



Effect of polymer blend matrix compatibility and fibre reinforcement content on thermal stability and flammability of eco-composites made from waste materials



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ABSTRACT

This paper was aimed to evaluate the effects of matrix types (with or without ethylene-glycidyl methacrylate compatibilizer) and rice husk (RH) loadings (40–80 wt.%) on RH-reinforced recycled high-density polyethylene/recycled polyethylene terephthalate (rHDPE/rPET) eco-composites. Results showed that the thermal stability and flammability resistance properties increased as the RH loadings increased. The addition of RH has effectively delayed the thermo-oxidation process of rHDPE/rPET matrix by 10 °C, and evident flame retardant effect has also been observed (decreased burning rate up to 24% in comparison to neat polymer blend). It is interesting to note that compatibilization of polymer blend matrix has further increased the thermal stability of eco-composites. SEM images confirmed the enhanced interfacial bonding of phases in the compatibilized matrix eco-composites. It can be concluded from this study that the used agro-waste material (RH) is attractive reinforcements in recycled plastics from the standpoint of their thermal and flammability properties.

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1. Introduction

During the period of 2010–2013, the average annual global rice production was 725 million metric tons and Asia region alone accounted for more than 90% of the total global rice production. Rice husk (RH) is an inexpensive by-product of rice processing where it is separated from rice grain during the rice milling process, and it is reported that, there is about 0.23 tons of RH is produced for every ton of rice produced [1]. The use of RH in the manufacture of composites panels provides advantageous characteristics, such as low density, biodegradability, toughness, resistance to weathering, and also making the final products more economically competitive. RH fibre-reinforced thermoplastic composites are now being commercialised in the green furniture, building construction for interior components, window and door frame, wall, partitioning and panelling industries as well as automotive industry [2].

The literature review reveals that PE, polypropylene (PP), polyvinyl chloride (PVC), polylactic acid (PLA) are among popular

choices as matrices for RH filled polymer composites [1]. However, the application of RH in thermoplastic composites is restricted by its hydrophilic nature and low thermal stability [3]. Because of polymers are mostly nonpolar (hydrophobic), the weak compatibility (polarity) between RH and polymer matrix must be solved to avoid the fiber-fiber agglomeration and bad mixing of composite materials. Polymer modification with a coupling agent containing polar groups, such as, maleic anhydride is commonly used to improve the matrix-fiber interfacial adhesion [4]. In fact, most of the natural fibers including RH have a relatively low degradation temperature of about 200 °C, which limits the processing temperature of composites and thus the melting point of polymer is an important consideration in choosing it as matrix materials for RH composites [5].

Thermal properties are crucial to understand the behaviour of the raw materials (either polymer or filler) and interfacial characteristics the end composite material [6]. Different thermal analysis techniques provide different essential information regarding the thermal stability of composites. Thermogravimetric analysis (TGA) can be used to determine the moisture content, thermal degradation temperature and thermal stability of composite materials. Differential scanning calorimetry (DSC) can be used for determining

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glass transition temperature (T_g), melting temperature (T_m), melting enthalpy (ΔH_m) and crystallinity level (χ_c) [7]. Flammability is an important criterion in material selection for consumer product, building and construction applications. Generally, synthetic polymers are highly flammable because of their petroleum source. However, Zhang et al. reported that addition of rice husk hold the potential to suppress the flammability of HDPE owing to the high content of silica present in RH [8].

In our previous research regarding on using recycled high-density polyethylene (rHDPE) and recycled polyethylene terephthalate (rPET) and RH, the investigation of physical, mechanical and thermal properties of RH-filled rHDPE/rPET biocomposite extruded via single screw and twin screw extruder [9], the effect of RH filler loading and matrix types (uncompatibilized and compatibilized rHDPE/rPET blend) on RH-filled rHDPE/rPET biocomposites on water absorption and mechanical properties [10] and the effect interfacial modifications between blend matrix and RH fillers on water absorption and mechanical properties [2] have been discussed. In spite of these studies, the thermal analysis and flammability properties on RH-filled recycled HDPE/PET blend with respect to the presence of compatibilizer in immiscible polymer matrix has not been discussed deeply. The objectives of the study were to investigate the effect of matrix types (recycled polymer blend with and without compatibilizer) and RH content (40–80 wt.%) and on the thermal properties and flammability behaviour of the RH-incorporated HDPE/PET ecomposites. The correlation between experimental and theoretical results for thermal properties are reported.

