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Recycled High Density Polyethylene/Ethylene Vinyl Acetate/Taro Powder Composites: The Effect of Polyethylene-grafted-Maleic Anhydride on Tensile Properties and Thermal Stability

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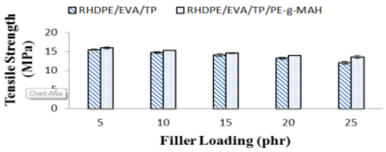
Graphical Abstract



Abstract

Taro powder (TP) filled composites based on recycled high density polyethylene (RHDPE)/ ethylene vinyl acetate (EVA) were prepared via melt blending method. Polyethylene-grafted-maleic anhydride (PE-g-MAH) was used as a compatibilizer to enhance interfacial bonding of RHDPE/EVA/TP composites. The effects of TP loading and the addition of PE-g-MAH on the tensile properties and thermal stability of RHDPE/EVA/TP composites were investigated and studied. Findings indicated that increased TP loading will reduce tensile strength and elongation at break while increasing the modulus of elasticity. The thermal stability of RHDPE/EVA/TP composites improved by increasing filler loading. With respect to both tensile properties and thermal stability, RHDPE/EVA/TP composites with PE-g-MAH performed better than the composites without the addition of compatibilizers.

Keywords: Polymer Composites, Recycled High Density Polyethylene, Ethylene Vinyl Acetate, Natural Fillers, Taro Powder, Polyethylene-grafted-Maleic Anhydride

Abstrak

Taro serbuk (TP) komposit diisi berdasarkan polietilena berketumpatan tinggi yang dikitar semula (RHDPE) / etilena vinil asetat (EVA) telah disediakan melalui leburan kaedah pencampuran. Polyethylene-dicantumkan-maleic anhidrida (PE-g-MAH) telah digunakan sebagai compatibilizer untuk meningkatkan ikatan antara muka komposit RHDPE / EVA / TP. Seterusnya kesan beban TP dan penambahan PE-g-MAH pada sifat-sifat tegangan dan kestabilan haba RHDPE / EVA / komposit TP telah disiasat dan dikaji. Dapatan kajian menunjukkan bahawa meningkatkan pengumpulan TP akan mengurangkan kekuatan tegangan dan pemanjangan pada takat putus manakala meningkat modulus keanjalan. Kestabilan terma komposit RHDPE / EVA / TP bertambah baik dengan peningkatan pengisi loading. Berkenaan dengan kedua-dua sifat tegangan dan kestabilan terma, komposit RHDPE / EVA / TP dengan PE-g-MAH prestasi yang lebih baik daripada komposit tanpa penambahan *compatibilizer*.

Kata kunci: Komposit Polimer, Polietilena dikitar semula berketumpatan tinggi, Ethylene vinyl acetate, Pengisi semula jadi, Serbuk Taro, Cantuman polyethylene-Maleic anhydride

1.0 INTRODUCTION

Polymer composites have been used to produce desired materials resulting in materials that are of outstanding physical properties [1]. A composite material is made by combining two or more materials with the main concept of existence of matrix materials and reinforcing fibers embedded in the matrix resin [2]. The main advantages of composite materials are its high strength to density and stiffness to density ratios, which give advantages to aircraft and aerospace industries [3]. Natural fiber reinforced composites have piqued scientific interests in the past decade since they offer both cost and weight reductions when compared to glass fiber composites, besides environmental advantages [4]. Natural fillers have several advantages such as environmental superiority, lower manufacturing energy, low density, low carbon dioxide emission, low cost, renewability, and biodegradability compared to inorganic-filler reinforced thermoplastic polymer composites [5]. A major problem encountered in polymer composites is the interface between hydrophilic natural fibers and hydrophobic polymer matrices [6]. With regards to this, compatibilization—the reducing of interfacial tension through an interfacial agent, compatibilizer—can be the most common way to achieve stabilization and reproducibility of the morphologies [7-9]. Compatibilizers will modify polymer interfaces by reducing the interfacial tension in the melt while stabilizing the dispersed phase, increasing the adhesion at phase boundaries and minimizing phase separation in solid state [10].

