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PROCESSING AND MECHANICAL PROPERTIES OF BIODEGRADABLE COMPOSITES

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SUMMARY: Continuous natural fiber reinforced thermoplastic materials are expected to replace inorganic fiber reinforced thermosetting materials. However, natural fiber itself has low strength and in the process of fabricating the composite, it is difficult to impregnate the thermoplastic resin into reinforcement fiber because of the high melt viscosity. In our previous studies, it has been clarified that a spun natural yarn had high strength. Moreover intermediate material, which allows highly impregnation during molding, can be obtained for fabricating continuous fiber reinforced thermoplastic composite by aligning resin fiber alongside reinforcing fiber with braiding technique. This intermediate material has been called "Micro-braided-yarn". In this investigation, completely naturally-derived micro-braided-yarn was fabricated by using thermoplastic biodegradable resin fiber (PLA) as the resin fiber and jute spun yarn as the reinforcement. Using jute spun yarn/PLA micro-braided-yarn, continuous natural fiber reinforced biodegradable resin composite plates were molded by hot press molding with various molding conditions. Observation of impregnation state and evaluation of mechanical properties were performed, and consequently, the relationship between the molding conditions and the mechanical properties of jute spun yarn/PLA composite was clarified. In addition, it is well known that the interfacial properties between natural fiber and resin is low, therefore, surface treatment on jute fibers was performed by using shellac resin (a natural surfactant) to improve the interfacial properties of jute fiber. Furthermore, jute spun yarn/PLA Micro-braided yarn textile was fabricated by using weaving techniques.

KEYWORDS: natural fibers, biodegradable resin, surface treatment, woven fabric

INTRODUCTION

Natural fibers are currently being developed as a possible substitute for conventional reinforcement materials such as glass fiber and carbon fiber because they have advantages of low cost, low density, ease of separation and biodegradability and recycle of thermoplastic is easier than that of thermosetting plastic. In this circumstance, investigation of natural fiber reinforced thermoplastic composite considering the environment have been carried out. [1-3] The natural fiber has also problems as follows:

(1) Mechanical properties are low.
(2) Fluctuation of fiber fineness and strength is wide.
(3) Impregnation of thermoplastic resin into reinforcement fiber bundle is difficult.

In series of study, the spun yarn was used as reinforcement fiber to solve (1) and (2). Natural fiber is short fiber and has no orientation. It also has wide fluctuation in fineness, length and strength. Usage of short fibers spun yarn is, fortunately, possible to reduce the fluctuation of the properties by spinning short fiber. Also spun yarn is available for continues fiber which can be used for textile process. Moreover, Micro-braided yarn was employed as an intermediate material for high impregnated continuous fiber reinforced plastics, in which traditional Japanese braiding technique was applied, to solve (3). Micro-braided yarn as shown in Fig. 1 was fabricated by braiding resin fibers alongside reinforcement fiber. Since resin fibers are located close to reinforcement fiber bundle, impregnation performance is excellent [4]. The interfacial adhesion between reinforcement and matrix is very important for mechanical properties of composite materials. However, the interfacial properties between natural fiber and most of biodegradable polymers such as aliphatic polyester polymers are inadequate [5]. In considering the interfacial properties, it is important to take into consideration natural fibers comprise several components, e.g. cellulose, hemi cellulose, and lignin, which are different from glass, carbon and other synthetic fibers that have uniform composition. In addition, the crystallinity of cellulose which is the primary framework of plants also plays significant role on the interfacial adhesion. Thus, in natural fiber reinforced plastics, one of the approaches to solve these problems to supply is surface modification onto the fibers with silane coupling agent or other synthetic chemical compounds [5, 6]. However, because of the mostly artificial substances which made of petroleum origin, any bio fiber reinforced bioplastics using these agents can not be considered as all-biobased or all-biodegradable. Shellac resin is secretion of lac insect which contains polar functional group. Because jute fiber also contains polar functional group, it is propriety to use treated jute fiber with shellac resin to improve interfacial adhesion.

