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Tensile and impact properties of fully green composites reinforced with mercerized ramie fibers

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Abstract The purpose of this study is to create a natural fiber-reinforced fully green composite with excellent toughness. By treating ramie plied yarns in a high concentration alkali solution, the reinforcements were mercerized. Results of tensile tests showed that unidirectional composites using mercerized ramie yarns exhibited two to three times larger fracture strain, without a marked decrease in strength, than composites using untreated yarns. In addition, mercerization for the ramie yarns brings a better interfacial strength to the composites. Laminated composites using mercerized ramie yarns also showed approximately twice larger impact energy than composites using untreated yarns. Thus, mercerization for natural fibers is expected for application to mechanical materials requiring a high toughness.

Introduction

Fully green composites [1] receive great attention nowadays because they can eliminate some environmental problems. Generally glass-fiber-reinforced plastics (GFRP) have enhanced thermal and mechanical properties, and because of such properties it is not easy to dispose of GFRP products after use. For that background, natural fiber-reinforced composites have recently been developed, in which plant-based high strength natural fibers are used in place of glass fibers [2, 3]. Although the natural fiber strength is inferior to glass-fiber strength, mechanical properties of natural fibers can be improved through mechanical and/or chemical treatments [4, 5]. Especially, it is expected that, through such treatments, the fibers will exhibit the high toughness necessary for structural components.

The purpose of this study is to create natural fiber-reinforced fully green composites with excellent toughness that is comparable to that of GFRP. In this study, toughness improvement in tensile and impact properties of the composites is observed after reinforcement with mercerized ramie fibers.

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Experimental procedure

Test materials and mercerization

A plied yarn of ramie fibers (ramie No. 16, five twists, TOSCO Co.), a representative high strength plant-based fiber, has been used for reinforcement of fully green composites. Table 1 shows physical and chemical properties of untreated ramie fibers [4]. According to Doi et al. [5] mercerized ramie fibers without any load application decrease in tensile strength, but their fracture strain increases dramatically, as compared to untreated ramie fibers, as shown in Table 2. This change also raised the toughness of the fibers. The present article thus explores if mercerized ramie yarns can raise tensile and impact properties of the composites.

Biodegradable thermoplastic resin (Randy CP-300, Miyoshi Oil and Fat Co. Ltd, Japan) has been used as a matrix material of the composites, which decomposes easily and naturally in soils. This hydrophilic resin is made from a

Table 1 Physical and chemical properties of untreated ramie fibers [4]

Density (Mg/m^3)	Cellulose (wt%)	Lignin (wt%)	Hemi-cellulose (wt%)	Pectin (wt%)	Wax (wt%)	Microfibrillar angle (°)	Moisture content (wt%)
1.50	68.6–76.2	0.6–0.7	13.1–16.7	1.9	0.3	7.5	8.0

Table 2 Tensile properties of untreated and mercerized ramie fibers [5]

Untreated			Mercerized[a]		
Fiber diameter (μm)	Tensile strength (MPa)	Fracture strain (%)	Fiber diameter (μm)	Tensile strength (MPa)	Fracture strain (%)
30.9	610	3.59	29.1	420	8.11

[a] Without load application

Table 3 Properties of cornstarch-based biodegradable resin

Density (Mg/m^3)	Melting point (°C)	Tensile strength (MPa)	Fracture strain (%)	Young's modulus (GPa)
1.16	58	10.6	6.5	0.531

blend of polycaprolactone and cornstarch. The resin was supplied in a water emulsion with micro-order particles. Mechanical properties of this resin are shown in Table 3.

For the purpose of toughening the reinforcement, the yarns were mercerized directly in alkali solution; then some yarns were done under load application in the same solution, because it is expected that this mechanical treatment does not decrease the yarn strength, as seen in ramie single fibers [5]. The alkali solution used was 15 wt% sodium hydroxide solution. The treatment time was 2 h. In the treatment under load application, a 37.2 N weight was applied to one end of the yarn suspended from a 2.0 m-height panel. Then the 15% sodium hydroxide solution was syringed from the top end of the yarn every 15 min for 2 h. Mercerized yarns with and without load application were washed in water containing a small quantity of acetic acid and then dried.

