluded that EU-MA5 was suitable to use due to its high enough tensile strength and high %elongation.

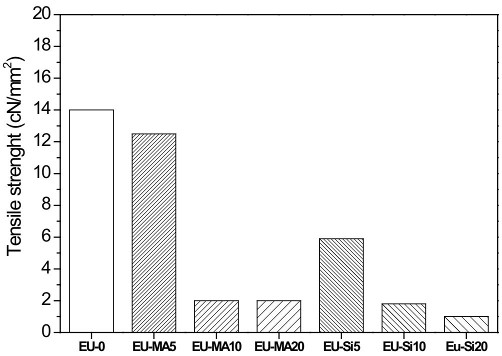

Figure 2. Tensile strength of HDPE and EU/HDPE composite with various content of EU from 5wt%, 10wt% and 20wt% in the present of PE-g-MA and Silane.

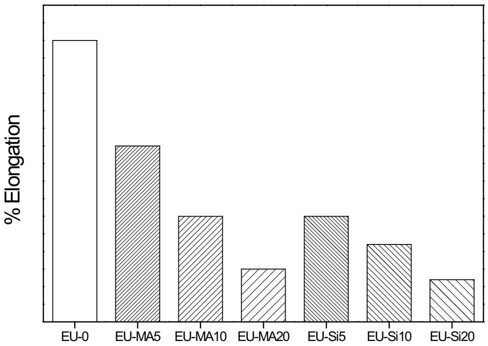

Figure 3. Elongation of HDPE and EU/HDPE composite with various content of EU from 5wt%, 10wt% and 20wt% in the present of PE-g-MA and Silane.

4 Conclusion

The wood plastic composites (WPCs) have been successfully investigated. The effect of the coupling agents, PE-g-MA (MA) and Silane (Si) on the thermal properties, and mechanical properties of polymer composite were also studied. The Eucalyptus flour was blend with the HDPE at 5wt%, 10wt% and 20wt%. The EU/HDPE composite slits were obtained from single screw extruder. The results showed that both of coupling agents significantly affect the mechanical, structure and thermal properties of the EU/HDPE composite. In additionally, the high content of Eucalyptus flour may affect the %crystalline of the polymer composite. The morphology of the EU/HDPE composite was also investigated by using SEM. It was noticed that the dispersion of fine Eucalyptus flour were observed with respected to the content of Eucalyptus flour. However, the PE-g-MA additive was presented the well mixing between Eucalyptus flour and polymer matrix.

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