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Effect of Molding Condition on the Mechanical Properties of Bamboo-Rayon Continuous Fiber/Poly(Lactic Acid) Composites

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Abstract

Natural fiber reinforced composite materials attract much attention in relation to the present global environmental problem. Continuous natural fiber reinforced thermoplastic composites have superior mechanical properties and have great potential as structural materials. However, it is difficult to impregnate reinforcement fibers with thermoplastics because of the high viscosity of molten plastics. In this study, continuous natural fiber reinforced thermoplastic composites were molded using intermediate materials fabricated through the micro-braiding technique. Bamboo-rayon fiber and poly(lactic acid) (PLA) fibers were used as reinforcement and matrix, respectively. Composite plates were fabricated by hot press molding under various molding conditions, including molding temperature, pressure and time. Tensile and shear tests were conducted to evaluate the effect of molding condition on mechanical properties of the composites. Tensile and shear strength increased with increasing molding temperature, whereas they are little affected by molding pressure and time. It was clarified that a molding temperature of 190°C, molding pressure of 1 MPa and molding time of 4 min are optimum molding conditions that lead to securing the best tensile strength ratio.

Keywords

Bamboo, poly(lactic acid), micro-braiding technique, continuous fiber reinforced thermoplastics

1. Introduction

Commodity plastics used in various fields have adequate strength and lower weight, and are inherently from petroleum. About 21% out of 1000 tons of plastic waste per

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year is incinerated and about 33% is landfilled. Considering this situation, the usage of commodity plastics presents a problem, such as exhaustion of resources, due to mass consumption. In addition, toxic gas generation during incineration and landfill shortage due to their non-degradability are also serious problems. Presently, natural fiber reinforced plastics attract much attention from these points of view. Natural fiber reinforced plastics are made with renewable resources and are expected to be much more widely used as materials for a recycling-oriented society. Especially, continuous natural fiber reinforced plastics are expected as alternatives to glass fiber reinforced plastics and many investigations have been conducted. Herrera-Franco and Valadez-Gonzales investigated the effect of alkali treatment on the mechanical properties of henequen fiber reinforced composites [1]. Hepworth *et al.* focused on the effect of the decortication process on the mechanical properties of hemp fiber reinforced composites [2]. Khondker *et al.* investigated the effect of fiber content on the mechanical properties of jute fiber reinforced composites [3].

On the other hand, the usage of biodegradable plastics as matrices of natural fiber composites has a possibility of solving the landfill shortage problem, because both reinforcement and matrix are degraded to molecular levels by the action of microorganisms in the field. In biodegradable plastics, poly(lactic acid) (PLA) is better suited for a matrix of natural fiber composites because it is naturally derived and has biodegradability. Examples of studies about natural fiber reinforced PLA include jute fiber composites by Bledzki and Jaszkiewicz [4], flax fiber composites by Bax and Müssig [5] and hemp fiber composites by Graupner *et al.* [6]. These studies, however, focused on the short fiber reinforced composites and there are few works about continuous fiber reinforced PLA [3]. This is because PLA is thermoplastic, that is, molten PLA has a high viscosity and there is therefore less impregnation to continuous fiber yarns.

In order to improve impregnation of thermoplastics to reinforcement fiber yarns, a micro-braiding technique has been developed based on the traditional braiding technique. In the micro-braiding technique, resin fiber yarns are braided around reinforcement fiber yarn and an intermediate material (micro-braided yarn) is obtained as a yarn shape. Since resin fiber yarns are located adjacent to a reinforcing fiber, better impregnation during melt molding is expected [3, 7, 8]. The micro-braided yarn as an intermediate material can be treated as a fiber yarn so that a successive weaving process is available.

In the present study, continuous fiber reinforced plastics were developed based on the micro-braiding technique. In order to fabricate micro-braided yarn, bamboo rayon fibers and thermoplastic PLA fiber yarns were used as reinforcement and matrix, respectively. Specimens were molded under various conditions and tested to clarify the effects of molding condition on the mechanical properties of the continuous bamboo fiber reinforced PLA composites.