Composite fabrication method

Untreated and mercerized yarns were wound around a thin metallic plate, and the matrix resin was pasted onto the yarn. Thereafter, it was dried and the yarn was cut as a pre-formed sheet. For the longitudinal tensile specimen, the sheet size was 15 mm wide and 100 mm long, in which the yarn was placed longitudinally. Two pre-formed sheets were placed into a metal die, and a slight pressure was applied at 150 °C using compression molding equipment. After evaporating water in the pre-formed sheets, the heating process was stopped; the pre-formed sheets were then pressed at 13.1 MPa. Pressure was applied until the temperature decreased to near room temperature. For the transverse tensile specimen, two pre-formed sheets were prepared in 100 mm × 100 mm size. Two pre-formed sheets were placed into a square metallic die and pressed in the same condition as the longitudinal tensile specimen. The fabricated specimen was then cut to 15 mm width, perpendicular to the fiber direction.

For the impact specimen, pre-formed sheets were prepared in 100 mm × 100 mm size in the same way as the transverse tensile specimen. In this case, three pre-formed sheets were placed with 0°/90°/0° lamination into the metallic die and pressed in the same condition as the tensile specimens. Additionally, plain woven fabrics of mercerized ramie yarns without load application were prepared by hand in 100 mm × 100 mm size as reinforcement. The fabric density was 9 warps per inch and 12 wefts per inch. The matrix resin was pasted onto the fabrics in the manner described above; it was then dried to produce pre-formed sheets. In order to vary water contents in the composites, some fabrics were placed directly into the die, the matrix resin was poured onto the fabrics, and these were pressed. In this case also, the temperature and press condition were identical to those described above.

Tensile and impact tests

Figure 1 shows the configuration and dimensions of longitudinal and transverse tensile specimens. Aluminum plates with 0.8 mm thickness were attached with epoxy adhesive on both ends of the composite. The gage length and width were 50 and 15 mm, respectively. On the center of the specimen surface, a strain gage was attached to measure uniaxial strain along the longitudinal direction.

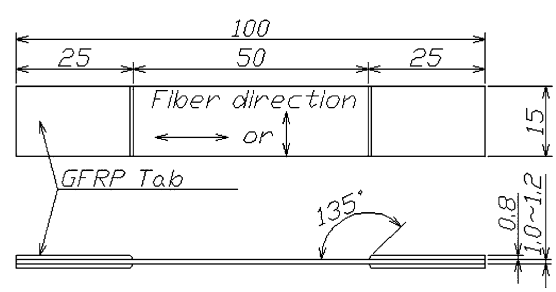

Fig. 1 Shape and dimensions of tensile specimen

Tensile testing of the specimens was carried out at cross-head speeds of 0.5, 5.0, and 100 mm/min using an Instron-type testing machine (Autograph IS-5000, Shimadzu Co. Ltd). These cross-head speeds correspond to strain rates of 0.01, 0.1, and 2.0/min.

Impact tests were also carried out under the potential energy of 7.36, 24.5, or 49.1 J using a drop-weight impact testing machine (IITM-23, Yonekura Mfg Co. Ltd). The impact specimen was fixed and given the impact test area of a circle of 80 mm diameter circle. The striker, with of 10 mm diameter hemisphere, was used for testing. The impact load was measured using an acceleration sensor attached near the striker. The impact energy was calculated from the area under the diagram of the impact load and striker's displacement, in case the striker completely penetrated the specimen.

Results and discussion

Tensile properties of mercerized ramie-fiber-reinforced green composites

Three unidirectional green composites reinforced by untreated ramie yarns, mercerized ramie yarns without load application, and mercerized ramie yarns with load application, denoted, respectively, as UT, T-0 and T-4, were tensile tested. Table 4 shows results of tensile tests for UT, T-0, and T-4 composites in the strain rate of 0.1/min; Figure 2a–c show typical stress–strain diagrams of these three composites. Values in Table 4 are all averages. The results show that the UT composite indicates almost linear behavior, similarly to that of untreated fibers [4, 5]; this composite is broken at around 2% strain. Such brittle behavior was also seen in other green composites reinforced with natural fibers, such as craua [6], flax [7], and Manila hemp [8], which had not been treated in alkali solution. On the other hand, the T-4 composite behaves with nonlinearity beyond 1% strain, similarly to the single-fiber behavior [4], and is broken at greater than 4% strain. The tensile strength of T-4 composites was about 300 MPa, and the Young's modulus was 21.6 GPa, which were