In this paper, jute spun yarn / PLA micro-braided yarn was fabricated. The micro-braided yarns were compression molded to produce natural fiber reinforced thermoplastic plates under various molding conditions. Relationship between molding condition and mechanical properties of specimen was investigated and the optimum molding condition was decided. Moreover, the jute fibers were treated with various concentrations of the aqueous solution of shellac resin. Coated jute spun yarn/PLA micro-braided yarns were molded by the optimum molding condition. Relationship between the surface treatment condition of shellac resin and mechanical properties of the specimen was investigated.

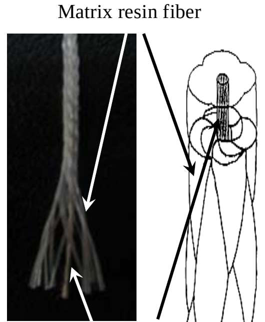

Fig.1 Micro-braided yarn

MATERIALS AND TESTING METHOD

Jute spun yarn was used as reinforcement fiber. As a resin fiber, Polylactic acid (PLA) fiber (Lactron, 16.7tex, Kanebo synthetic fiber), naturally-derived biodegradable thermoplastic resin fiber, was used. The jute spun yarn was treated with an aqueous solution of shellac resin (Gifu Shellac Ltd., Japan). The concentration of shellac resin was varied (0.5wt %, 2.0wt %, 5.0wt %). After coating with shellac resin, the coated jute yarn was dried at 80^{o}C, various drying times (0hr,0.5hr,1hr,2hr,3hr). By using Jute spun yarn coated with shellac resin as axial yarn and PLA resin fiber as braiding yarn, Jute/PLA Micro-braided yarn was fabricated by tubular braiding machine.

Micro-braided yarns were wound parallel at 30 (for tensile test) or 60 (for bending test) times onto a metallic frame. The metallic frame has a spring mechanism so as to adjust the tension caused by the thermal shrinkage of fiber during processing. Then the metallic frame was placed in a heated die with 20×200 mm concave and molded to produce unidirectional test specimens by changing molding conditions.

Tensile test of moldings was performed by using INSTRON universal testing machine (model 4206). The specimen size was 200 mm in length, 20 mm in width and 0.8 ~ 1 mm in thickness, and aluminum tabs (thickness 0.5 mm, width 20 mm, length 50 mm) were put on both ends. The span length was 100 mm and the crosshead speed was 1mm/min. At the same time, strain was measured by strain gage (gage length 10 mm). A three point bending test of moldings was performed by using INSTRON universal testing machine (model 4206). The specimen size was 20 mm in length, 10 mm in width and 2.0 mm in thickness. The span length was 16 mm and the test speed was 1 mm/min.

OPTIMUM MOLDING CONDITION

First, the effect of molding condition on mechanical properties was investigated using non-coated Jute spun yarn/PLA Micro-braided yarn. The specimen with Jute spun yarn which was not treated with shellac resin was called “None-coated”. The thickness of the moldings was changed by changing the molding temperature and as a consequence the fiber volume content (Vf) was varied. Since the increase in molding temperature led to decreasing specimen thickness, the fiber volume fraction in the composite plates could be different. Thus, the achievement ratio was used to compare the tensile properties among the specimens with different moding conditions. Achievement ratio was calculated by normalizing the experimental value with theoretical value. Theoretical value was calculated by rule of mixture.

Relationship between the achievement ratio of tensile strength and molding conditions (molding temperature and time) is shown in Fig. 2. When the molding temperature was changed in molding pressure of 1.33 MPa and molding time of 8 min., the achievement ratio of tensile strength was increased until 190^{o}C and decreased over 190^{o}C.