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2. Experimental

2.1. Molding

The materials used in this study were a bamboo rayon fiber (Tex: 18.5) and PLA fiber yarns (Tex: 8.2). Micro-braided yarn was fabricated as fiber volume fraction of 50% with a tubular-braiding machine. In the present study, since bamboo rayon fiber was very fine and was tender, four fibers were bundled with a quiller machine and were used in the micro-braiding process. Fabrication and structure of micro-braided yarn are illustrated in Fig. 1.

Micro-braided yarn obtained was wound on the metallic frame 50 × 2 times. Then, the micro-braided yarns wound on the frame were placed on the pre-heated mold die and were molded with a hot-press system. Figure 2 shows a schematic view of the molding process. After a given molding cycle, the heating platens were

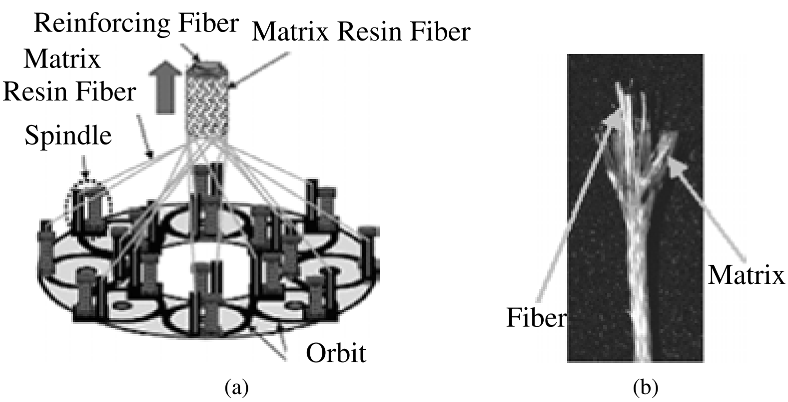

Figure 1. Fabrication of micro-braided yarn. (a) Schematic view of a braiding machine. (b) Micro-braided yarn.

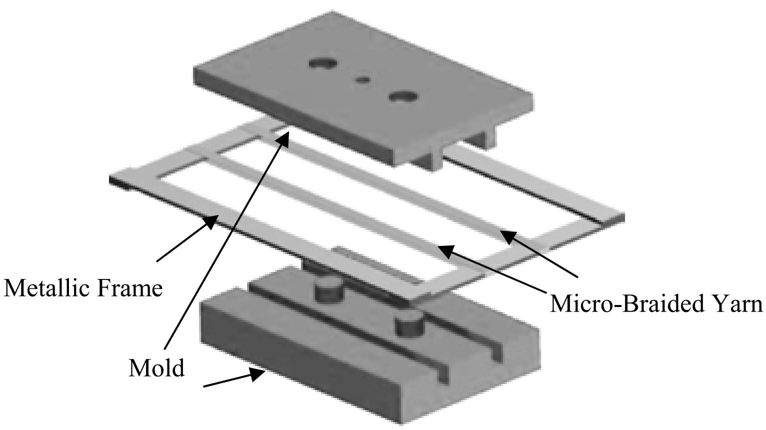

Figure 2. Alignment of micro-braiding for compression molding. This figure is published in color in the online version.

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cooled by the water flow in the pipes equipped through the platens. During cooling, molding pressure was applied until the temperature of the platens became 40°C and the molded pieces were obtained. The molding piece was a unidirectional composite with 200 mm length and 20 mm width. Here, molding pressure, molding temperature and molding time are defined as the pressure during compression molding, the temperature of the heating platens and the duration of molding temperature, respectively. In the present study, molding temperature, molding time and molding pressure were selected as 170 · 190°C, 4 · 8 min and 1 · 3 MPa, respectively, which resulted in the 8 molding conditions.

2.2. *Observation of Impregnation*

In order to observe the impregnation, the molding pieces were embedded in an epoxy resin system. After the cross-sections were polished, they were observed with an optical microscope. The resin system consisted of Epikote 828 (JER Co.) and triethylenetetramine and was cured for 24 h under atmosphere pressure.

