

The Use of Agro Waste *Alpinia galanga* Fibers as Reinforcement in Polyethylene Composites

[M. Awang, Rohani. M., Asmadi A., Mohd Aidil A. A. & Nora'aini A.]

Abstract— Employing fibers particularly those from renewable materials as an alternative to inorganic fibers in composites is part of the effort to promote bio-based products as well as to broaden recycling of solid wastes. In this work, the use of agro waste *Alpinia galanga* (AG) as composite reinforcement was investigated in terms of tensile properties, thermal stability, morphology and water absorption. Composites based on AG fibers and high density polyethylene (HDPE) were prepared together with an additive, eco-degradant (ECO) and a compatibilizer, polyethylene-g-maleic anhydride (PE-g-MA) in a mixer, Brabender Plastograph EC at various AG fiber loadings. The results reveal that the composite formulation enabled AG fibers to function as reinforcement in HDPE-based composites. The presence of ECO at 15 wt% fiber loading improved tensile strength up to 33.6 MPa, about 21 % higher than that of pristine HDPE (27.8 MPa). The addition of MA-g-PE at 10 wt% fiber loading also indicates comparable tensile strength to pristine HDPE of about 27.5 MPa. The Young's modulus of AG/HDPE composites with the addition of ECO was enhanced up to 4874 MPa as compared to 1012 MPa of pristine HDPE. The elongation at break of the AG/HDPE composites however is somewhat lower than that of pristine HDPE. The water absorption of AG/HDPE composites with ECO at 15 wt% fiber loading was improved to 0.96 % as compared to 1.40% of the AG/HDPE composites. The proposed formulation containing ECO and PE-g-MA also slightly improved thermal stability of AG/HDPE composites. Scanning electron microscopy examination indicates a much better adhesion between AG fibers and HDPE matrix with the presence of ECO. The composite formulation with ECO, an environment-friendly additive which helps degrade polyolefin composites to the basic elements with no harmful effects contributed to noticeable improvements in properties of AG/HDPE composites.

Keywords—Agro waste, *Alpinia galanga*, composites, eco-degradant, properties

I. Introduction

Over the last few years, the demand on the use of inorganic fibers as reinforcement or filler in polymer composites particularly from renewable resources as a viable alternative to synthetic fiber i.e. glass, carbon and aramid have accelerated as it meets the need that confront environmental friendly characteristics. Natural fibers such as kenaf, hemp, baggase, cotton, pineapple and palm fibers have an edge over synthetic fibers because they are abundantly available, low in density, less toxic and recyclable [1-2]. They also have high specific strength and modulus suitable for applications such as in the automotive, building, packaging as well as furniture industries [3-4]. The incorporation of natural fibers in thermoplastic based composites may assist to minimize the dependence on synthetic fibers generally derived from petroleum and thus help to reduce the amount of plastic used and reduce the cost of resulting products. Moreover, natural fibers are mostly obtained from vegetal biomass. Therefore, the use of natural

fibers can promote recycling activity and cause less environmental pollution, particularly on solid waste disposal problems and air pollution.

In general, natural fibers consist of cellulose, hemicellulose together with matrix such as lignin or pectin. These constituents may differ from plant to plant depending on the origin, climate, age, part of plants and soil conditions [5-6]. Various natural fibers have been studied by many researchers as reinforcement or filler in thermoplastic or thermoset composites. However, little attention was paid on the potential of *Alpinia galanga* (AG) fibers in polyethylene based composites. AG fibers are derived from agricultural waste from galanga cultivation and this plant is locally available and can be found in other countries such as in China, India, Thailand and Indonesia.

As inferred by other researchers, the main contributor of the strength of natural fiber is cellulose [7]. However the cellulose has disadvantages mainly due to its hydrophilic properties that make them incompatible with non-polar polymer matrix, thus weaken the tensile properties of the composites. Therefore, the modification or treatment either chemically or physically must be employed to improve interfacial surface adhesion between fiber and polymer matrix.