Here, the effect of molding temperature on jute fiber was clarified. TGA measurement of Jute fiber and tensile test of heated jute single yarn were performed. From results of TGA measurement, reduction rate of weight of jute fiber became more than 5 % at nearly 200^{o}C. In addition, tensile strength of jute single yarn decreased at above 200°C of molding temperature. Next, cross-sectional observation was performed to investigate the internal structure of the moldings. The higher molding temperature became, the better impregnation advanced, and unimpregnation region could not be seen at 190°C of molding temperature. Consequently, strength of molding was increased with increasing molding temperature until molding temperature reached 190°C because unimpregnation ratio was decreased, and the strength of the jute fiber did not decrease until 190°C. In the case that molding temperature was higher than 200°C, although the impregnation of PLA resin into the jute fiber bundle was enough, strength of molding was decreased due to heat deterioration of jute fiber with increasing molding temperature.

When the molding time was changed in molding pressure of 1.33 MPa and molding temperature of 190°C, which was optimum molding condition from the above-mentioned result, the achievement ratio of tensile strength was increased until 8 min. and decreased over 8 min. Cross-sectional observation of moldings at various molding time was performed. Unimpregnation region could not be seen at any molding time. Moreover, dispersion of jute fibers was observed with increase in molding time. The bundle area was increased until 8 min. and decreased over 8 min. Because of increasing molding time, PLA resin impregnated into jute fiber bundle and then the jute fiber was dispersed. In the case of above 8min., since the increase in molding time led to decreasing in specimen thickness, so the jute fiber could not be dispersed enough above 8 min. With increasing molding time, tensile strength of jute single yarn decreased as the jute fiber was deteriorated by heat. Consequently, in molding pressure of 1.33 MPa and molding temperature of 190°C, if molding time was increased until 8min., strength of molding was increased because dispersion of jute fiber was advanced. In the case that molding time was longer than 8 min., strength of molding was decreased due to the heat deterioration of jute fiber.

As described above, because the strength was affected by impregnation of resin into fiber bundle, heat deterioration of jute fibers and dispersion of jute fibers, optimum molding condition was limited. For example, in molding pressure of 1.33 MPa, the molding condition that the achievement ratio of tensile strength became higher than 85 % was that molding temperature was 190 ~ 195°C, molding time was about 5 ~ 8 min.

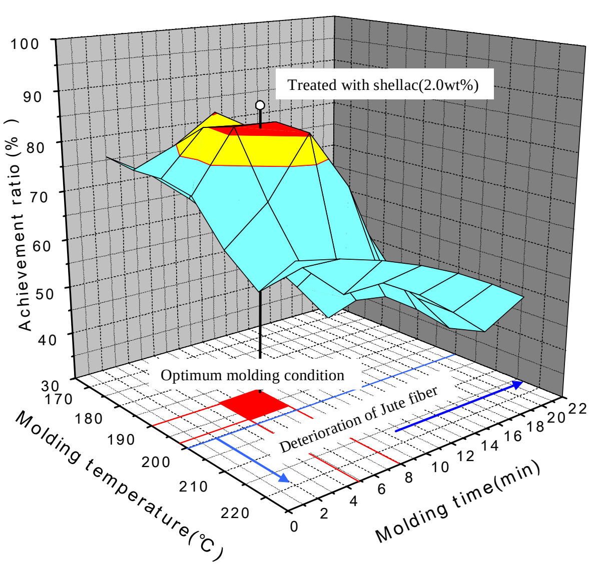

Fig. 2 Relationship achievement ratio and molding conditions.

SURFACE TREATMENT WITH SHELLAC RESIN: EFFECT OF CONCENTRATIONS OF SHELLAC RESIN

Coated jute spun yarn with various concentrations of aqueous solution of shellac resin/PLA micro-braided yarns were molded under the optimum molding condition (molding pressure of 1.33 MPa, molding temperature of 190^{o}C and molding time of 8 min.). Relationship between tensile properties of the specimen and the concentration of the aqueous solution of shellac resin is shown in Fig. 3. The achievement ratios of elastic modulus and tensile strength were increased from 0.5wt % until 2.0wt % and decreased over 2.0wt %.