2.3. *Tensile Tests (Composites)*

Tensile tests were conducted on the molding pieces to clarify the effect of molding condition on the mechanical properties. For tensile tests, a universal testing machine (AGS-1000A, Shimadzu Co.) was used. Aluminum tabs were glued on the end of the specimen, which resulted in the 140 mm parallel section. Cross-head speed was 1 mm/min. Load and tensile strain were recorded using a load cell (10 kN capacity) and a strain gage (5 mm gage length), respectively.

2.4. *Tensile Tests (Bamboo Fibers)*

In order to evaluate the strength of bamboo fibers, tensile tests were also conducted on the bamboo rayon fibers using a universal testing machine (AGS-1000A, Shimadzu Co.). The capacity of the load cell was 500 N. The length of the specimen was 160 mm. Paper tabs were glued on the both end of the specimen, which result in the 100 mm reduced length. Cross-head speed was 1 mm/min.

There is a possibility of the effect of heating and pressure during molding on the properties of bamboo fibers in the composites. In order to evaluate the effect of molding condition on the fiber strength in the composites, tensile tests were conducted on the fibers extracted from the central portion of the composites using chloroform. The length of the specimen was 85 mm. Paper tabs were glued on both ends of the specimen, which resulted in the 25 mm reduced length. Cross-head speed was 1 mm/min.

2.5. *Shear Tests*

Shear tests were also conducted on the specimens based on the Iosipescu method [9]. An Iosipescu fixture equipped on the universal testing machine (AGS-1000A, Shimadzu Co.) was used. The capacity of the load cell was 10 kN. The specimen geometry was 60 mm length and 18 mm height with isosceles triangle shape notches

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with 4.5 mm height and 4.5 mm base at both sides of the center of a specimen. Cross-head speed was 1 mm/min.

3. Experimental Results and Discussion

3.1. Moldability

Cross-sectional observation results for the composites molded in the present study are shown in Fig. 3. The melting point of a PLA fiber used was about 170°C as

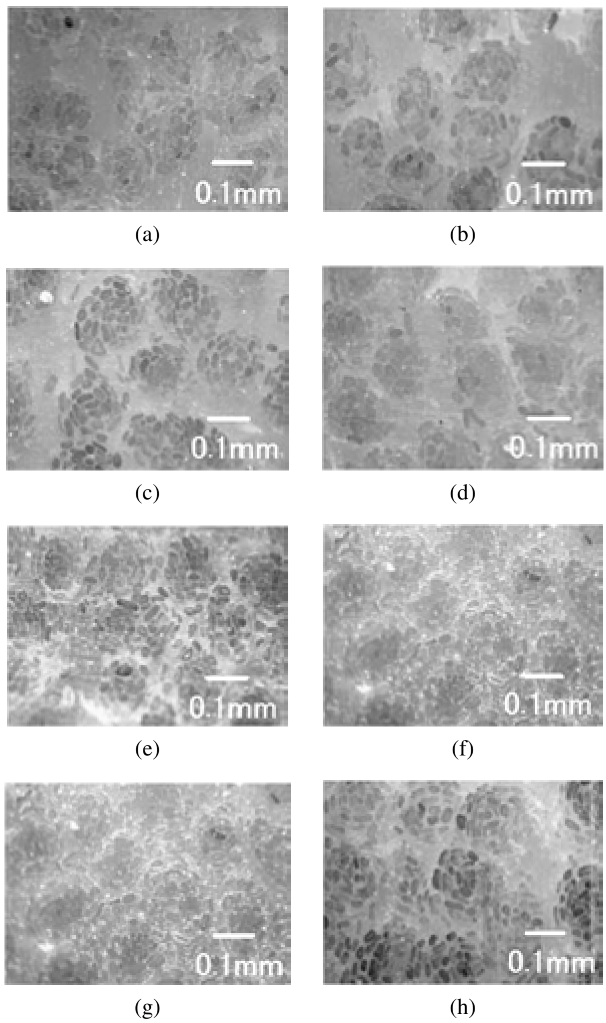

Figure 3. Results of cross-sectional observation of bamboo-rayon/PLA composites molded at different temperatures. (a) 170°-1 MPa-4 min; (b) 170°-1 MPa-8 min; (c) 170°-3 MPa-4 min; (d) 170°-3 MPa-8 min; (e) 190°-1 MPa-4 min; (f) 190°-1 MPa-8 min; (g) 190°-3 MPa-4 min; (h) 190°-3 MPa-8 min. This figure is published in color in the online version.

