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Examination of heat resistant tensile properties and molding conditions of green composites composed of kenaf fibers and PLA resin

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Abstract—Disposing of conventional fiber-reinforced polymers (FRPs) poses an environmentally challenging problem. Disposal of FRPs by combustion discharges carbon dioxide in the air because the resin of FRPs is made of fossil fuel. When they are disposed of in the ground, FRPs remain semi-permanently without decomposing. In response to these problems, green composites are now being developed and are extensively studied as a material that produces a lower environmental burden. In this paper, green composites using kenaf fiber yarn bundles and PLA (poly(lactic acid)) are fabricated and their tensile properties are evaluated in the experiment. The tensile Young's modulus of all of the laminations is larger than that of PLA alone and the tensile strength of some laminations is larger than that of PLA alone. In particular, the value of UD composite of 0° shows double the tensile strength of PLA alone. Furthermore, the molding conditions for fabricating with a hot press are investigated and the heat resistant tensile properties of green composites are also reported.

Keywords: Green composite; kenaf; PLA; tensile properties; heat resistance molding condition; hot press.

1. INTRODUCTION

Fiber-reinforced polymers (FRPs) pose environmental problems when they are disposed of because the resin of FRPs is made of fossil fuel. One of the problems

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is that carbon dioxide increases in the air owing to the combustion disposal of FRPs. Another problem is that the FRPs remain without decomposing in the ground because of the semi-permanent durability of FRPs. Therefore, a development of new composites made of the natural fibers and biodegradable resins is desirable. We call this new composite as a green composite.

In previous papers about the green composites reinforced by kenaf fibers, the water treatments on the surface of kenaf fibers were examined [1] and their effect on the flexural properties of the green composite using poly(lactic acid) (PLA) resin was discussed. The tensile strength of the green composite using an immersion type of PLA was examined [2]. The mechanical properties of the green composite were investigated [3], in which immersion type of PLA and the short kenaf fibers with the high aspect ratio composed the green composite. The composites of unsaturated polyester (UP) resin reinforced by long kenaf fibers [4] or chopped kenaf fibers [5] were fabricated with a hand layup or a injection molding. Their mechanical properties were also reported.

In this paper, the molding conditions for fabricating green composite are examined, in which the kenaf fiber yarn bundles as the natural fiber and PLA as the biodegradable resin are prepared. The mechanical properties of green composite fabricated with a hot press are investigated. Various kinds of surface treatments are carried out on the surface of kenaf fiber yarn bundles and these effects on the transverse strength of composite are also reported. Finally, the heat resistant tensile properties of green composites are examined.

2. SPECIMENS

2.1. Natural fibers

Kenaf fibers are made of kenaf bast. A kenaf textile (Unipacs CO.) is made of kenaf fiber yarn bundles and it is shown in Fig. 1. The interval of the fiber yarn bundles of textile was 1.9 mm and this textile was used as the reinforcement in the cross-ply

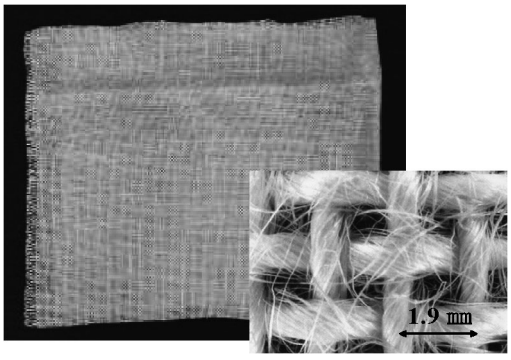

Figure 1. Kenaf textile.

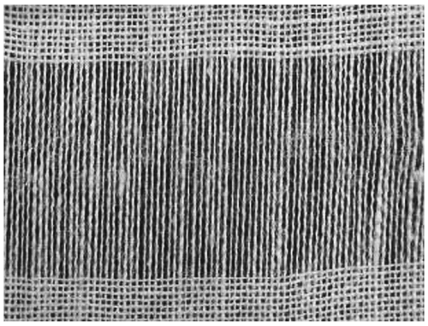

Figure 2. UD kenaf sheet.

