Injection Molded of Bio-Micro-Composites from Natural Fibers and Polylactic Acid

Subyakto, Euis Hermiati, Nanang Masruchin, Ismadi, Kurnia Wiji Prasetiyo, Wida Banar Kusumaningrum, and Bambang Subiyanto

Abstract

Green composites were needed by automotive industries because they are environmentally friendly, recyclable, lightweight and strong. Natural fibers such as bamboo and sisal are potential source of these materials and can be used as substitutes of fiber glass which is hard to recycle and not renewable. In this experiment, bio-composites made from micro fibers of betung bamboo (Dendrocalamus asper) and sisal (Agave sisalana) mixed with a natural polymer of polylactic acid (PLA) were developed that may used for automotive application. Bamboo or sisal fibers were converted into pulp and processed using a disc refiner to produce microfibrillated cellulose (MFC) with fiber diameter around 10 µm. MFC was mixed with PLA and triacetin and dried. The mixture was processed in a mixer at temperature of 170°C, speed of 60 rpm for 20 min. The compound mixture was removed and processed into pellets using a pelletizer at 170°C. Pellets were processed using injection molding machine. The compositions of fibers/PLA were 10/90, 20/80, and 30/70. The mechanical properties were tested in accordance with ASTM standards. Result shown that optimum composition ratio of bamboo fibers/PLA was 20/80 which gave flexural strength of 62.30 MPa, flexural modulus of 3.89 GPa, tensile strength of 44.55 MPa, tensile modulus of 1.20 GPa, and hardness of 112.90 R. While the optimum composition ratio of sisal fibers/PLA was 30/70 which gave flexural strength of 67.83 MPa, flexural modulus of 4.43 GPa, tensile strength of 48.18 MPa, tensile modulus of 1.13 GPa, and hardness of 110.50 R.

Key words: natural fibers, micro size, polylactic acid, injection, composites

Introduction

Green composites made from natural polymer reinforced with natural fibers has been subjected to many studies. This environmentally sound composites can be used for many applications, includes for automotive parts. Automotive industries grow rapidly in the world. In 2007 world production of cars is 52.1 million units, increased from 49.1 million units in 2006. If trucks are included the total production in 2007 is 74.1 million units and predicted to increase to 84 million units in 2008 (Renner 2008). Therefore materials to support these industries will be very huge and important. Now there is a trend for automotive industries to reduce utilization of materials that polluted the environment such as fiber glass, carbon and aramid fibers and substitute them with “green” materials such as natural fibers. The European Union End of Life of Vehicles (ELV) program requires that in the year of 2015 all new cars should have 95% recyclable materials (Marsh 2003). This means composites reinforced with natural fibers will play important role and might be a revolutionary material of this century (Marsh 2003). Some advantages using natural fibers compared to synthetic fibers are renewable, biodegradable, recyclable, non toxic to environment and health, lighter density, better mechanical properties, non abrasive to tools, and lower price (Leao et al. 1998; Mohanty et al. 2002; Oksman et al. 2003; Wambua et al. 2003; Mueller and Krobilowski 2003; Zimmermann et al. 2004; Suddell and Evans 2005; Bismarck et al. 2005; Bogoeva-Gaceva et al. 2007; John and Thomas 2008). Utilization of natural fibers reduce car weight up to 40%, lower energy to produce natural fiber (4 GJ/ton) compare to glass fiber (30 GJ/ton), and production of glass fiber release toxic gases such as CO2, NOx, SOx and dust (Marsh 2003; Suddell and Evans 2005). While many advantages are obtained of using natural fibers for composite, some drawbacks are realized. Natural fibers are hydrophilic in character, when it combined with polymer matrix that are hydrophobic then they have a lower compatibility. Natural fibers are also required low processing temperature to about 200°C to prevent fiber degradation (Nakagaito et al. 2005).

To overcome the drawbacks, addition of coupling agent in the matrix and improve processing methods are applied. Biocomposite from natural fibers and polymer matrix for automotive materials have been studied intensively (Leao et al. 1998; Mohanty et al. 2002; Wambua et al. 2003, Misra et al. 2004, Suddell and Evans 2005, Biedzki et al. 2006). Research on composite from raw fibers of sisal and bamboo have been done (Li et al. 2000; Mohanty et al. 2004a; Mohanty et al. 2004b; Okubo et al. 2004; Shibata et al. 2008; Okubo et al. 2009; Huang et al. 2009).