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the endothermic peak point obtained by differential scanning calorimetry. From this fact, the viscosity of molten PLA at 190°C was considered as lower than at 170°C so that the impregnation was considered as better at 190°C. However, good impregnation was achieved in all conditions, whereas the area of resin rich region varied at each molding condition. These results indicate that volume fraction varies with molding condition. Thus, the fiber volume fraction was calculated using the following equation,

$$V_\mathrm{f} = \frac{\rho_\mathrm{c} - \rho_\mathrm{m}}{\rho_\mathrm{f} - \rho_\mathrm{m}}, \tag{1}$$

where V_f is fiber volume fraction, ρ is density and subscript c, f and m indicate composites, fiber and matrix, respectively. Each density was measured based on the Archimedes' principle. The equation used in the measurements is,

$$\rho = \frac{m_\mathrm{a}}{m_\mathrm{a} - m_\mathrm{w}} \rho_\mathrm{water}, \tag{2}$$

where m is weight of a composite and subscripts a, w and water are in air, in water and for water, respectively.

Since there is a possibility of change in density during molding, density measurements were conducted on the fiber extracted from composites using chloroform. The relation between density of bamboo rayon fiber and molding condition and the relation between density of the composites and molding condition are shown in Figs 4 and 5, respectively. For comparison, density of bamboo rayon fiber without chloroform treatment was also measured and shown in Fig. 4. The density of PLA fiber was 1.26 g/cm^3.

From Fig. 4, there is little effect of the chloroform treatments on the density of bamboo rayon fiber. No effect of molding condition on the density was also confirmed. In contrast to the results of Fig. 4, an increase in the density of the

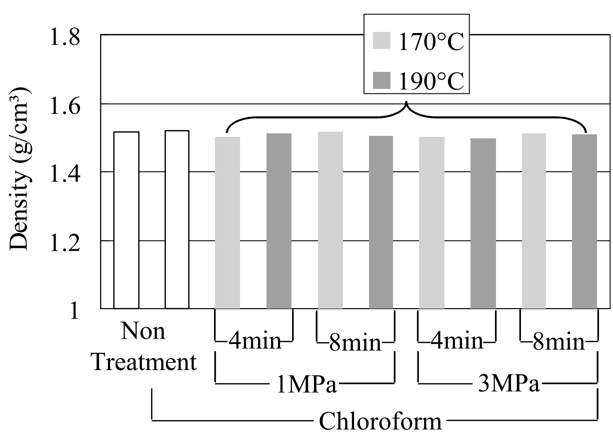

Figure 4. Density of bamboo-rayon fiber extracted from bamboo-rayon/PLA composites molded at different conditions.

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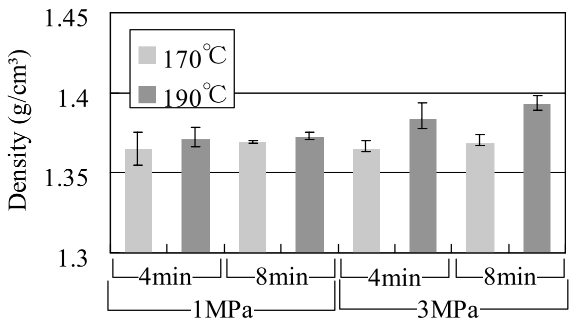

Figure 5. Density of bamboo-rayon/PLA composites molded at different conditions.