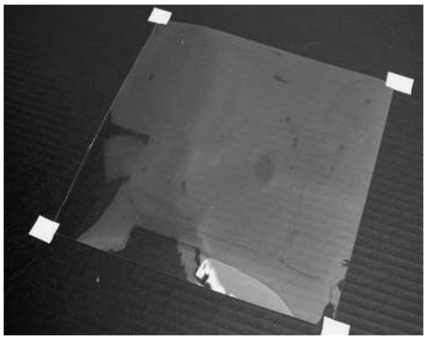

Figure 3. PLA sheet.

and quasi-isotropic composites. The unidirectional (UD) kenaf sheet (Fig. 2) was made by pulling out the weft kenaf yarn bundles from the textile. The UD kenaf sheets were used as the reinforcement in the UD reinforced composite.

2.2. Biodegradable resin

PLA resin used as the matrix of the composites is Laycia H-440 (Mitui Chemicals) and is formed into a sheet of 0.25 mm thickness (Fig. 3). In order to find the molding temperature of green composite, the thermal properties of PLA were examined by the use of a DSC instrument. From the result of the test as shown in Fig. 4, the melting and glass transition temperatures were 148°C and 58°C, respectively, and the resolution of PLA was not recognized until 200°C.

2.3. Laminates

Three kinds of laminated green composite plates were used in the experiment and their sizes were 300 × 300 ×2 mm. One kenaf textile was inserted between PLA sheets and then the cross-ply and quasi-isotropic laminated plates were composed of five PLA sheets and four kenaf textiles. For the case of UD laminated plate, two UD kenaf sheets were inserted between the PLA sheets. As a result, the fiber volume content was kept at a constant value of 38% for all the laminated plates.

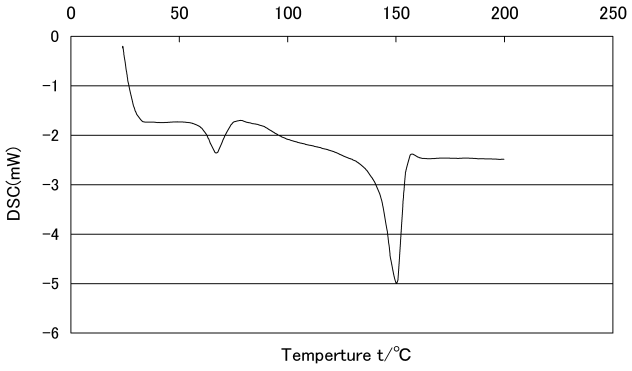

Figure 4. Results of DSC for PLA.

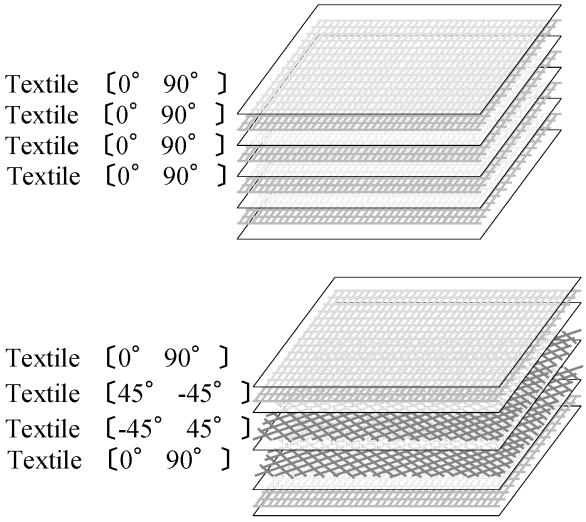

Figure 5. Laminate composition of textile composite A (top) and B (bottom).

Four kenaf textiles were laminated in the same direction. This textile composite with the fiber direction of 0° and 90° cross-ply was designated as Textile Composite A (Fig. 5, top).