Compression molding is the main process in the production of automotive parts when using natural fiber composites, while only a little using injection molding process (Nystrom et al. 2007). However some research had been done on natural fiber composites using injection molding for general purposes (Chow et al. 1998; Arzondo et al. 2004; Arzondo et al. 2005; Godavarti 2005; Mutje et al. 2006; Panthapulakkal and Sain 2007).

Polylactic acid is polymer made from renewable materials such as corn (Oksman et al. 2003). Composites made from this polymer which reinforced with natural fibers
will produce totally green composites (Ljungberg and Wesslen 2002; Mathew et al. 2005; Iwatake et al. 2008; Suryanegara et al. 2009). The purpose of this research is to develop injection molded materials from natural fibers of sial, bamboo and green polymer of polyactic acid that may used for automotive application. Effect of fiber ratio on the mechanical properties and morphological characteristic are observed.

### Materials and Methods

#### Microfibrillated Cellulose Preparation

Betung bamboo (*Dendrocalamus asper*) and sial (*Agave sisalana*) were obtained from Bogor and Bilirat, respectively. Bamboo and sial fibers were processed into pulp using chemicals and bleached. Sial fibers were cut into 3–5 cm fibers and pulped using kraft process with 20% active alkali and 30% sulfidity. Cooked liquors were 18.06% NaOH and 7.55% N₂S with ratio of materials to liquor was 1 : 5. Cooking time was 3 hours at 160°C. Bamboo was crushed to obtain 5 cm length fiber bundles. Kraft pulping was processed with 15–17% active alkali and 22.5–25.0% sulfidity. Cooked liquors were 18.06% NaOH and 7.55% N₂S with ratio of materials to liquor was 1 : 4. Cooking time was 3.5 hours at 165°C. Bleaching process was conducted at three steps. For sial, first step using 2.14% Cl₂ at room temperature for 60 min, second steps using 1.5% NaOH at 70°C for 90 min, and the last step using 4% hypochlorite at 40°C for 180 min. For bamboo, first step using 4.68% Cl₂ at room temperature for 60 min, second steps using 1.5% NaOH at 60°C for 60 min, and the last step using 4% hypochlorite at 40°C for 180 min. Bleached pulp was processed further in a disc refiner using water for 5 cycles to produce microfibrillated cellulose (MFC) with fiber diameter around 10 μm.

#### Composites Preparation

Polyactic acid (Lacea H400, Mitsui Co. Japan) was dissolved in Dichloromethane in a reaction jar using stirrer. After PLA dissolved, wet pulp and Triacetin were put in and stirred until homogenous. The mixture was dried in an oven 60°C for 6 h. Dried mixture was processed further in a mixer (Haake Reocord 90) at temperature of 170°C, speed of 60 rpm for 20 min. The mixture was removed and processed into pellets using a pelletizer (Laboplastomill 30R150) at 170°C. Pellets were processed using injection molding machine (Nissei Plastic Industrial Co. Ltd. PS60E9ASE) to make test samples of bending, tensile, and hardness. Injection parameters were: injection temperature of 140–150°C, injection pressure of 45–50%, injection speed of 30–40%, injection time of 10s. The compositions of fibers/PLA were 10/90, 20/80, and 30/70. Triacetin added was 7% of the composite weight.

### Testing

The mechanical properties were tested in accordance with ASTM standards (ASTM D790: flexural test, ASTM D638: tensile test, ASTM D785: hardness test). Scanning electron microscope (JEOL JSM 5310LV Japan) was used to observe the fracture surface of the composites.