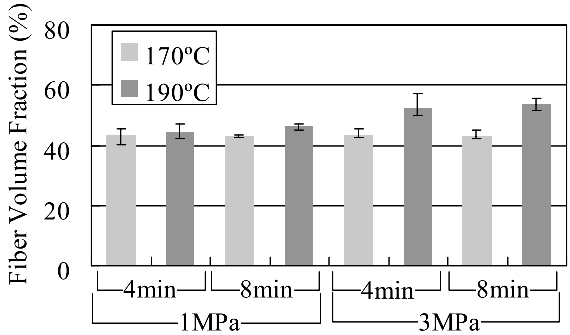

Figure 6. Fiber volume fraction of bamboo-rayon/PLA composites molded at different conditions.

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composites under 190°C-3 MPa-4, 8 min was observed from Fig. 5. Considering the volume fraction of the reinforcements in a micro-braided yarn used in the molding, it was due to matrix pouring out from the mold die. This tendency depended more on molding temperature and pressure than molding time.

Figure 6 shows the relation between volume fraction calculated using the above results and molding condition. From Fig. 6, an increase in the volume fraction under 190°C-3 MPa-4, 8 min was observed, which was the same tendency as with Fig. 5. From these results, an increase in the volume fraction was attributed to the amount of matrix pouring out from the mold die.

3.2. Mechanical Properties

Stress–strain curves of molding temperature 170 and 190°C are shown in Figs 7 and 8, respectively. The curves are offset in the strain axis. From Figs 7 and 8, mechanical properties were affected by the molding condition.

For quantitative discussion, the relation between Young's modulus and molding condition obtained from Figs 7 and 8 is shown in Fig. 9. From Fig. 9, Young's modulus increased with increasing molding temperature. This was attributed to the increase in fiber volume fraction.

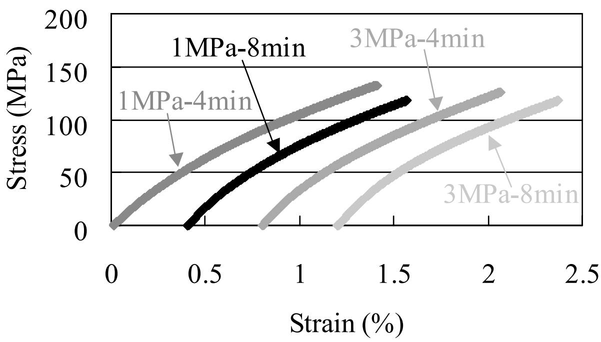

Figure 7. Stress–strain curve of bamboo-rayon/PLA composites (molding temperature: 170°C).

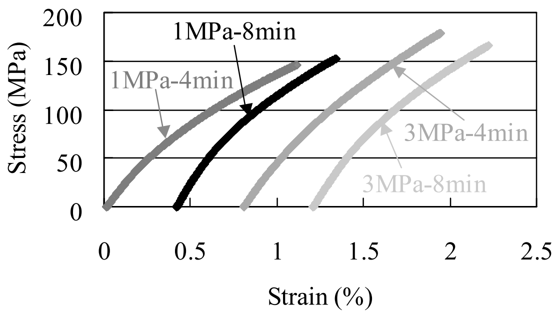

Figure 8. Stress–strain curve of bamboo-rayon/PLA composites (molding temperature: 190°C).

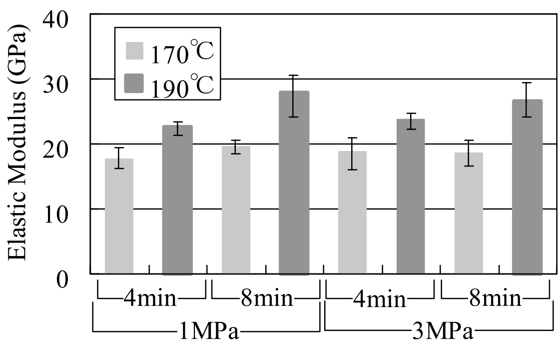

Figure 9. Elastic modulus of bamboo-rayon/PLA composites molded at different conditions.