The two kenaf textiles inside were inclined in the 45° and −45° directions to the two kenaf textiles outside. This composite has the quasi-isotropic property and was designated as Textile Composite B (Fig. 5, bottom).

Eight UD kenaf sheets were used and a pair of two UD sheets was inserted in the same direction between the PLA sheets for keeping the same fiber volume fraction as those of other composites. Three kinds of specimens (UD composites of 0°, 45° and 90°) were obtained from the UD composite by changing the cutting direction (Fig. 6).

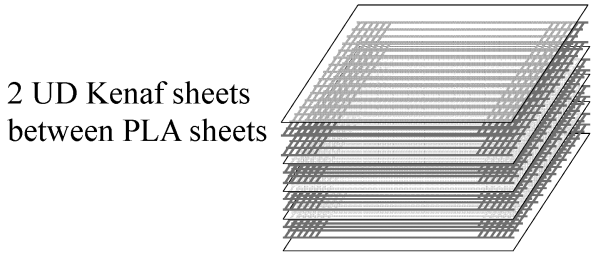

Figure 6. Laminate composition of UD composite.

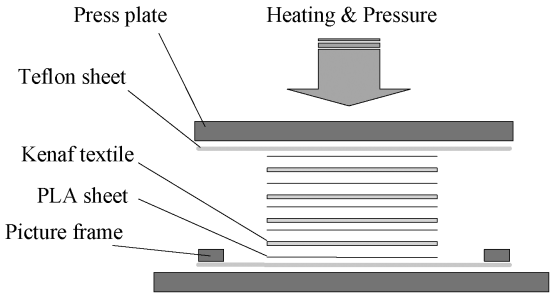

Figure 7. Hot press molding of green composite.

Table 1.
Molding condition of hot press

Process	Clearance removal ⇨	Melting ⇨	Impregnation ⇨	Cooling
Temperature [°C]	185			−5 [°C/min]
Pressure [MPa]	10	1	10	1
Time	10 s	20 min	10 s	

The laminated plates composed of PLA and kenaf sheets stated above were molded with a hot press (Fig. 7). From the result of DSC, the temperature and time of the melting process were fixed at 185°C and for 20 min under 1 MPa (Table 1).

3. STATIC TENSILE TESTS

3.1. Method of static tensile tests

The test pieces were cut into 250 × 25 × 2 mm specimens from the plate as shown in Fig. 8 by using a precision sawing machine. The tabs were bonded to both ends. The static tensile test was executed based on the JIS K7113 and the test speed was 1 mm/min.

The results of the tensile test are shown in Fig. 9. The tensile strength of Textile Composite A (cross-ply laminate) was higher than that of PLA alone. However,

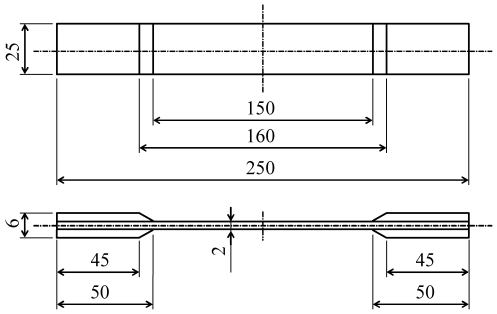

Figure 8. Dimension of tensile test specimen.

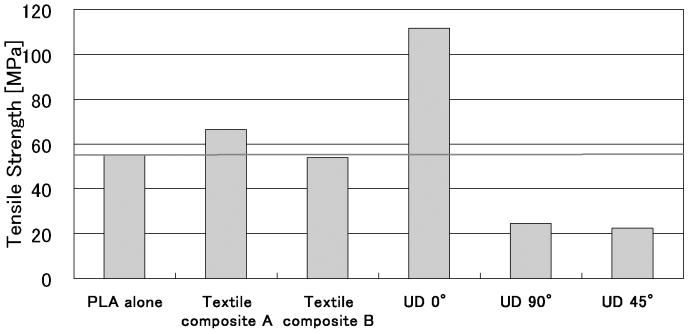

Figure 9. Tensile strength of specimens.

