

Preparation and Characterization of Cassava Fiber-Based Polypropylene and Polybutylene Succinate Composites

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ABSTRACT

Cassava root fiber was used as a bio-reinforcement in a polymer composite. A composite of polypropylene (PP) and polybutylene succinate (PBS) with cassava root fiber was prepared on a twin-screw extrusion. Maleic anhydride-polypropylene (MAPP) compatibilizer was used to improve the interfacial strength between the fibers and the polymer matrix. The effects of untreated fiber content and compatibilizer on the mechanical properties of composite were investigated. An increase in fiber loading increased the Young's modulus and flexural modulus, indicating higher stiffness in the fiber-filled composites, whereas the tensile strength and flexural strength decreased. The addition of MAPP compatibilizer in the PP composite with 50% fibers improved the flexural strength to a higher level than pure PP, therefore giving rise to a stronger but less flexible material. Studies with a scanning electron microscope revealed better interfacial adhesion between the fibers and the polymer matrix in the composite containing MAPP compatibilizer. Thermogravimetric analysis showed that the thermal stability and degradation temperature of PP composites increased with the presence of MAPP, suggesting enhanced interfacial interaction and compatibility due to the treatment of compatibilizer.

Key words: cassava, fibers, polypropylene, polybutylene succinate, MAPP, composites, extrusion

INTRODUCTION

Natural cellulosic fibers have gained considerable attention as reinforcing filler in polymeric materials. Besides their low cost and density, the plant-based natural fibers are renewable and biodegradable. They are also abundantly available as a waste from agricultural production. Thailand generates large quantities of cassava root fiber, approximately 1.5 million tons/

year, which can serve as a potential source of bio-fiber materials. Recently, the development of fiber-reinforced polymer composite has focused on using biodegradable plant-based fibers as reinforcing materials due to their exceptional specific properties (Mishra *et al.*, 2004; Reddy and Yang, 2005). New materials that provide both economic and environmental benefits have been developed for application in the packaging, building, furniture and automotive industries. The

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use of polyolefins, such as PP and PE as a matrix polymer in composites has been studied extensively due to their excellent mechanical and thermal properties (Kim *et al.*, 2006; Beckermann and Pickering, 2008). Biocomposites containing natural fiber and biodegradable polymer matrixes, (such as aliphatic polyester), for instance polylactic acid (PLA), polycaprolactone (PCL) and polybutylene succinate (PBS), are being developed widely to achieve fully renewable and environmentally friendly materials as substitutes for plastics from petroleum (Kim *et al.*, 2005; Arbelaz *et al.*, 2006; Huda *et al.*, 2008). The desirable properties of a composite depend greatly on the interfacial compatibility between the fibers and the polymer matrix. Strong interfacial adhesion is crucial to achieve efficient fiber reinforcement. Accordingly, the use of compatibilizer helps to reduce the interface tension between the hydrophilic cellulose fibers and the hydrophobic polymer matrix. Maleic anhydride grafted polypropylene (MAPP) has been demonstrated to be effective in improving the interfacial interaction by the esterification of the anhydride group of MAPP and the hydroxyl groups of the fiber (Yang *et al.*, 2007a,b).

The availability of inexpensive, natural, cellulosic cassava root fiber in Thailand provided the opportunity to investigate the possibility of its utilization in the preparation of a fiber-reinforced composite. This study therefore aimed to use cassava root fibers for the preparation of PP- and PBS- based composites and to evaluate the effect of the fibers on the mechanical properties of the materials. Moreover, the effect of MAPP on the characteristics of the composite as the compatibilizer in the PP composite was investigated.

MATERIALS AND METHODS

Materials

Cassava root fibers (CS), called cassava

pulp, were obtained from cassava starch processing factories. The fibers were washed, dried and pulverized without chemical treatment. Polypropylene (PP) and polybutylene succinate (PBS Bionolle #1000 series), used as the matrix polymer, were supplied as homopolymer pellets by GS Caltex Corp. (South Korea) and by Showa Highpolymer Co. (Tokyo, Japan). The maleic anhydride-polypropylene (MAPP) compatibilizer was obtained from Crompton Corporation (Chemtura Corporation, Middlebury, CT, USA) in the form of Polybond 3200 (1% weight maleic anhydride).