Table 4 Longitudinal tensile properties of unidirectional green composites reinforced with UT, T-0, and T-4 yarns in the strain rate of 0.1/min

Composite type	Number of samples	Fiber volume fraction (%)	Tensile strength (MPa)	Fracture strain (%)	Young's modulus (GPa)
UT	6	58.2	309	2.45	24.0
T-0	5	66.1	284	5.69	15.3
T-4	5	67.2	303	4.06	21.6

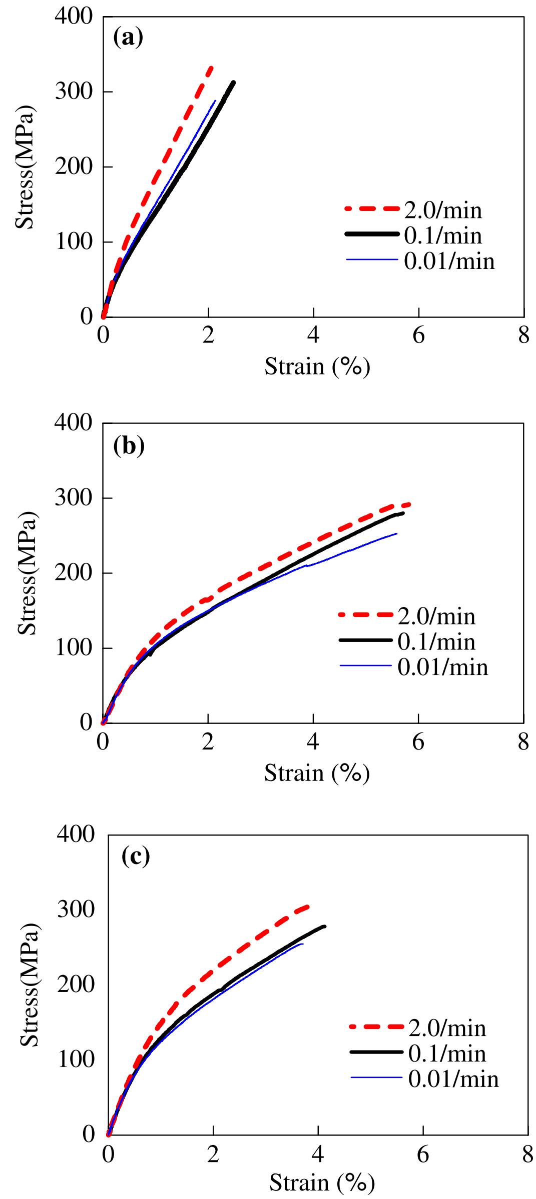

Fig. 2 Typical stress–strain diagrams of fully green composites reinforced by **a** UT yarns, **b** T-0 yarns, and **c** T-4 yarns

almost comparable to those of the UT composite. On the other hand, the T-0 composite behaves with nonlinearity earlier than the T-4 composite. It deforms greatly and is broken at more than 6% strain. Young's modulus of T-0 composite was 15.3 GPa, less than that of UT and T-4 composites. The strength of T-0 composite was also decreased in comparison to that of UT and T-4 composites.

Broken specimens revealed that large deformations in T-0 and T-4 composites arose from their plastic strains. The mechanism of plastic deformation in mercerized fibers is explained from slippage between cellulose microfibrils of the fibers, which occurs because of partial remove of hemicellulose through alkali treatment [4]. Because the area under the stress–strain diagram obtained from tensile test expresses the degree of toughness, such plastic deformation engenders increased toughness of the composites. Therefore, the results depicted in Fig. 2 show that mercerized ramie yarns with and without load application impart a dramatic toughness increase to fully green composites.