the increase of the tensile strength was not as much higher as the expected value. For the quasi-isotropic Textile Composite B, its strength was a little smaller than that of PLA alone. On the other hand, the tensile strength of the UD composite of 0° was significantly larger than that of PLA alone, which was double the tensile strength of PLA. The strength of the UD composite of 90° decreased considerably more than that of PLA alone, which was one-half the tensile strength of PLA. The specimen of 45° showed about the same results as the UD composite of 90°. The reason for the strength reduction of the UD composites of 90° could be explained by the observation of fracture modes.

However, the tensile Young's modulus for all the specimens was larger than that of PLA alone as shown in Fig. 10.

3.2. Fracture surface observation

Figure 11 shows the failure aspects of Textile Composite A and the UD composites of 0° and 90° after the test. The fracture aspect of the UD 0° showed zigzag breakage of fiber bundles. In order to produce continuous fibers, the kenaf fiber yarn bundles were twisted and interlaced and this twisting effect contributed to an increase of the tensile strength for the UD composite of 0°. The pull-out of the twisted yarn bundles from the resin seemed to receive a larger resistance.

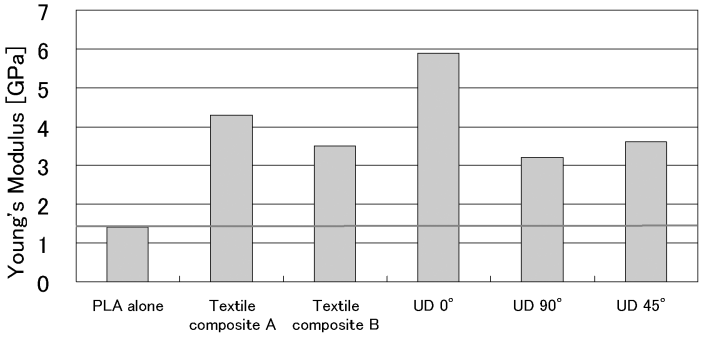

Figure 10. Young's modulus of specimens.

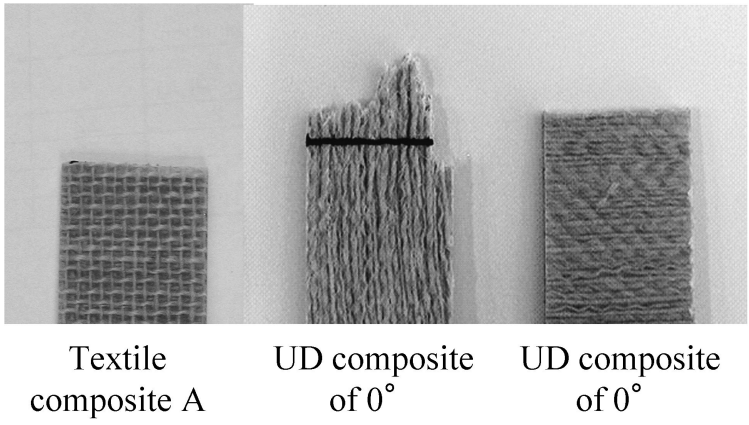

Figure 11. Typical aspects of tensile failure.

Textile Composite A and the UD composite of 90° broke along the fiber bundles of 90°. The optical photograph by a micro-scope showed the perfectly exposed the fiber bundles of the UD composite of 90° as shown in Fig. 12. Since the interface strength between the transverse fiber bundles and the matrix of PLA was not strong enough, the strength of UD composite of 90° was smaller than that of PLA.