Preparation of composite

The matrix polymers, PP and PBS, were blended with cassava fibers and compatibilizing agents in a laboratory-sized, co-rotating, twin screw extruder (model BA-19, Bau Technology, Uijungbu, Kyungki, South Korea). The temperature of the mixing (central) zone in the barrel was maintained at 185°C (PP) and 145°C (PBS) with a screw speed of 200 rpm. The extruded strand was cooled in a water bath at room temperature and pelletized using a pelletizer (Bau Technology, Uijungbu, Kyungki, South Korea). Extruded pellets were oven dried at 70°C for 1 h and compounded a second time in the extruder in order to ensure good blending, before drying again at 70°C for 12 h, followed by storage in sealed polyethylene bags. Extruded pellets were injection molded into tensile (ASTM D638), Izod impact (ASTM D256), and three-point bend test bars (ASTM D790) using an injection molding machine (Bau Technology, Uijungbu, Kyungki, South Korea) at 185°C (PP composites) and 140°C (PBS composites) with an injection pressure of 1200 psi and a device pressure of 1500 psi.

Composite characterization

The tensile test for the composites was conducted according to ASTM D638-99 with a Universal Testing Machine (Zwick Co., Ulm,

Germany) at a crosshead speed of 100 mm min^{-1} . The Izod pendulum impact resistance was measured on an impact tester (model DYD-103C, DaeYeong Precision Co., Kunpo, Kyungki, South Korea) by ASTM method D256-97. The three-point bend tests of the composites were carried out according to ASTM D790 with a Universal Testing Machine at a crosshead speed of 5 mm.min^{-1} . Five replications were conducted for all measurements. The thermal degradation of the composites was characterized using a thermogravimetric analyzer TGAQ500 (TA Instruments, New Castle, DE, USA) on 10 mg samples, over a temperature range from 25 to 700°C at a heating rate of 20°C/min . Composite morphology was observed on tensile fracture surfaces under scanning electron microscopy (SEM; Sirion, FEI, Hillsboro, OR).

RESULTS AND DISCUSSION

Effect of fiber loading on mechanical properties

Figure 1 shows that the Young's modulus of composites increased with an increasing amount of cassava fiber, for both PP and PBS. The flexural modulus also followed the same trend (Figure 2). This suggested that the increased fiber loading in the polymer matrix resulted in composites becoming more rigid and stiffer. The trend of an improvement in Young's and the flexural modulus as the fiber load increased was more significant in the case of PP composites. The decrease in tensile and flexural strength of the PP and PBS composites with different levels of fibers is shown in Figures 3 and 4. For both PP and PBS, the tensile strength decreased with increasing fiber loads, whereas the flexural strength showed a relatively small change

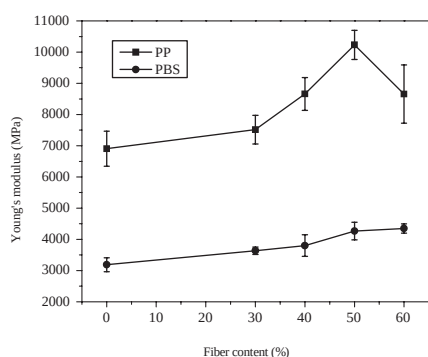


Figure 1 Young's modulus of PP and PBS composites.

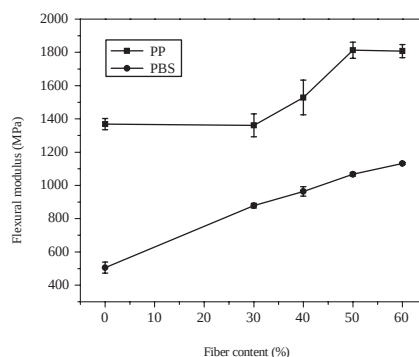


Figure 2 Flexural modulus of PP and PBS composites.