Polyethylene-g-maleic anhydride (PE-g-MA) is a common compatibilizer used to improve the compatibility of the fiber and matrix. As reported in other work, micrograph images indicated the effectiveness of PE-g-MA as a compatibilizer to improve the interfacial bonding between hydrophilic fibers and hydrophobic polymer matrix [8-9]. The positive enhancement in the adhesion of fibers and matrix was also reported with incorporation of eco degradant [10]. Eco degradant (ECO) is an additive that has environmental advantages as it can degrade polyolefins to basic elements with no harmful residue. Therefore, the polyolefin with ECO has biodegradability properties.

Hence, in this research work, the *Alpinia galanga* fiber-high density polyethylene (AG/HDPE) composites with and without addition of PE-g-MA and ECO were prepared. The effects of addition of PE-g-MA and ECO on tensile properties, morphology, thermal and water absorption were studied.

M. Awang, Rohani. M., Asmadi A., Mohd Aidil A. A. & Nora'aini A
Universiti Malaysia Terengganu
Malaysia

II. Materials and Methods

A. Materials

HDPE resin with a density of 0.957 gm/cm³ and melt flow index of 4 g/min at 190°C was supplied by Polyethylene Malaysia Sdn. Bhd., Malaysia. AG stalks were obtained from local sources in Terengganu, Malaysia and the fibers were extracted manually. The fibers were then dried at 80°C for 8 h prior to cut to the average length of 15 mm. The compatibilizer, PE-g-MA was purchased from Sigma-Aldrich (Malaysia). An additive, ECO was procured from Behn Meyer Polymer Sdn. Bhd.

B. Preparation of AG/HDPE Composites

AG/HDPE composites with and without the addition of ECO or PE-g-MA was prepared via melt blending method using Brabender Plastograph EC mixer at 135°C and 50 rpm. HDPE resin was charged first and melted for 2 min. Then, AG fibers at different fiber loadings at 3, 6, 10 and 15 wt% were charged and blended for another 4 min. The compatibilizer or additive at 5 phr (based on weight of HDPE) was added during the blending process and mixing was continued until the total mixing time of 10 min. The composite formulation is described in Table 1. Composites were then taken out after the completion of mixing process and were cooled down prior to molding into dumbbell shape specimen using an injection molding machine.

TABLE I. TABLE TYPE STYLES

Materials	AG/HDPE	AG/HDPE+PE-g-MA	AG/HDPE+ECO
AG	0, 3, 6, 10, 15	0, 3, 6, 10, 15	0, 3, 6, 10, 15
HDPE	100, 97, 94, 90, 85	100, 97, 94, 90, 85	100, 97, 94, 90, 85
MAPE	-	3	-
ECO	-	-	3

C. Analysis and Testing

Tensile properties of AG/HDPE with and without PE-g-MA and ECO were studied using universal testing machine Instron 3366 in accordance with ASTM D638. Five replicates of samples were tested for every composite formulation. A crosshead speed and gauge length used was 10 mm/min and 35 mm, respectively. The morphology study of fractured surfaces of the composites was evaluated using Scanning Electron Microscopy (SEM-ISM-6380LA) with an accelerating voltage of 5 kV. The thermal stability analysis of composites was carried out using Mettler Toledo TGA (SDTA851). Water absorption study was performed using type v injection-molded dumbbell samples. The initial readings of sample weight were taken and then samples were soaked in distilled water for a specific interval time, after that were taken out and reweighed. The calculation of the percentage of water absorption is as follows:

$$\text{water absorption (\%)} = \frac{M2 - M1}{M1} \times 100 \quad (1)$$

where M1 and M2 were the initial weight and final weight of sample after immersion, respectively.