4. IMPROVEMENT OF INTERFACE STRENGTH

4.1. Methods of surface treatments

In order to examine the interface strength of the UD composite of 90°, simple specimens were fabricated in which one UD kenaf sheet and 8 PLA sheets were laminated. The surface of the UD kenaf sheet received the following treatments:

(1) Boiling treatment: The UD kenaf sheet was put into the distilled and boiled water for 5 min, 30 min and 3 h, respectively, and then dried in the air.

(2) Silane treatment: The UD kenaf sheet was soaked in 2% aminosilane solution for 5 min and then dried in the air.

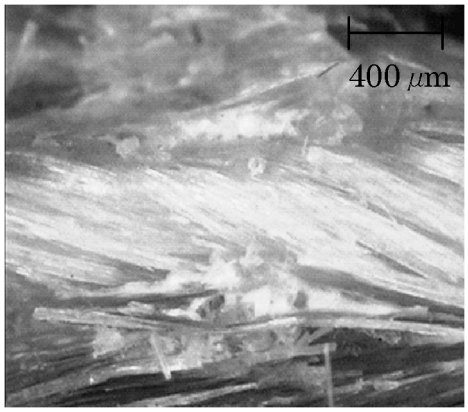

Figure 12. Failure aspect of UD composite of 90°.

(3) Solvent washing: The UD kenaf sheet was soaked in 99.5% ethanol as solvent for 5 min and then dried in the air.

4.2. Test result

The tensile properties of the simple specimens with surface treatments are listed in Table 2 and that of the simple UD composite of 90° without surface treatments is also listed in order to compare the effects of each surface treatment.

The tensile strength of the specimen treated by silane did not improve due to the discoloration during the heating process of molding. Otherwise, the strength of the specimen with boiling treatment for 30 min and with solvent washing showed a larger value. For the Young's modulus, no distinct change was observed among the specimens subjected to various surface treatments. Consequently, it was confirmed that the solvent washing and the boiling treatment for 30 min were effective for the improvement of interfacial adhesion between PLA and kenaf fibers.

All of the specimens with solvent washed textiles were fabricated by the same method illustrated in Figs 5 and 6. Table 3 lists the comparison of these specimens with and without solvent washing for the tensile strength. The strengths of Textile Composites A and B with solvent washing were larger than those without solvent washing and the strength of Textile Composite B was larger than that of PLA alone.

However, the strengths of the UD composites of 0° and 90° were smaller than those without solvent washing. The comparison of the cross-sections in the UD composite of 0° and Textile Composite A with a microscope is shown in Fig. 13. In the UD composite of 0° specimen, the fiber bundles expanded compared with those of Textile Composite A by the effect of solvent washing and the fiber yarn bundles of 0° specimen were loosened without the constrain of transverse fibers. The failure aspects of UD composite of 0° specimens with and without solvent washing are compared in Fig. 14. The fiber bundles of the fractured the UD composite of 0° specimen with solvent washing showed the combination of pullout and breakage of

Table 2.
Results of surface treated specimens

Treatment method	Tensile strength [MPa]	Young's modulus [Gpa]	Breaking strain [%]
Not treated	31.6	2.51	1.31
Boiling (5min) + dry	32.6	2.55	0.73
Boiling (30min) + dry	41.6	2.45	2.01
Boiling (3h) + dry	34.2	2.75	0.84
Silane + dry	17.8	2.55	0.73
Boiling (30 min) and silane + dry	24.5	2.80	0.99
Solvent + dry	41.3	2.48	2.0

Table 3.
Comparisons of tensile strength of PLA not treated and ethanol treated

	Method of processing	Tensile strength [MPa]	Young's modulus [GPa]	Breaking strain [%]
PLA alone		55.4	1.4	4.8
Textile	not treated	66.5	4.2	2.3
composite A	ethanol	70.4	5.3	1.9
Textile	not treated	54.0	3.5	2.2
composite B	ethanol	63.1	4.7	2.0
UD composite	not treated	111.6	5.9	2.7
of 0°	ethanol	104.9	6.1	2.4
UD composite	not treated	24.5	3.2	1.0
of 45°	ethanol	37.7	4.1	1.1
UD composite of 90°	not treated	22.4	3.6	0.9
	ethanol	14.9	2.8	0.5

the fiber bundles. Otherwise, most of the fiber bundles showed the breakage in the UD composite of 0° specimen without solvent washing.

