Development of Recycle-Board by Spinning of Plastics and Blending with Wastepapers. 1: Influence of Processing Methods

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Abstract: Composites consisting of a high-density polyethylene (HDPE) and wastepaper powders were prepared by two different processing methods: the spinning-press method and the roll mixing method. The influence of the processing method on mechanical properties of wastepaper/HDPE composites was studied. The compatibilizing effect of a maleic anhydride-grafted polyethylene (MAHPE) between the wastepaper and HDPE was also investigated. It was found that the flexural properties of spinning-pressed composites were lower than those of roll mixed composites, but spinning-pressed composites showed improved fracture toughness than roll mixed composites. Mechanical measurements and the scanning electron microscopy (SEM) showed that the addition of 5 to 10wt% MAHPE into the HDPE matrix was effective to enhance the mechanical properties of wastepaper/HDPE composites and to improve interfacial adhesion between the wastepaper powders and HDPE matrix.

Keywords: Wastepaper/polyethylene composite, Maleic anhydride-grafted polyethylene, Processing, Flexural properties, Fracture toughness

1. INTRODUCTION

The expanding use of thermoplastic polymers, for example, polyethylene, polypropylene, polystyrene, poly(acrylonitrile-butadiene-styrene), and so on, in wide production areas is an indisputable fact nowadays. Although these polymers have the advantage of low cost, low density, and processability, these are nondegradable in the natural environment. Accordingly, disposal of these polymers after their intended use, so-called plastic wastes, is a serious problem in the world.

In the last decade, a great deal of attention has been focused on recycling of plastics; the material recycling, the chemical recycling, and the thermal recycling. In the view point of energy-saving, resources-saving, and waste reduction, the material recycling is presumably most effective among these recycling methods [1].

In the field of composites, the fiber reinforcement of thermoplastic matrices was initially developed using man-made fibers such as glass, carbon, aramid, etc. in order to take the advantage of their high specific strength and modulus [2]. In recent years, however, a large number of studies have been devoted to fibers of vegetable origin, with the scope of replacing man-made fibers [3]. The use of natural fibers, such as cellulose, sisal, jute, kenaf, bamboo, offers several benefits, including low cost, high specific stiffness and strength, and biodegradability. The wastepaper made from mainly cellulose fibers is one of the most abundant materials among the paper products, therefore, it has attracted much research interest. Many attempts have been made to use wastepaper [3] and plant fibers [4, 5] as reinforcement for thermoplastic polymers.

The aim of the present study is to prepare and characterize the wastepaper/polyethylene composites. Composites were prepared by the spinning-press and the roll mixing methods, and influence of the method on mechanical properties was investigated. Moreover, to increase the fiber-matrix interfacial adhesion, a maleic anhydride-grafted polyethylene was employed as a compatibilizer, and its effect on mechanical properties of the composites was investigated.

2. EXPERIMENTAL

2.1. Materials

The polymeric matrix used in this study was a commercially available high-density polyethylene (HDPE), HIZEX 2100J (supplied by Mitsui Chemicals, Inc.). It has an MFR of 6 g/10min, a density of 0.956 g/cm³ and a melting point (T_m) of 127°C. The wastepaper was obtained in the form of waste newspaper. The waste newspaper was grounded into wastepaper powders, as shown in Fig. 1, by the grinder made by Topia. A maleic anhydride-grafted polyethylene (MAHPE) was obtained from Sigma-Aldrich.

Fig. 1. Optical photograph of wastepaper powders. The scale is graduated in millimeters (1mm increments).
2.2. Preparation of wastepaper/HDPE composites

The wastepaper/HDPE composites were prepared by two different processing methods, viz. (1) spinning-press and (2) roll mixing. In the spinning-press method, HDPE was blended with MAHPE as a compatibilizer to the specified concentration of 0, 3, 5 or 10wt% and mixing was performed in a twin-screw extruder (Technovel Corp., KZW 15-30 MG) with a screw diameter of 15mm and an L/D ratio of 30/1. The operating temperature was 170-175°C. The HDPE/MAHPE blended compound was extruded, from a single-nozzle die (3mm diameter), as a mono-filament with a diameter of about 0.18mm. After the melt spinning, obtained mono-filaments were chopped into short fibers with about 15mm long. The wastepaper powders were added to short fibers. The mixture proportion of wastepaper, PE, was 50:50 by mass. Then, the composite was compression molded at about 11MPa pressure and at 160°C in a parallel plate press for 5min.