### Results and Discussion

#### Microfibrillated Cellulose of Sial and Bamboo

Fibers in natural fibers usually formed in bundles (50–100 μm in diameter) contains of single fiber with diameter size of 10–20 μm and can be fibrillated further into crystal cellulose with diameter size of 4–10 nm. The smaller diameter of fiber the stronger (Zimmermann et al. 2004, Nakagaito and Yano 2004). For example fiber of wood pulp has modulus elasticity of 10 GPa, however in the form of crystal structure the modulus elasticity is 130–250 GPa. Mechanical process combined with chemical process are used to make micro or nano fibers (Abe et al. 2007). Microfibrillated cellulose is first introduced by Turbak et al. (1983). Microfibrillated cellulose of sial and bamboo were prepared from bleached kraft pulp that processed using disc refiner for 5 cycles. The diameter size of fiber is less than 10 μm (Subyakto et al. 2009) therefore called micro fibers. While the fiber length of sial is about 1.4 mm (Munawar 2008) and bamboo is about 0.5–2.0 mm. This gave high aspect ratio (length/diameter) and when use for reinforcement will result in strong composites (Zimmermann et al. 2004; Zhang et al. 2010). Zhang et al. (2010) studied deeply the properties of microfibrillated cellulose from bamboo (*Phyllostachys pubescens*) pulp.

#### Mechanical Properties of Bio-micro Composites

Results of bending, tensile, and hardness properties are presented in Table 1, 2, and 3, respectively. It is shown that composites made from sial or bamboo yielded in almost similar properties, even though in some properties sial performed better compare to bamboo.

Flexural (bending) properties of composites increases with increasing fiber ratio in sial/PLA composites, while in the bamboo/PLA composites performed optimally in ratio of 20/80 (Table 1). The flexural strength of sial/PLA composites ranges from 61.78 MPa to 67.83 MPa, and the flexural modulus ranges from 3.11 GPa to 4.43 GPa. In the bamboo/PLA composites, the flexural strength ranges from 55.59 MPa to 62.30 MPa, and the flexural modulus ranges from 3.77 GPa to 3.89 GPa. These flexural properties obtained from injection molded specimens were lower than hot press molded specimen using the same materials (Subyakto et al. 2010). Using hot press molding, the flexural strength and flexural modulus of sial/PLA are 61.54–100.42 MPa and 5.77–8.07 GPa, respectively.
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Table 1. Results of flexural properties of micro-composites using injection molding.

<table>
<thead>
<tr>
<th>Composite Type</th>
<th>Max Load (N)</th>
<th>Flexural Strength (MPa)</th>
<th>Flexural Modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sisal/PLA: 10/90</td>
<td>212.6 (6.4)</td>
<td>62.49 (2.03)</td>
<td>3.11 (0.23)</td>
</tr>
<tr>
<td>Sisal/PLA: 20/80</td>
<td>208.3 (2.8)</td>
<td>61.78 (1.12)</td>
<td>3.37 (0.09)</td>
</tr>
<tr>
<td>Sisal/PLA: 30/70</td>
<td>227.2 (18.6)</td>
<td>67.83 (4.94)</td>
<td>4.43 (0.28)</td>
</tr>
<tr>
<td>Bamboo/PLA: 10/90</td>
<td>195.1 (14.6)</td>
<td>58.15 (4.14)</td>
<td>3.88 (0.08)</td>
</tr>
<tr>
<td>Bamboo/PLA: 20/80</td>
<td>208.6 (11.7)</td>
<td>62.30 (3.50)</td>
<td>3.89 (0.07)</td>
</tr>
<tr>
<td>Bamboo/PLA: 30/70</td>
<td>187.3 (8.2)</td>
<td>55.59 (2.56)</td>
<td>3.77 (0.16)</td>
</tr>
</tbody>
</table>

* Figures in the brackets are standard deviations

Table 2. Results of tensile properties of micro-composites using injection molding.

<table>
<thead>
<tr>
<th>Composite type</th>
<th>Yield Strain (%)</th>
<th>Tensile Strength (MPa)</th>
<th>Tensile Modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sisal/PLA: 10/90</td>
<td>5.68 (0.13)</td>
<td>43.04 (0.88)</td>
<td>0.95 (0.08)</td>
</tr>
<tr>
<td>Sisal/PLA: 20/80</td>
<td>5.49 (0.08)</td>
<td>42.69 (2.53)</td>
<td>1.14 (0.16)</td>
</tr>
<tr>
<td>Sisal/PLA: 30/70</td>
<td>5.52 (0.14)</td>
<td>48.18 (0.62)</td>
<td>1.13 (0.12)</td>
</tr>
<tr>
<td>Bamboo/PLA: 10/90</td>
<td>5.95 (0.23)</td>
<td>41.58 (4.09)</td>
<td>1.05 (0.06)</td>
</tr>
<tr>
<td>Bamboo/PLA: 20/80</td>
<td>5.53 (0.18)</td>
<td>44.55 (0.66)</td>
<td>1.20 (0.14)</td>
</tr>
<tr>
<td>Bamboo/PLA: 30/70</td>
<td>5.41 (0.10)</td>
<td>43.79 (1.54)</td>
<td>1.23 (0.09)</td>
</tr>
</tbody>
</table>

* Figures in the brackets are standard deviations.