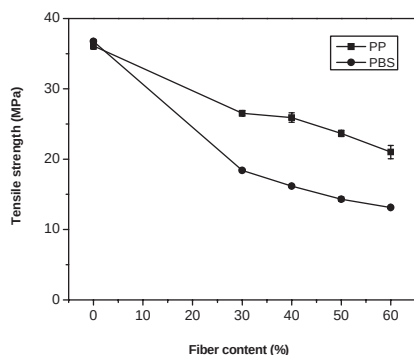


Figure 3 Tensile strength of PP and PBS composites.

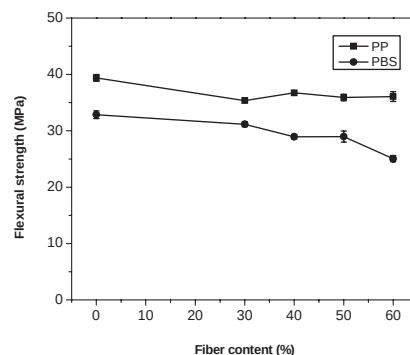


Figure 4 Flexural strength of PP and PBS composites.

with the fiber loading. Similar results were obtained with pineapple leaf fibers (Hujuri *et al.*, 2007). Figure 5 shows the strain at maximum force of the PP and PBS composites with different fiber loadings. As the polymer matrix was stiffened by fibers, elongation (the ability of material to stretch before breaking) decreased. The impact strength of the PP and PBS composites with different fiber loads is shown in Figure 6. It was observed that the impact strength decreased with increasing fiber loads.

Effect of MAPP compatibilizer

The MAPP compatibilizer enhanced the tensile strength of the PP composites containing 50% fiber, for which the tensile strength increased slightly from 23.67 to 27.7 MPa with 3% MAPP

(Figure 7). Upon increasing the compatibilizer content to 5, 7 and 9%, the material strength was not significantly different, and the tensile strength was 28.21, 28.76 and 29.03 MPa, respectively. The tensile strength of the composites with MAPP was lower than that of the pure PP. Figure 8 shows the effect of MAPP on the flexural strength improvement of the PP composites. The addition of MAPP in the PP composite with 50% fiber improved the flexural strength to a higher level than in the pure PP, therefore giving rise to a stronger, but less flexible material. The increasing level of MAPP had no statistically significant effect on the mechanical properties. The enhancement in the fiber-matrix interfacial interactions gave better stress transfer from the matrix into the fibers, leading to higher tensile

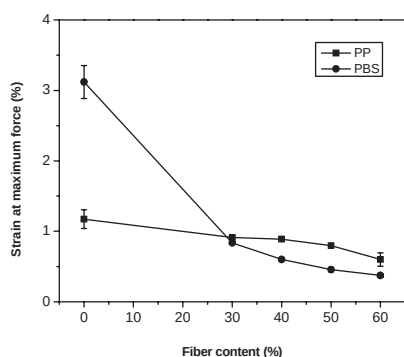


Figure 5 Strain of PP and PBS composites.

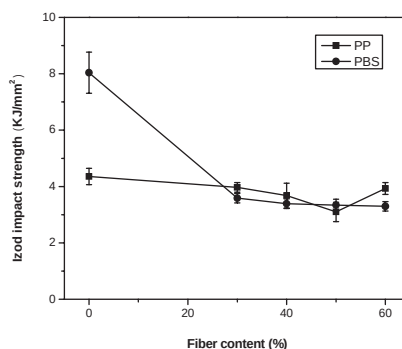


Figure 6 Impact strength of PP and PBS composites.

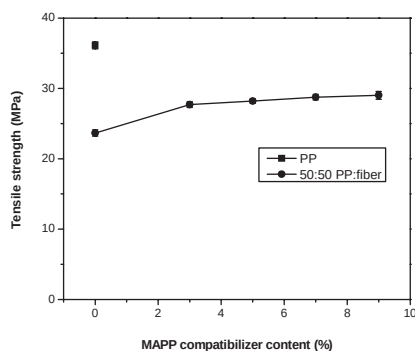


Figure 7 Tensile strength of compatibilized PP composites.