2. Experimental

2.1. Materials

Recycled high-density polyethylene (rHDPE; density of 923 kg/m³, melt flow index of 0.72 g/10 min at 190 °C, 2.16 kg load) and recycled polyethylene terephthalate (rPET; T_g of 74.1 °C, cold crystallization peak temperature of 119.9 °C, melting peak temperature of 252.5 °C and intrinsic viscosity of 0.68 dL/g) were obtained from a local plastic recycling plant. To improve the compatibility between immiscible polymer blend components of rHDPE and rPET, ethylene-glycidyl methacrylate copolymer (E-GMA) with a trade name of Lotader AX8840 (melt flow index of 5 g/10 min at 190 °C, 2.16 kg load and a glycidyl methacrylate content of 8%) was used as compatibilizer. Rice husk (RH) with particle size of 100-mesh was used as agro-filler in the experiment. Maleic anhydride polyethylene (MAPE) with a melting peak temperature of 135.2 °C was utilized as coupling agent. All the raw materials were supplied from a local factory namely BioComposites Extrusion Sdn. Bhd.

2.2. Preparation of RH filled ecomposites

The rHDPE and rPET were compounded using a laboratory scale co-rotating twin screw extruder (model Thermo Prism TSE 16PC, $D = 16$ mm, $L/D = 25$). The screw rotating speed was fixed at 30 rpm. The four barrel temperatures from the feeding to die zones were set as 250, 270, 240 and 190 °C, respectively. The uncompatibilized polymer blend with the weight ratio of rHDPE/rPET at 75/25 (wt/wt) is labeled as rPB. Meanwhile, the compatibilized polymer blend with the same ratio of both plastics and 5% of E-GMA (based on the total weight of blend) is referred as rPB/E-GMA. The pre-extruded polymer blend (rPB and rPB/EGMA, respectively) pellets were melt-blended with RH and 3% of MAPE at temperatures profiles 170, 215, 210 and 195 °C with the same screw speed as extrusion of recycled polymer blend. The loading level of RH was

varied at 40, 50, 60, 70 and 80 wt.%. In order to remove the trapped moisture inside RH flour, it was oven-dried at 90 °C for 24 h before extrusion. After extrusion, the extrudates were cooled and granulated into pellets by a crusher. The fine granules were compression molded at 200 °C under a pressure of 1000 psi by using a hot/cold press machine (LP50, LABTECH Engineering Company). The pre-heating, venting, full pressing and cold pressing times were set to 3, 2, 5 and 5 min, respectively.

2.3. Thermal analysis

Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) was conducted using a Mettler Toledo TGA/SDTA851^e and DSC 882^e, respectively, on the samples of about 10–15 mg. Samples of TGA were tested at a heating rate of 10 °C/min over the temperature range from 25 °C to 600 °C, the temperature of complete degradation. While the DSC samples were scanned from 25 °C to 300 °C at heating rate of 10 °C/min, under atmospheric air flow condition. The melting temperature (T_m) values were taken as the maximum of the endothermic melting peak. In order to confirm the moisture content of raw and dried RH determined by TGA, moisture analysis was carried on both RH using moisture analyzer (MS-70, A&D Company).

2.4. Flammability properties

Burning test was performed in accordance to ASTM D 5048-90 (Procedure A – test of bar specimens) to determine the relative burning characteristics and flame resistance properties. The burning rates of specimens are calculated using the Eq. (1):

$$V = 60 \frac{L}{t} \quad (1)$$

where V is the burning rates (mm/min), L is the burned length (mm), t is the burning time (seconds). All the reported results of the burning tests are the average of five replicates for each formulation.

2.5. Scanning electron microscopy (SEM)

The morphology of the fracture surface of broken sample from tensile testing was analysed using SEM (VPSEM Philips XL-30). The samples were sputter-coated with gold before examination of SEM at 2000 \times .

3. Results and discussion

3.1. TGA

The thermo-oxidative degradation of rPB and rPB/EGMA ecomposites with different RH loadings was studied using TGA under atmosphere air condition. The corresponding TGA curves are illustrated in Fig. 1(a) and (b). As can be seen in Fig. 1, the neat polymer blend matrix (rPB and rPB/EGMA samples without RH filler) experienced a dramatic weight loss through one-step degradation process from 400 °C to 500 °C. This result can be explained that neat polymer blend comprises a series of interchained small molecular species which easily undergo thermal decomposition at elevated temperature, which results in the formation of small-molecule radicals with a great activity, capable of initiating various reactions of macromolecules, first of all their thermo-oxidative degradation, depolymerization and destruction that preferably occur at the weak sites of the polymer chains. The maximum decomposition temperature of the rPB and rPB/EGMA samples was at 471 °C and 469 °C, respectively (Table 1). The lower decomposition temperature for rPB/EGMA than that of rPB was due to an increase of rHDPE-rPET interaction as a result of the incorporation of EGMA compatibilizer

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