Figure 10 shows the relation between tensile strength and molding condition. Tensile strength increased with increasing molding temperature, which was also attributed to increasing volume fraction. In addition, increase in interfacial shear strength between fiber and matrix, which was not measured in the present study, might also improve the tensile strength. In addition, molding pressure and time had little effect on the tensile strength. It is noted that increase in strength and modulus with increasing temperature under 1 MPa molding pressure was confirmed, regard-

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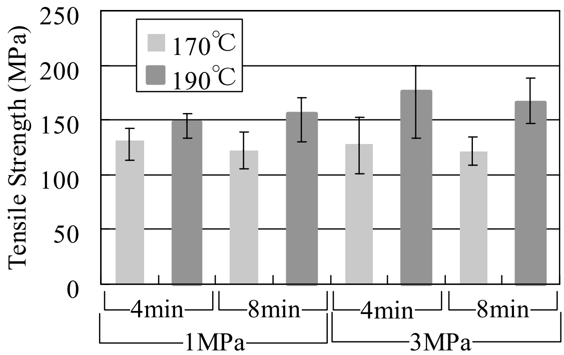

Figure 10. Tensile strength of bamboo-rayon/PLA composites molded at different conditions.

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less of almost constant fiber volume faction. The reason for this phenomenon is unclear for now and will be discussed in the future works.

In the present study, achievement ratio was calculated to evaluate the degree of fiber strength in the composites. Achievement ratio is calculated as

$$P = \frac{\sigma_f}{\sigma_{fc}} \times 100, \tag{3}$$

where P is achievement ratio, σ_f is stress which reinforcing fibers carried at the composite failure and is calculated using a rule of mixture, and σ_{fc} is the strength of the fiber extracted from the composites using chloroform. The equation of rule of mixture is

$$\sigma_c = \sigma_f V_f + (1 - V_f) E_m \varepsilon_c, \tag{4}$$

where σ_c is strength of the composite, V_f is volume fraction, σ_c is maximum strain of the composites and E_m is modulus of the matrix (PLA).

The strength of the fiber extracted from the composites is shown in Fig. 11. For comparison, strength of bamboo rayon fiber without chloroform treatment was also measured and shown in Fig. 11. Achievement ratio calculated from the results of Figs 10 and 11 is shown in Fig. 12. From Fig. 11, chloroform treatment did not affect the strength of the bamboo rayon fiber. Molding conditions selected in this study also did not affect the strength of the fiber. From Fig. 12, achievement ratio increased with increasing molding temperature. Molding pressure and time had little effect on the achievement ratio. One reason for the improvement in achievement ratio with temperature was improvement in interfacial condition between fiber and matrix. Thus, shear strength, which is more affected by interfacial strength, were measured. Figure 13 shows the relation between shear strength and molding condition. Shear strength increased with increasing molding temperature. This was considered as increasing fiber/matrix shear strength. This tendency was similar to Figs 10 and 12. From these results, increases in tensile strength and achievement ratio were also attributed to the increasing fiber/matrix shear strength. A mechanism of increasing interfacial strength with molding temperature was unclear. However,

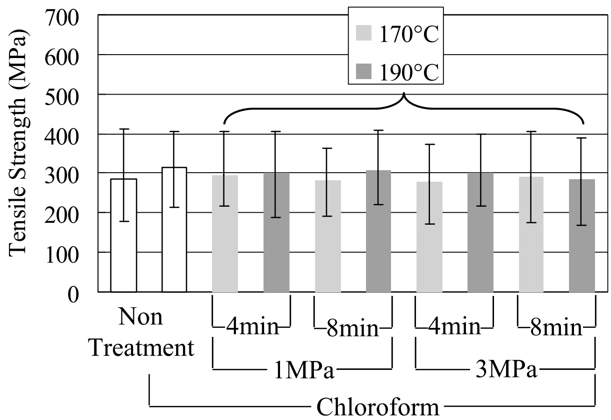

Figure 11. Tensile strength of bamboo-rayon fibers extracted from bamboo-rayon/PLA composites molded at different conditions.