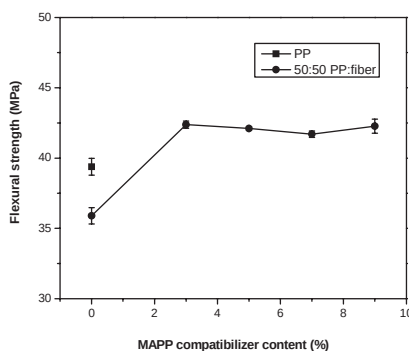


Figure 8 Flexural strength of compatibilized PP composites.

strength and flexural strength. Hujuri *et al.* (2007) reported an increase in flexural strength for PP composites with 5-20% pineapple leaf fiber compatibilized with MAPP.

Microstructure study by scanning electron microscope (SEM)

The morphology of the tensile fracture surfaces of composites was analyzed by SEM to investigate the extent of fiber and polymer matrix adhesion. Figure 9 shows SEM images of the PP-cassava fiber composites in the non-compatible samples compared to those in the compatibilized ones. It can be clearly seen in the composites without MAPP that there was evidence of fiber debonding with large gaps between the fibers and

matrix, as well as a large number of holes in the matrix where the fibers were pulled out (Figures 9(a) and (b)). Contrarily, the tensile fracture surface of composites containing MAPP showed no clear gap in the interfacial region between the polymer matrix and the fibers, having short broken fiber ends (Figures 9(c) and (d)). This indicated that MAPP promoted efficient fiber and matrix interfacial bonds, resulting in enhanced mechanical properties.

Thermogravimetric analysis (TGA)

Figure 10 presents the dynamic TGA and the derivatives of the thermogravimetric curves of the PP composites with 50% fiber, containing 0, 3 and 7% MAPP respectively. The peaks on the