In the roll mixing method, HDPE was blended with MAHPE as a compatibilizer to the specified concentration of 0, 3, 5, 10 or 20wt% and precompounding was carried out in a two-roll mill (Eto Seisakusyo Co., Ltd.) at 170°C for 3min. Once the polymer was melted, wastepaper powders were added (50% by weight), and mixed with the polymer at 170°C for about 5min. The mixture was preheated by compression molding at 180°C for 15min. Then, the compound was compression molded at 2.2MPa pressure and at 180°C in a parallel plate press for 1min. Finally, the composite plate of 2mm thickness was compressed by another mold at 25°C.

2.3. Mechanical Test

Samples for the flexural tests were cut to the size of 50mm x 15mm x 2mm. The flexural properties of composites were investigated by the tensile tester (Tensilon UTM-4-100, Orientec Co.) with a crosshead speed of 1mm/min according to JIS K7171 method. The fracture toughness of composites was measured by using the compact tension specimens (35mm x 35mm x 2mm) at the same measurement condition. The fracture toughness value (Jm) was estimated by the J-integral value at the maximum load on the load-displacement diagram. J-integral value was calculated from the equation derived by Rice et al. [6]. Average values of more than five measurements were taken for each sample.

2.4. Morphological Observation

For comparison of the morphology of each composite, the cryogenically fractured surfaces were examined by using Hitachi S-2150 scanning electron microscope (SEM). Specimens were coated with a thin layer of gold to reduce the charge build-up before SEM observation. The accelerating voltage was 20kV.

3. RESULTS AND DISCUSSION

3.1. Mechanical Properties

Figure 3 shows the effect of wastepaper on the flexural properties of wastepaper/HDPE composites. The flexural modulus and strength of composites filled with wastepaper, regardless of two different processing methods, were higher than those of polyethylene matrix (HDPE/MAHPE). Because wastepaper powders were added 50wt% as the reinforcement in composites, the high specific stiffness and strength of wastepaper influenced the flexural modulus and strength of the wastepaper/HDPE composites.

The influence of processing methods on mechanical
Table 1. Mechanical properties of wastepaper/polyethylene composites (50/50) and matrix.

<table>
<thead>
<tr>
<th>Processing method</th>
<th>Flexural modulus $E_f$, GPa</th>
<th>Flexural strength $\sigma_f$, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spinning-press</td>
<td>1.62 ± 0.35</td>
<td>24.8 ± 0.88</td>
</tr>
<tr>
<td>Roll mixing</td>
<td>2.43 ± 0.15</td>
<td>31.0 ± 0.87</td>
</tr>
<tr>
<td>Polyethylene matrix</td>
<td>0.98 ± 0.05</td>
<td>23.3 ± 0.35</td>
</tr>
</tbody>
</table>

properties of the composites is shown in Table 1 (when MAHPE content is 0wt% in Fig. 3). It can be seen that flexural properties of the composites are significantly affected by the processing methods. The flexural modulus and strength of roll mixed composites were 25% to 50% higher than those of spinning-pressed composites. The result is due to the fact that the roll mixing method enhanced the dispersion of wastepaper in polyethylene matrix in comparison with the spinning-press method. Moreover, as shown in our previous report [7], in the case of spinning-pressed composites, the flexural properties were higher than those of composites made of HDPE pellets and wastepaper powders by compression molding, which possessed the flexural properties equal to or less than HDPE matrix. It is found that by means of spinning of HDPE, it is possible to blend the HDPE matrix with wastepaper powders effectively.