III. Results and Discussion

A. Tensile Properties

Figure 1 shows the effects of addition of PE-g-MA and addition of ECO in AG/HDPE composites on tensile strength, Young's modulus and elongation at break, respectively. It can be seen that the tensile strength of the composites generally increases with the increase of AG fiber loading up to 15 wt%. However, the tensile strength of AG/HDPE composites is lower than that of AG/HDPE composites with addition of PE-g-MA and addition of ECO. This occurs because of the lack interaction and adhesion between AG fiber and HDPE matrix. This result is consistent with the SEM findings in Figure 4 indicating poor interaction due to the existence of gaps between AG fibers and HDPE matrix. The AG/HDPE composites with addition of PE-g-MA and addition of ECO showed improved tensile strength. A maximum tensile strength was observed at 15 wt% for composites with addition of ECO with the tensile strength of 33.6 MPa which is 20.9% higher than that of pristine HDPE (27.8 MPa). These results show that the addition of ECO has been found to be beneficial in increasing the tensile strength even at low fiber loading (3 wt%). This is attributed to the establishments of strong interaction between ECO with the AG fiber and HDPE. A similar observation was reported by Ismail *et al.* (2009) [10], when incorporating ECO in polyethylene/chitosan composites.

The composites with the addition of PE-g-MA also showed an improvement in the tensile strength value, but the increase is minimal. The tensile strength of these composites was close to those of pristine HDPE at most fiber loadings. On the basis of these results, the incorporation of PE-g-MA and ECO promotes the increment in tensile strength of AG/HDPE composites to some extent due to the improvement in the interfacial bonding between AG fibers and HDPE matrix.

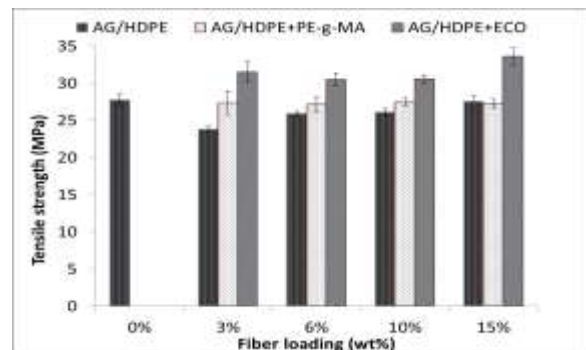


Figure 1. Tensile strength of AG/HDPE composites without and with addition of PE-g-MA and addition of ECO.

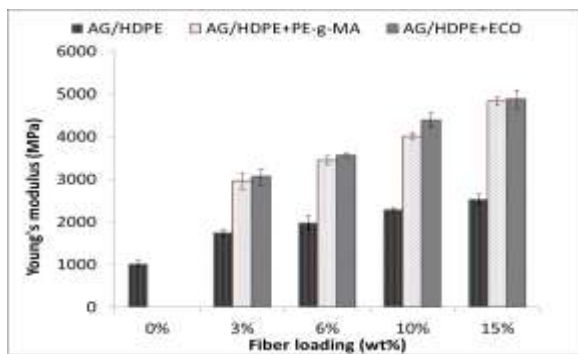


Figure 2. Young's modulus of AG/HDPE composites without and with addition of PE-g-MA and addition of ECO.

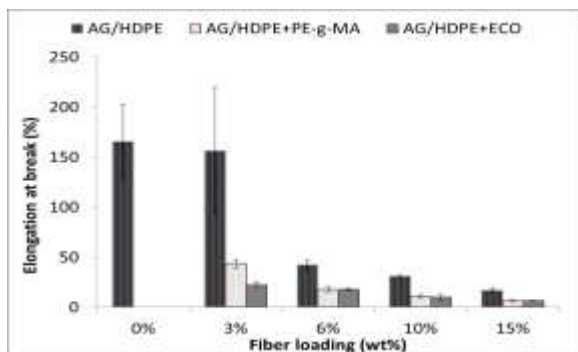


Figure 3. Elongation at break of AG/HDPE composites without and with addition of PE-g-MA and addition of ECO.