2.0 METHODOLOGY

2.1 Materials

RHDPE with melt flow index of 0.7g/10min (190°C) and density of 939.9kg/m^3 was used. EVA contains 18.1wt% VA, with melt index of 2.5g/10min (80°C, 2.16kg) and density of 0.93g/cm^3 was supplied from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia. Taro powder filler was obtained from a local village in Selangor. The ingredients of Taro are shown in Table 2.1. Polyethylene-grafted-maleic anhydride containing 0.85 wt% of maleic anhydride and dibenzoyl peroxide (DBP) with 75% of water was also obtained from A.R. Alatan Sdn. Bhd., Kedah Darul Aman, Malaysia.

2.2 Sample Preparation

For the TP preparation, the stems from Taro plants were cut, washed, dried and ground to powder by using a grinder. Taro fillers with average sizes of 75 µm were dried in a vacuum oven at 80°C for one hour. For composites preparation, the compounding of the blends was carried out by melt blending in a Brabender internal mixer. The RHDPE was first mixed in the internal mixer at 160 °C with a speed of 50 rpm for 2 min, followed by addition of EVA and mixed until homogenous. The compatibilizer, PE-g-MAH, dibenzoyl peroxide (DBP) and TP were added into the mixer for the remaining minutes. Each compounding was compression molded into sheets of 2 mm thickness using a hydraulic press at 160 °C for 6 min and cooled under pressure for 4 min. Table 2.2 shows the formulation used in this study.

2.3 Characterization and Measurements

Mechanical properties of the composites were measured from tensile test using Universal Testing Machine Instron 5582 with crosshead speed of 30 mm/min. Dumbbell shaped specimens were conditioned at ambient temperature before testing. Thermogravimetry analysis of the composites was done by using Perkin-Elmer Pyris 6 TGA analyser. Samples of about 10 mg were scanned from 50 to 650°C with a heating rate of 10°C/min by using constant nitrogen gas flow of 50ml/min to prevent thermal oxidation process of the polymer sample.

Table 2.1 Ingredients of Taro filler was tested from Laboratory Department of DXN Holdings Bhd., Jitra, Kedah.

Content	Quantity
Calories (Kcal)	274
Carbohydrate (%)	52.6
Fat (%)	1.2
Protein (%)	13.1

Table 2.2 Formulation of RHDPE/EVA/TP composites and RHDPE/EVA/TP/PE-g-MAH composites.

Composite Code	RHDPE [phr]	EVA [phr]	TP [phr]	PE-g-MAH [phr]	DBP [phr]
RHDPE/EVA/TP	80	20	5	-	-
RHDPE/EVA/TP	80	20	10	-	-
RHDPE/EVA/TP	80	20	15	-	-
RHDPE/EVA/TP	80	20	20	-	-
RHDPE/EVA/TP	80	20	25	-	-
RHDPE/EVA/TP/PE-g-MAH	80	20	5	6	1
RHDPE/EVA/TP/PE-g-MAH	80	20	10	6	1
RHDPE/EVA/TP/PE-g-MAH	80	20	15	6	1
RHDPE/EVA/TP/PE-g-MAH	80	20	20	6	1
RHDPE/EVA/TP/PE-g-MAH	80	20	25	6	1

3.0 RESULTS AND DISCUSSION

3.1 Tensile Properties

Figure 3.1 presents the effect of the filler loading and compatibilizer on tensile strength of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites. Both composites progressively decreased with increasing filler loading. This was due to the filler content and increasing agglomeration of the filler particles, consequently reducing the tensile strength of the composites. Quiroz-Castillo *et al*. reported that tensile strength showed a decreasing trend with the increase of chitosan content because of the brittleness of chitosan, which reduced the ductility [11]. Adrikha & Shapovalov revealed that the addition of micro-sized fillers introduced weak interfaces because of poor wetting and negligible adhesion that resulted in reducing the tensile strength proportional to the filler content [12].

By comparing both graphs, with similar TP loading, RHDPE/EVA/TP/PE-g-MAH composites exhibited higher tensile strength than RHDPE/EVA/TP composites. This can be explained by the presence of PE-g-MAH, which made easy the transfer of slight stress at the interfacial adhesion between the matrix and filler through the deformation. The accumulation of compatibilizer enhanced the interfacial adhesion of the matrices while providing interaction between anhydride groups of maleic anhydride to the hydroxyl groups of TP, thus improving matrix-filler interaction.