Regarding the effect of the strain rate on mechanical properties of the composites, all composites showed a similar effect. Fracture strain is unaffected by the strain rate, irrespective of a large change in the strain rate, although the composite stress increases somewhat with an increased strain rate. The toughness of the composites increases with an increased strain rate. The effect of the strain rate on the matrix strength was also investigated. In this case, the specimen size of the matrix resin was equal to that of the composite specimen. The result showed that the matrix strength was 8–14 MPa. This variation is too small to change the composite stress in Fig. 2 because the matrix volume fraction is only 30–40% in the composites. Therefore, the effect of the strain rate on the composite stress and strength must be affected by the reinforcement.

Averages of tensile properties of transverse tensile specimens are shown in Table 5. The strain rate of transverse tensile specimens was 0.01/min. The Young's modulus of the T-0 composite is not so different from that of the UT composite, but the tensile strength is different. The tensile strength of UT composites perpendicular to the fiber direction is 8.2 MPa, but that of T-0 composites is 9.6 MPa, about 10% higher than the UT composite. It can be concluded from this result that mercerization can increase the bonding strength between the fibers and matrix resin, as well as increase the fracture strain of the longitudinal tensile specimen. Table 2, the mercerized fibers decrease in strength, about 30% less than the untreated fibers. However, the decreasing rate in strength of T-0 composites, which is reinforced with the mercerized fibers, is only 8%, as calculated from Table 4. It is considered that this improvement is caused by the enhanced interfacial strength mentioned above.

Table 5 Transverse tensile properties of unidirectional green composites reinforced with UT and T-0 yarns in the strain rate of 0.1/min

Composite type	Number of samples	Fiber volume fraction (%)	Tensile strength (MPa)	Fracture strain (%)	Young's modulus (GPa)
UT	4	64.3	8.23	0.759	1.85
T-0	4	51.5	9.63	0.787	1.78

Impact properties of mercerized ramie-fiber-reinforced green composites

Effects of mercerization on the impact properties of a ramie-fiber-reinforced green composite were explored. Table 6 shows maximum loads and impact energies of the laminated composites with unidirectional layers using untreated and mercerized ramie yarns. To determine the effect of mercerization, the mercerized yarn without load application was used as the reinforcement of a laminated composite. These laminated composites are denoted as UT laminate and T-0 laminate, respectively. As shown in the table, the UT laminate was penetrated with the potential energy of 7.36 J, and the impact energy was 5.71 J. But these T-0 laminate specimens were not penetrated despite the same potential energy. Therefore, the energy was increased to 24.5 J, and the impact test was carried out using a new specimen of T-0 laminate. In this case, T-0 laminate was penetrated with the impact energy of 12.7 J. That is to say, toughness improvement through mercerization for ramie yarns increases their impact properties to almost twice as high as that of laminated composites reinforced with untreated yarns.

Figure 3a and b, respectively, show the representative damage morphology of UT and T-0 laminates that suffered penetration. The UT laminate is damaged with yarn breaks perpendicularly to the yarn direction, as shown in arrows of Fig. 3a, which means that the yarn cannot be deformed greatly. In contrast, the T-0 laminate is damaged with an interfacial crack of about 60 mm length between yarn fibers without any yarn breakage, which signifies that the yarn absorbs more impact energy because of toughness improvement caused by mercerization. Consequently, mercerization for ramie yarns increases impact properties of the composites.

Effect of water content on impact properties of mercerized ramie fiber reinforced green composites

Fully green composites are hydrophilic. Therefore, the effect of water content on their mechanical properties must be discussed. In this study, to clarify the effect of water

Table 6 Impact properties of laminated composites reinforced with UT and T-0 yarns

	Number of samples	Thickness (mm)	Fiber volume fraction (%)	Maximum load (kN)	Impact response	Impact energy (J)
UT laminate	2	2.24	42.7	0.699	Penetration	5.71
T-0 laminate	3	2.33	55.9	1.90	Rebound	–
	1	2.21	57.1	1.95	Penetration	12.7