Furthermore, the effect of MAHPE on the flexural properties of wastepaper/polyethylene composites is shown in Fig. 3. The flexural properties of polyethylene matrices were gradually decreased by the addition of MAHPE. The result is due to the fact that the molecular weight of MAHPE is much lower than that of HDPE. In contrast, the flexural modulus of composites, regardless of different processing methods, was decreased up to 3wt% of MAHPE but was remarkably increased by further adding MAHPE above 5wt%. Similarly, the flexural strength of composites was gradually increased with increasing MAHPE contents except for the drastic increase in flexural strength of the roll mixed composite at 3wt% of MAHPE. Botev et al. [5] reported that chemical interactions existed in the composite consisting of polypropylene/maleic anhydride grafted polypropylene/basalt fibers. Therefore, the result of Fig. 3 strongly suggests that the reaction of the maleic anhydride groups on MAHPE with cellulose-components of the wastepaper improves the interfacial adhesion between the HDPE matrix and the wastepaper in composites.

The fracture toughness values ($J_{\infty}$) of both wastepaper/HDPE composites and HDPE matrices are given in Fig. 4. The fracture toughness of HDPE matrix decreased by stepwise addition of MAHPE. Under the influence of low-molecular-weight MAHPE in contradiction to HDPE, the property of HDPE matrix is considered to turn from ductile to brittle. On the other hand, within the range of MAHPE content investigated, the fracture toughness of spinning-pressed composites was higher than that of roll mixed composites. In the case of spinning-pressed composites, the fracture toughness remained stable up to 3wt% of MAHPE but was remarkably increased at 5wt% MAHPE content. This is mainly due to the improvement of interfacial adhesion between HDPE matrix and wastepaper, which interrupt the crack propagation in composites. At 10wt% MAHPE content, the reduction in the fracture toughness might be due to the residue of MAHPE, which could not react with wastepaper. For roll mixed composites, the fracture toughness was decreased with the increase of MAHPE contents. This result may be explained by the decrease in strength of wastepaper, which is undergone considerable damage as a result of the high shear force during roll mixing in a two-roll mill.

3.2. Blend Morphology

The cryogenically fractured surfaces of composites are shown in Figs. 5 and 6. In the SEM micrographs of wastepaper/HDPE composites by the spinning-press method (Fig. 5), there exist many clusters of wastepaper powders in the composite as compared with composites by roll mixing method (Fig. 6). The fractured surfaces of composites with or without MAHPE were compared. In the composites without MAHPE, as shown in Figs. 5 (a) and 6 (a), the fibers derived from wastepaper were pulled out from HDPE matrix. The vacant spaces, from which fibers were pulled out, are also observed. The fracture occurred in the interface of wastepaper and HDPE. This phenomenon indicates that the adhesion between wastepaper and HDPE matrix is poor. On the other hand, with increasing the MAHPE content of the composite, as shown in Figs. 5 (b and c) and 6 (b and c), the length of wastepaper fibers, which were pulled out from the matrix, were shorter than that without MAHPE. Typical magnified-photographs are shown in Figs. 6 (a) and 6 (b). Moreover, the dispersion of wastepaper in the HDPE matrix was enhanced by adding MAHPE as a compatibilizer. The fracture occurred in the matrix material. All SEM observations illustrate that the addition of MAHPE improved the compatibility of wastepaper and
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Fig. 5. SEM photographs of the freezed-fracture surfaces of spinning-pressed wastepaper/polyethylene composites (50/50) with MAHPE contents; (a) 0wt%, (b) 5wt%, and (c) 10wt%.

Fig. 6. SEM photographs of the freezed-fracture surfaces of roll mixed wastepaper/polyethylene composites (50/50) with MAHPE contents; (a) 0wt%, (b) 5wt%, and (c) 10wt%. A magnified photograph is at the upper right; magnification x300.

4. CONCLUSIONS

Composites consisting of a high-density polyethylene (HDPE) and wastepaper powders were prepared by two different processing methods: the spinning-press method and the roll mixing method. The influence of processing method on the mechanical properties of wastepaper/HDPE composites was studied. The compatibilizing effect of a maleic anhydride-grafted polyethylene (MAHPE) for the wastepaper/HDPE composites (50/50) was also investigated.

The concluding resumes are the following:
1. The use of wastepaper powders for the reinforcement of PE is effective to improve the flexural properties.
2. The flexural properties of roll mixed composites are higher than that of spinning-pressed composites.
3. The spinning-pressed composites exhibit improved fracture toughness than those of roll mixed composites.
4. The addition of 5wt% MAHPE into the HDPE matrix is very effective to enhance the fracture toughness of spinning-pressed composites.

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REFERENCES