As depicts in Figure 2, the Young's modulus of AG/HDPE composites are higher than pristine HDPE and increases with increasing fiber loadings. The increase could be due to the stiffening effect of AG fibers on the HDPE matrix. This is consistent with several findings which also indicate that the inclusion of cellulosic fibers in polymer matrix enhances the stiffness of the composites^[11-12]. The Young's modulus value of AG/HDPE composites was further improved with addition of PE-g-MA and with addition of ECO and the improvement is up to 4874.2 MPa and 4832.9 MPa which is 381% and 377% higher than that of pristine HDPE (1012.6 MPa), respectively. Minimal difference was noticed in the Young's modulus value for AG/HDPE composites with addition of PE-g-MA and composites with addition of ECO.

For elongation at break, a significantly lower value was observed with the inclusion of AG fibers than that of pristine HDPE. On the contrary, the elongation at break is reduced with increasing fiber loading as shown in Figure 3. As reported in most findings, the inclusion of cellulose fibers would rigid interface and reduce the ductility of the composites^[13-14].

B. Morphological Study

Figure 4 shows the micrograph of the tensile fracture surface of AG/HDPE composites without and with the addition of PE-g-MA and the addition of ECO. From the Figure 4(a), it's clearly seen that the AG fiber is loosely embedded in HDPE matrix and there are a few areas spotted the fiber pull outs from the HDPE matrix. This proved the

poor adhesion and interaction between AG fibers and HDPE matrix as there is no modification or chemical treatment involved in the preparation of composites. Meanwhile, the AG/HDPE composites with the presence of PE-g-MA and ECO exhibited an improvement in surface adhesion between the fiber and matrix as shown in Figure 2(b) and 2(c), respectively.

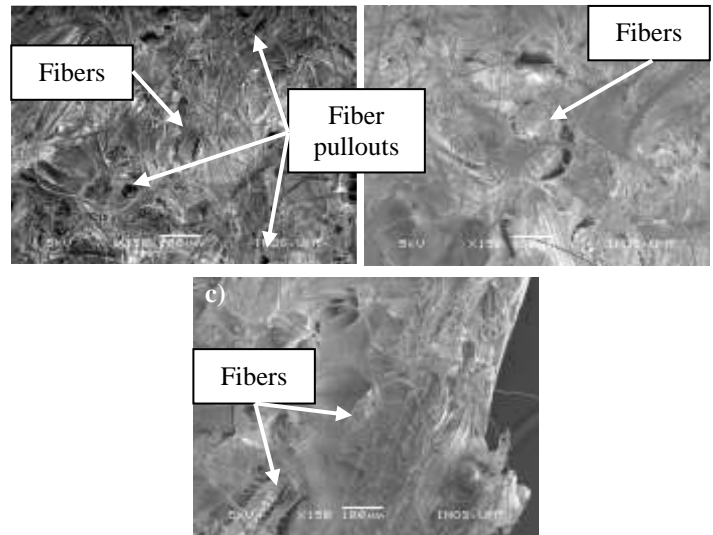


Figure 4. SEM micrograph of tensile fracture surface of a) AG/HDPE, b) AG/HDPE with addition of PE-g-MA and c) AG/HDPE with addition of ECO composites at 15 wt% under x150 and magnification.

C. Thermal Analysis

The thermogram of AG/HDPE shows that pristine HDPE has higher thermal stability than AG/HDPE composites as illustrated in Figure 5. The inclusion of AG cellulose-based fibers decreased the thermal stability of composites. As explained in other findings, celluloses will degrade at high temperature, thereby causing a reduction in the thermal stability of the composites^[11-12]. Therefore, higher fiber loading composites (15 wt%) had lower thermal stability than lower fiber loading composites (3 wt%). The addition of PE-g-MA and the addition of ECO enhanced slightly the thermal stability of AG/HDPE composites. When the 5% of weight loss is selected as a comparison point, the decomposition temperature of AG/HDPE composites with addition of PE-g-MA and ECO is approximately 2 to 3% higher than that of AG/HDPE composites, respectively.