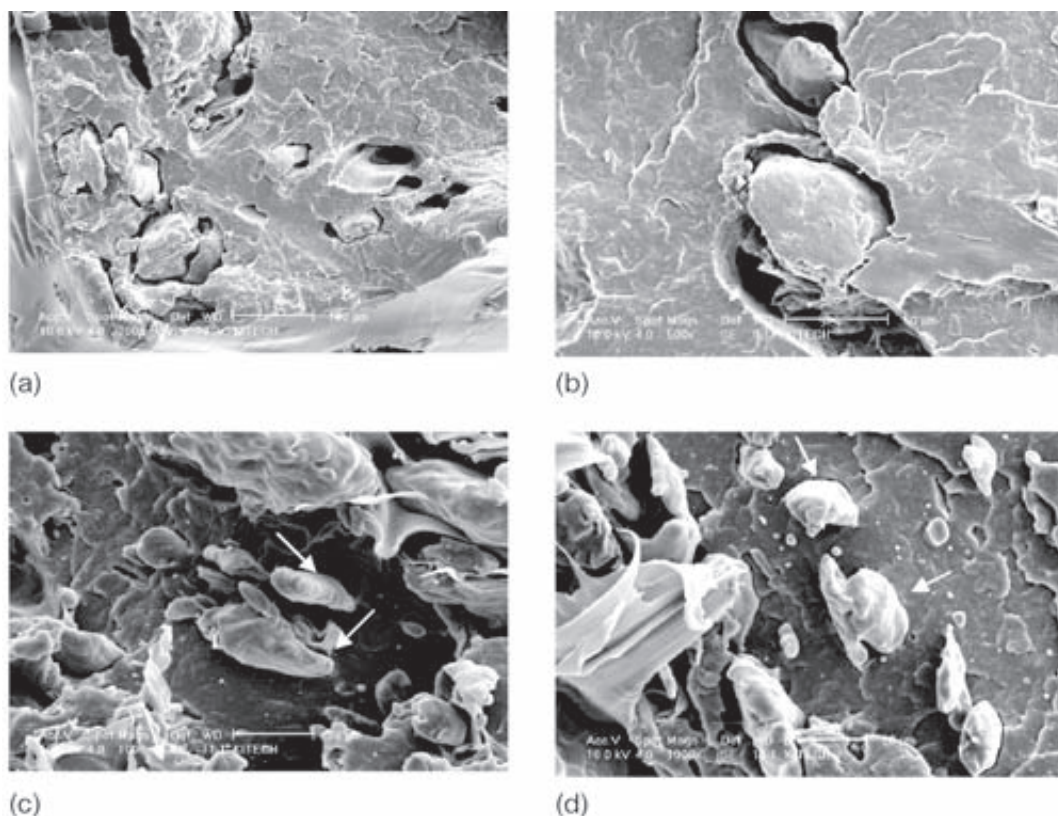


Figure 9 SEM micrographs of the tensile fracture surfaces of ((a) and (b)) PP- cassava fiber composites without MAPP compatibilizer showing the presence of cavities and large gap between fiber and matrix and ((c) and (d)) composites containing MAPP compatibilizer showing better interfacial adhesion with short broken fiber ends.

derivative curves, corresponding to the highest weight loss rates, were used to determine the degradation temperatures of the fiber and polymer fractions. The thermal decomposition characteristics of cassava fiber are also represented. The distinct peak at 250-400°C corresponded to the thermal degradation of hemicelluloses, cellulose and lignin constituents in natural cassava fiber. In the fiber filled composites, two thermal-degradation regions were observed. The first region was the result of thermal depolymerization of the constituents in the fiber. The main weight loss corresponding to the degradation of the polymer matrix occurred in the range of 460-470°C. It was evident that the thermal stability and degradation temperature of the composites increased with the addition of MAPP. Similar results were obtained with rice husk flour-filled polyolefin composites (Kim *et al.*, 2006). This behavior may be attributed to the improved fiber-matrix interfacial adhesion with the

formation of the covalent linkage at the interface, leading to better compatibility between the hydrophilic fibers and the hydrophobic PP matrix.

CONCLUSION

The incorporation of cassava root fiber into PP and PBS composites increased the stiffness or modulus of the materials. The addition of MAPP compatibilizer resulted in an improved composite strength, compensating the loss in mechanical properties caused by the fibers. The tensile fracture surface of the PP composites containing MAPP compatibilizer examined by SEM revealed a stronger interfacial bonding between the fibers and the PP matrix, which was evident from the short broken fiber ends without any clear gap around the fibers. The presence of cavities (indicating easy fiber pull out) and large gaps between the fibers and the matrix were observed in composites without compatibilizer. The improved thermal stability of the compatibilized PP composite was presumably due to the enhanced interfacial adhesion between the fibers and the matrix polymer.

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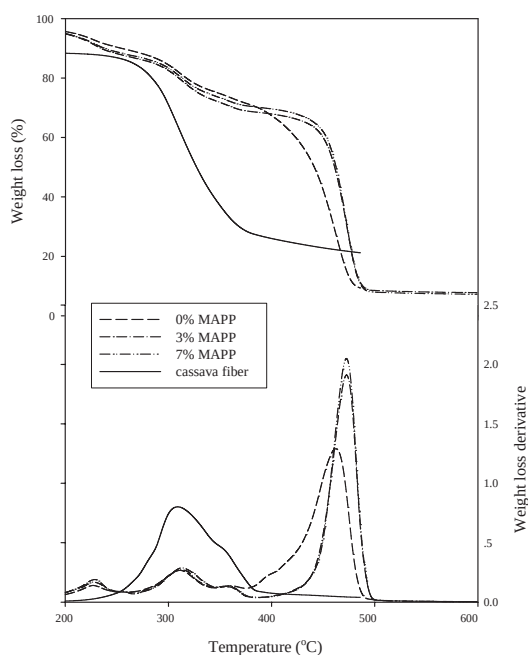


Figure 10 Thermogravimetric analysis of PP composites with 0%, 3% and 7% MAPP compatibilizer.

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