From result of cross sectional observation, in the case of 0wt % and 0.5wt %, the area of fiber bundle was almost same. Whereas, in the case of 5.0wt %, the fiber bundle area was 90 % of other concentrations, namely, dispersion of the jute fibers could not be seen. Observation of fracture surface was performed using SEM to investigate the condition of the interface of the specimen. In the case of 0wt % and 0.5wt %, adhesion of the resin on the jute fiber could not be seen. In the case of 2.0wt %, adhesion of the resin on jute fiber was observed. In the case of 5.0wt %, more amount of resin on the jute fiber than 2.0wt % was obtained.

From these results, tensile properties of 2.0wt % were improved by increasing the interface property. Tensile properties of 5.0wt % became low without dispersion of the jute fibers since jute fiber bundles were bound with shellac resin, and PLA resin could not impregnate the fiber bundles. The ratio of tensile strength with the concentration of 2.0wt % was added in Fig. 2. In the case of the jute yarn treated with aqueous solution of shellac resin, the ratio of tensile strength was improved.

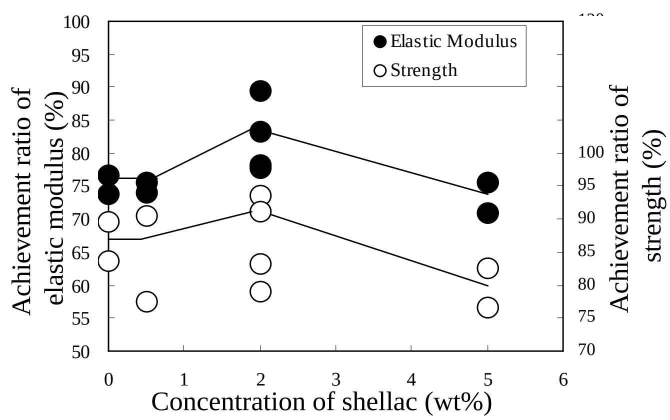

Fig. 3 Relationship between achievement ratio of mechanical properties and concentration of shellac resin.

EFFECT OF DRYING TIME OF SHELLAC RESIN

Coated Jute spun yarn with 2.0wt % of aqueous solution of shellac resin was dried during various drying times, and then the coated jute spun yarn/PLA micro-braided yarns were molded under the optimum molding condition. The ratios of elastic modulus and tensile strength remained almost the same for various drying times. From the observation of cross-sectional area and fracture surface, the specimens for any drying conditions were almost in the same condition. Therefore, in the case of surface treatment by using 2.0wt % of aqueous solution of shellac resin, the drying time was not so important.

OPTIMUM SURFACE TREATMENT CONDITION

Relationship between the achievement ratio of tensile strength and surface treatment conditions (concentration of shellac resin and drying time) is shown in Fig. 4. Because the strength was affected by improving of interfacial property and dispersion of jute fibers, optimum molding condition was limited. For example, the surface treatment condition that the achievement ratio of tensile strength became higher than 90 % was that concentration of shellac resin was around 2.0wt %.

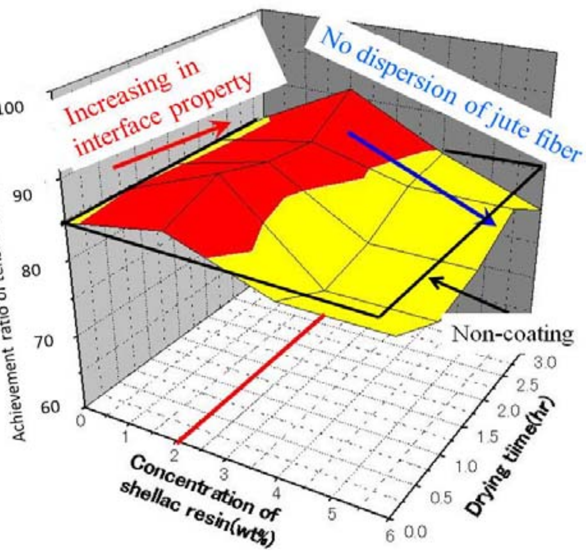

Fig. 4 Relationship between achievement ratio of tensile strength and surface treatment conditions.