Table 3. Results of hardness of micro-composites using injection molding.

<table>
<thead>
<tr>
<th>Composite Type</th>
<th>Hardness (R)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sisal/PLA: 10/90</td>
<td>118.0 (0.35)</td>
</tr>
<tr>
<td>Sisal/PLA: 20/80</td>
<td>117.5 (0.94)</td>
</tr>
<tr>
<td>Sisal/PLA: 30/70</td>
<td>110.5 (1.32)</td>
</tr>
<tr>
<td>Bamboo/PLA: 10/90</td>
<td>118.1 (1.29)</td>
</tr>
<tr>
<td>Bamboo/PLA: 20/80</td>
<td>112.9 (0.96)</td>
</tr>
<tr>
<td>Bamboo/PLA: 30/70</td>
<td>112.7 (0.45)</td>
</tr>
</tbody>
</table>

* Figures in the brackets are standard deviations.

While that of bamboo/PLA are 73.31–105.23 MPa and 6.11–7.06 GPa, respectively. The decrease may due to the processing method, where in injection molding process, the material processed in pelletizer before injection which may reduce the bond between fibers and PLA. For flexural properties, the optimal ratio of sisal MFC/PLA is 30/70, while that of bamboo MFC/PLA is 20/80.

Table 2 showed that tensile properties of composites are optimal in ratio of sisal MFC/PLA of 30/70 and bamboo MFC/PLA of 20/80. The yield strain decreased with increasing fiber ratio or decreasing of PLA as matrix, because PLA has higher yield strain compare with sisal or bamboo fibers. The tensile strength obtained in this experiment are range of 42.69–48.18 MPa for sisal MFC/PLA and 41.58–44.55 MPa for bamboo MFC/PLA. Lee et al. (2004) obtained lower value about 35 MPa of tensile strength of composite PLA-bamboo fiber using hot press molding. This may due to the size of bamboo fiber they used (fiber bundle with diameter of 70 µm). In this experiment we used smaller diameter of fiber around 10 µm. The tensile modulus of this experiment range from 0.95–1.14 GPa for sisal MFC/PLA composites and 1.05–1.23 GPa for bamboo MFC/PLA composites. This values of tensile modulus are lower compare to that obtained by Lee et al. (2004) about 2.7 GPa.

Table 3 presented the hardness results of composites of sisal MFC/PLA and bamboo MFC/PLA at various fiber ratios. For sisal MFC/PLA composites the highest value of hardness obtained from ratio of 10/90, similarly with bamboo MFC/PLA composites. This my due to the character of fibers which has high aspect ratio of length to diameter. In the flexural and tensile properties, this aspect ratio has significant effect as reinforced agent of the composites. However in hardness this effect may not significant, where PLA as matrix is more dominant.

Scanning Electron Microscope Observation

SEM observation of fracture surface of composites are presented in Figure 1 for sisal MFC/PLA and Figure 2 for bamboo MFC/PLA. It is shown that in all fiber ratios the mixture between fibers and PLA are homogenous.
Conclusions

Bio-micro composites from micro fibers of sisal, bamboo and polylactic acid was developed using injection molding machine. Effect of fiber ratio on the mechanical properties and morphological characteristics were observed. The results showed that the optimum composition of sisal MFC/PLA is 30/70, while that of bamboo MFC/PLA is 20/80. SEM observation showed that in all compositions the mixture between fibers and PLA are homogenous.

References


Subyakto; E. Hermiati; D.H.Y. Yanto; Fitria; I. Budiman; Ismadi; N. Masruchin; B. Subiyanto. 2009. Process Development to Produce Cellulose Nanofibers from Sisal (Agave sisalana) and Betung Bamboo (Dendrocalamus asper), Berita Selulosa 44 (2): 57-65.


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