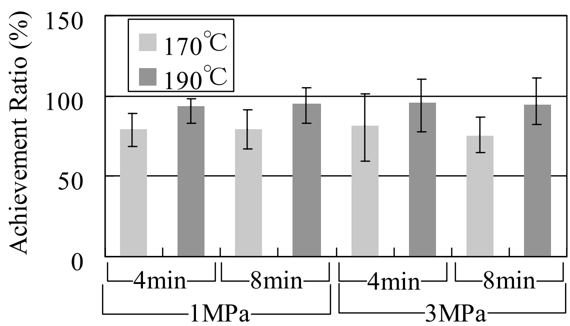

Figure 12. Achievement ratio of bamboo-rayon/PLA composites molded at different conditions.

there was the possibility of chemical interaction between constituents of the fiber and matrix.

3.3. Optimum Molding Condition

From the result of Sections 3.1 and 3.2, the optimum molding condition of bamboo fiber reinforced composites is discussed. Tensile and shear strength were improved with increasing molding temperature. Therefore, molding at 200°C was conducted. However, tensile strength was reduced to about half. In order to discuss the reason, bamboo rayon fiber was extracted from the composites using chloroform and was observed by scanning electron microscopy. The observation result is shown in Fig. 14. Cracks formed in the fiber were observed. This was due to the pyrolysis of the constituent of the fiber. The crack formation reduced the composite strength. This result corresponded with the report by Madsen *et al*. [10, 11]. Thus, the optimum molding temperature for bamboo rayon fiber PLA composites in this study is 190°C.

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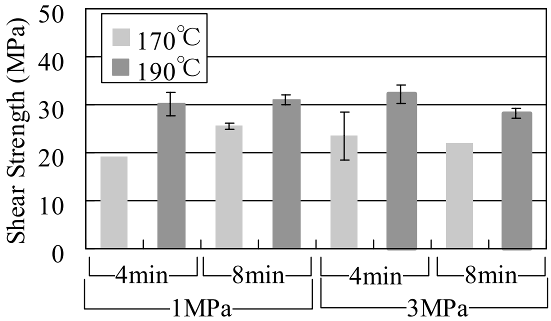

Figure 13. Shear strength of bamboo-rayon/PLA composites molded at different conditions.

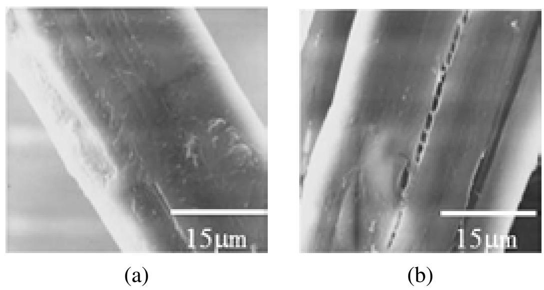

Figure 14. Fiber surface of bamboo-rayon fiber molded at different conditions. (a) 190°-3 MPa-8 min; (b) 200°-1 MPa-4 min.

There was little effect of molding pressure and temperature on the achievement ratio. For this reason, the optimum molding pressure and time are 1 MPa and 4 min, respectively, whereas fiber volume fraction, which affects mechanical properties, increases with increasing molding pressure. This result suggests that mechanical properties can be controlled by adjusting the fraction of the reinforcement fiber in micro-braided yarn to some degree.

4. Conclusion

Unidirectional bamboo rayon fiber reinforced PLA composites were molded and the relation between mechanical properties and molding condition was discussed. Conclusions obtained are as follows.

1. Good impregnation was achieved using micro-braided yarn as an intermediate material.
2. The density and the fiber volume fraction increased under the molding condition of 190°C-3 MPa-4, 8 min.
3. Tensile strength, modulus, achievement ratio and shear strength increased at the molding temperature of 190°C.

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4. Density and strength of the bamboo rayon fiber were not affected by chloroform treatment and molding condition.
5. The optimum molding condition for bamboo rayon fiber PLA composites used in the present study is molding temperature of 190°C and there was little effect of molding pressure and time.

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