TABLE II. FORMULATION OF ALPINIA GALANGA/HIGH DENSITY POLYETHYLENE (AG/HDPE) COMPOSITES WITH PE-G-MA/ECO

Samples	Fiber loading (wt %)	Temperature at 5 % weight loss (°C)
HDPE	-	353.0
AG/HDPE	3	317.6
AG/HDPE	15	285.1
AG/HDPE+PE-g-MA	15	290.0
AG/HDPE+ECO	15	292.4

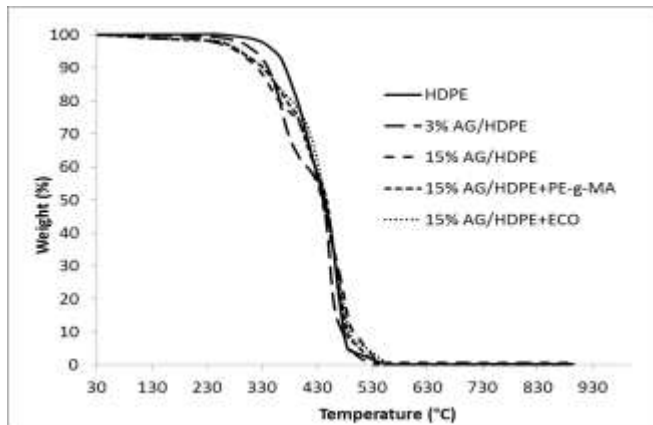


Figure 5. TGA thermograms for AG/HDPE composites without and with addition of PE-g-MA and addition of ECO

D. Water Absorption Study

Water absorption of AG/HDPE composites with and without addition of PE-g-MA and addition of ECO is depicted in Figure 6. From the graph, it is clearly seen that the introduction of AG fibers into HDPE matrix increased the water absorption. Water absorption for AG/HDPE composites at 3 wt% fiber loading and pristine HDPE was about 0.4% and 0.09%, respectively. The higher fiber loading composites (15 wt%) have higher water absorption than that of 3 wt% fiber loading composites. PE-g-MA and ECO modified composites show a reduction in water absorption rates of about 0.96 and 0.89%, respectively as compared to 1.40% water absorption of AG/HDPE composites. This might be due to a better interfacial bonding between AG fibers and HDPE matrix as shown in SEM micrograph (Figure. 4(a & b) which hinders the rate of absorption of water.

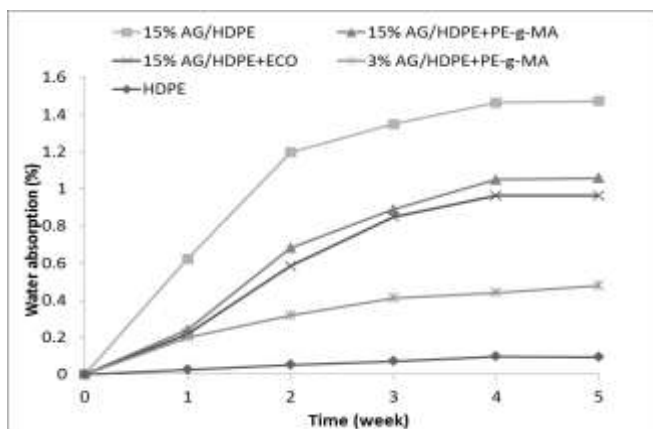


Figure 6. The water absorption of AG/HDPE composites without and with addition of PE-g-MA and addition of ECO

iv. Conclusion

The incorporation of PE-g-MA and ECO exhibited better improvement in tensile strength, Young's modulus, thermal

and water absorption than that of AG/HDPE composites. The tensile strength of AG/HDPE with addition of ECO improved up to 33.6 MPa which is 20.9% higher than that of pristine HDPE (27.8 MPa). The highest tensile strength of composites with PE-g-MA at 15 wt% fiber loading was about 27.5 MPa comparable to that of pristine HDPE. The addition of PE-g-MA and ECO minimally improved thermal stability and reduced water absorption of the composites. SEM results support the findings indicating the presence of better interfacial adhesion with the incorporation of PE-g-MA and ECO.

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About Author (s):



Mohamad Awang is a lecturer at the School of Ocean Engineering, Universiti Malaysia Terengganu (UMT), Malaysia.