Figure 3.2 exhibits elongation during breaking of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites. The elongation at break of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites decreased as filler loading increased. This can be attributed to the restriction of elastic properties for the composites with reduction in matrix due to the increase of fillers. The deformation of TP is generally less than RHDPE/EVA matrix, therefore the addition of filler forces the matrix to deform more than the total deformation of the composites.

From Figure 3.2, elongation at break for RHDPE/EVA/TP composites is higher than RHDPE/EVA/TP/PE-g-MAH composites. The results were explained by high stiffness of filler than matrix which can increase the tensile modulus but commonly cause a significant reduction in elongation at break of the composites. The decreasing trend of elongation at break for RHDPE/EVA/TP/PE-g-MAH composites was a recognizable sign of improved interfacial adhesion between RHDPE/EVA phases and TP filler after the addition of PE-g-MAH as compatibilizers.

The effect of filler loading and compatibilizer on modulus of elasticity of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites was denoted in Figure 3.3. Both graphs of the composites revealed an increase in modulus of elasticity in a manner proportional to the TP content. As TP is stiff in character, an increase in TP content will result in a decrease in ductility of the composites, hence increasing tensile modulus. On the other hand, Adhikary *et al*. reported that tensile modulus of HDPE/wood flour increased by increasing the fiber content due to the composite material beccoming stiffer [13].

In comparison between uncompatibilized and compatibilized composites, RHDPE/EVA/TP/PE-g-MAH composites displayed higher modulus of elasticity than RHDPE/EVA/TP composites at different filler loading. The addition of PE-g-MAH was preferentially located at the interface between the two polymers and resulted in lowering the interfacial tension; improving the compatibility of TP and the matrix. Improvement in stiffness of the composites affects the modulus of elasticity values of both composites. According to Ismail *et al*., addition of white rice husk ash (WRHA) to NR/LLDPE blends will increase the tensile modulus and higher tensile modulus will be achieved by addition of poly-(propylene-ethylene-acrylic acid) (PPEAA) as compatibilizer, which improves the filler-matrix bonding by efficient stress transfer from the matrix to the filler phases [14].

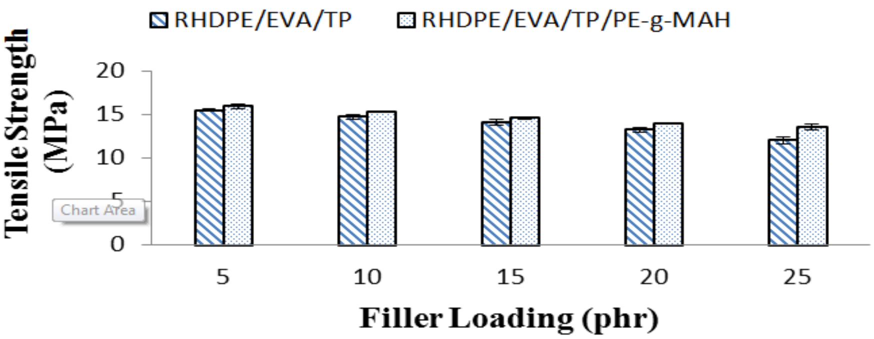


Figure 3.1 Effect of filler loading and compatibilizer on tensile strength of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites

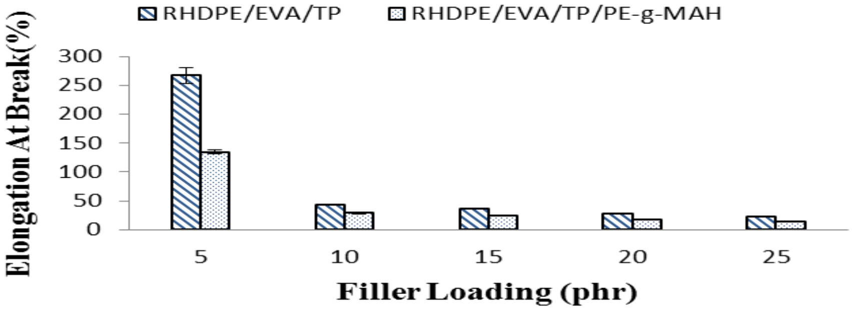


Figure 3.2 Effect of filler loading and compatibilizer on elongation at break of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites

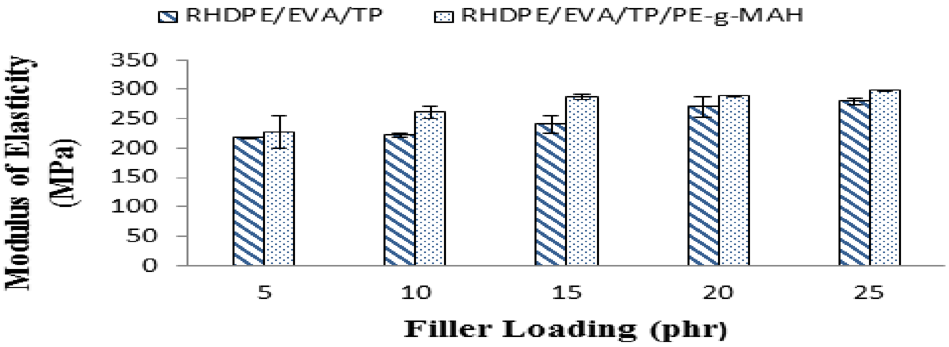


Figure 3.3 Effect of filler loading and compatibilizer on modulus of elasticity of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites

3.2 Thermal Degradation

Figure 3.4 shows the typical thermal degradation curves of RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites with different filler loadings. Table 3.1 shows that the temperature of 50 % weight loss and residual mass for RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites increased with the increasing of filler loading. This result indicated that higher filler loading gives the RHDPE/EVA/TP composites more thermal stability than lower filler loading.

From Table 3.1, it can be seen that temperature of 50 % weight loss (T-50%wt) and residual mass of RHDPE/EVA/TP/PE-g-MAH composites are higher than RHDPE/EVA/TP composite. This was due to the presence of good interfacial adhesion between RHDPE/EVA and TP phase as a result of uniform dispersion of fillers throughout RHDPE/EVA matrix. The better dispersion of filler acts as a barrier against the release of volatile gases during thermal degradation. It may also be attributed to an adsorption effect of these gases at the filler surface, which slows down polymer degradation [15].

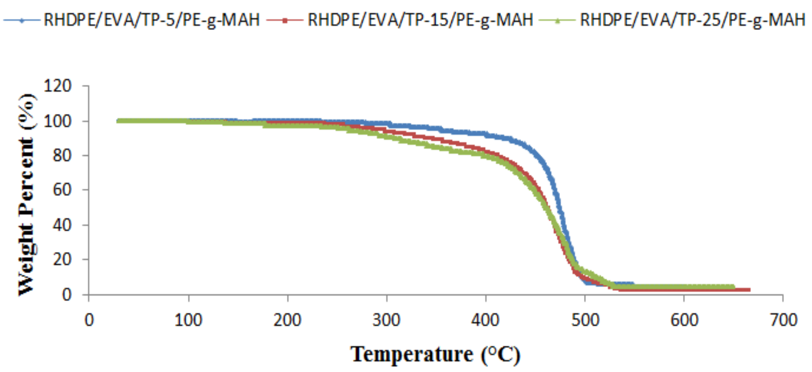


Figure 3.4 Thermogravimetric curves of RHDPE/EVA/TP/PE-g-MAH composites at different filler loading

Table 3.1 Temperature of 50% weight loss (T-50%wt) and residual mass for RHDPE/EVA/TP and RHDPE/EVA/TP/PE-g-MAH composites

Blend Composition	$T_{-50\%wt}$ (°C)	Residual Mass (%)
RHDPE/EVA	440.12	2.051
RHDPE/EVA/TP-5	475.57	2.100
RHDPE/EVA/TP-15	475.82	4.299
RHDPE/EVA/TP-25	476.29	8.059
RHDPE/EVA/TP-5/PE-g-MAH	475.86	3.501
RHDPE/EVA/TP-15/PE-g-MAH	476.03	5.111
RHDPE/EVA/TP-25/PE-g-MAH	476.21	8.947

4.0 CONCLUSION

The increase in filler loading for taro powder in RHDPE/EVA/TP composites reduced tensile strength and elongation at break while increasing modulus of elasticity. The addition of PE-g-MAH as a compatibilizer improved interfacial adhesion, thus, improving tensile strength and modulus of elasticity while decreasing elongation at break. Both the addition of taro powder and compatibilizer has enhanced the thermal stability of RHDPE/EVA/TP composites.

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