BENDING TEST RESULT

0 degree and 90 degrees bending tests were performed to clarify that the surface property was improved by using surface treatment with shellac resin. Fig. 5a, b shows the relationship between 0 degree and 90 degrees bending properties and concentration of shellac resin.

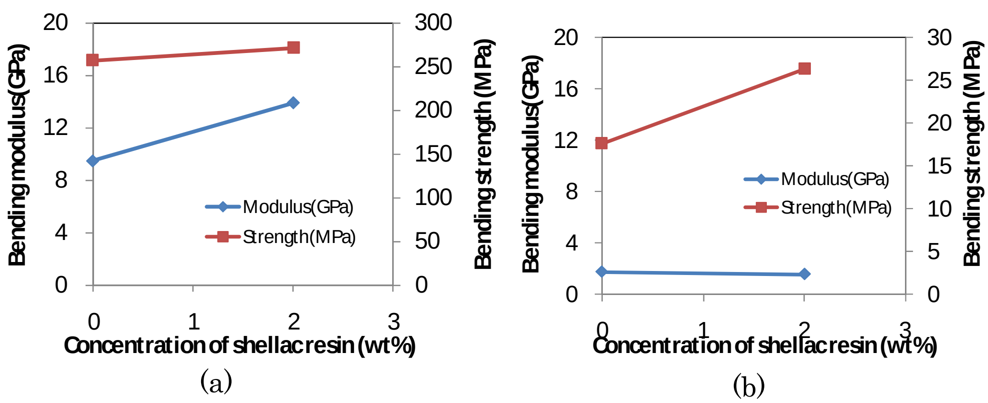

Fig. 5 Relationship between bending properties and concentration of shellac resin: (a) 0 degree direction to reinforcement fibers, (b) 90 degrees direction to reinforcement fibers.

The bending strength of surface treated with 2.0wt % shellac resin was higher than that of without shellac resin surface treatment. In the case of 90 degrees bending test, the difference of the bending strength of without shellac resin surface treatment and 2.0wt % shellac resin surface treatment appeared conspicuously. From these result, it was clarified interfacial property were increased by shellac resin surface treatment.

WOVEN FABRIC

The Jute/PLA MBY woven fabric with a 18(warp) x 14(weft) per inch was fabricated by using weaving technique. Then, the jute/PLA woven fabric composite plate was molded by compression molding in optimum molding condition (in molding pressure of 1.33 MPa, molding temperature of 190^{o}C and molding time of 8 min.). Fig. 6 shows the fabrication method of jute/PLA woven fabric composite.

Weaving machine

Woven fabric

Compression molding

Fig.6 Fabrication of woven fabric composite.

Table 1 shows the mechanical properties of jute/PLA woven fabric composite. It was clarified the bending properties of warp direction were lower than that of weft direction. The bending properties of woven fabric composite in Warp direction were lower than that in Weft direction because during molding the fiber density of weft became low due to the resin flowing in warp direction only.

Table 1 Mechanical properties of jute/PLA woven fabric composites

	Elastic modulus (GPa)	Tensile strength (MPa)	Bending modulus (GPa)	Bending strength (MPa)
Warp direction	17.5	61	4.7	142
Weft direction	-	-	2.2	84

CONCLUSION

By combining spun yarn and micro-braided yarn, problems in natural fiber reinforced thermoplastic resin composites such as low mechanical properties and difficulty in the impregnation could be solved. Because the strength was affected by impregnation of resin into fiber bundle, heat deterioration of jute fibers and dispersion of jute fibers, optimum molding condition was limited. The tensile properties of treated reinforcement fiber with shellac resin were improved. But, the jute fiber could not be dispersed when the amount of shellac on the reinforcement fiber was too much. The jute fiber was binded with shellac resin, and PLA resin could not be impregnated into the jute fiber bundle. It was clarified that there is the optimum concentration of the shellac resin. From these results of bending tests, it was clarified interfacial property were increased by shellac resin surface treatment. Furthermore, the woven fabric reinforced composite could be molded by using combination of braiding and weaving techniques.

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