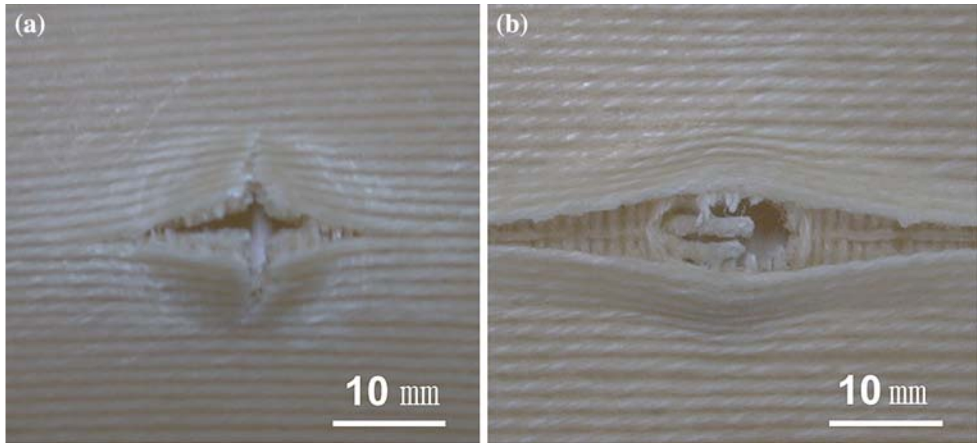

Fig. 3 Impact damage morphology of fully green composites reinforced by **a** UT yarns and **b** T-0 yarns

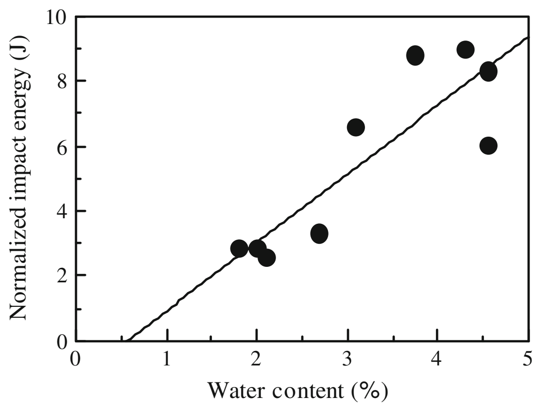

Fig. 4 Effect of water content of fully green composites reinforced with mercerized ramie woven fabrics

content on the impact properties of the composites, laminated composites reinforced with plain woven fabrics of ramie yarns were additionally impact-tested. Water contents were measured through an electronic moisture analyzer. Figure 4 shows the effect of water content (wt%) on impact energy of the composites, in which the perpendicular axis was normalized by dividing the impact energy by the number of layers. In this study, the water content of the composites varied from 2% to 5%. The result shows that higher water contents increase the normalized impact energy of the composites. It was observed from the specimens that the matrix resin was softened with increased water content. Such softening of the matrix resin increases the impact properties, but softening often reduces deformation resistance and strength of the composites. That is to say, it is anticipated that the impact energy of the composites would decrease with further increased water content.

In general, the drop-weight impact energy of GFRP using glass-fiber roving cloth and epoxy resin is 25–30 J, if the thickness of the composite is about 1.6–2.0 mm [9, 10]. In this study, the impact energy of the laminated composite with three woven fabric layers was 26.5 J. The average thickness of the laminated composite with three layers was 2.67 mm, but its weight was almost equal to that of the GFRP laminate. Consequently, although these values are not directly comparable because of different matrices, fully green composites using mercerized ramie woven fabrics are comparable to practical GFRP in terms of impact properties.

Conclusion

Tensile and impact properties of green composites reinforced with mercerized ramie yarns were explored. The obtained results are summarized as follows:

(1) A unidirectional green composite reinforced by mercerized ramie yarns was improved dramatically in toughness because of the increased fracture strain of ramie fibers through mercerization. It is considered that improved interfacial bonding between the fibers and matrix through mercerization is also related to the increased toughness of the composites. In addition, the deformation resistance of the composite increased with an increased strain rate.
(2) A laminated green composite using unidirectional layer sheets made of mercerized ramie yarns absorbs almost twice as much impact energy as the composite using untreated reinforcement. Impact energies of the laminated green composites using mercerized ramie woven fabrics increased with increased water contents.
(3) The above points indicate that if mercerized ramie fibers are used as reinforcement of fully green composites, the composites can show improved toughness, especially in terms of impact properties.

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