5. EXAMINATION OF MOLDING CONDITION

5.1. Melting temperature and time

In the molding conditions listed in Table 1, the temperature and duration of the melting process were fixed at 185°C and for 20 min under 1 MPa. Here, we examined these conditions for changing the temperature from 160 to 190°C and the duration from 5 to 25 min. After Textile Composite A was fabricated under these molding conditions, the tensile strength of these specimens was compared.

From the results as shown in Table 4, the combination of the temperature at 185°C and the duration of 15 min gave the highest tensile strength of Textile Composite A. This implies that the required temperature must be higher than the

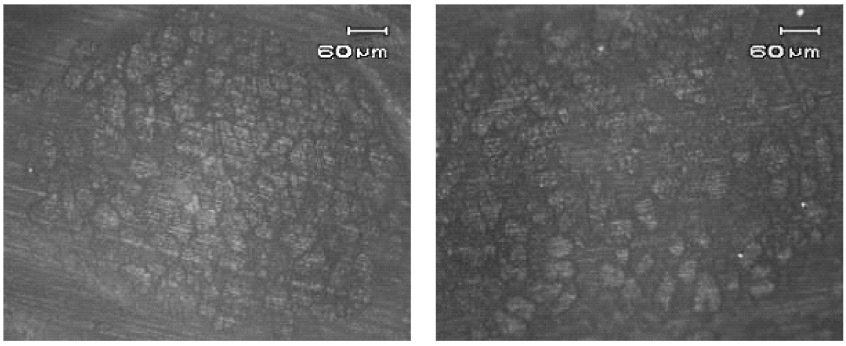

Figure 13. Comparison of cross-sections.

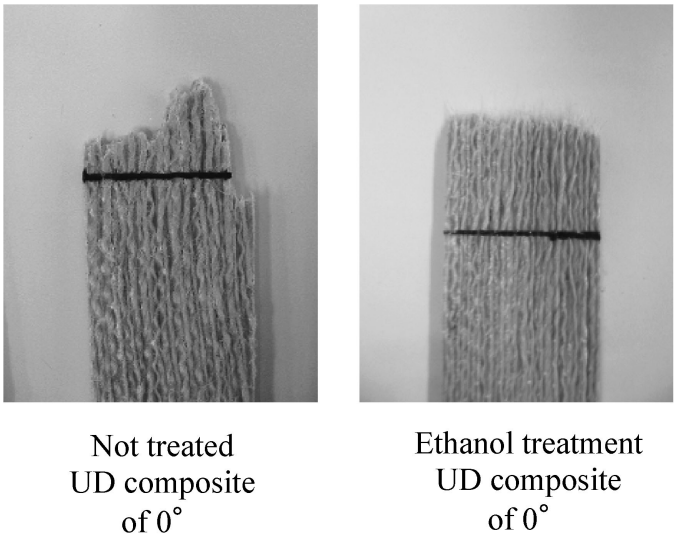

Figure 14. Comparison of tensile fracture.

melting temperature of PLA (148°C) but lower than 190°C and that the longer duration was not necessarily required to prevent searing kenaf fibers. Figure 15 shows the difference of the cross-sections of the green composites fabricated with three curing durations of 10, 15 and 20 min at 185°C. Insufficient impregnation was brought by a shorter time and a longer time caused to sear the kenaf fibers.

Table 4.
Effects of melting temperature and time on tensile strength

Melting temperature [°C]	Melting time [min]	Tensile strength [MPa]		
		Ave	MAX	MIN
160	5			
	10			
	15	70.99	72.58	68.08
	20	75.15	77.88	72.57
	25			
170	5			
	10			
	15	72.30	73.57	71.47
	20	76.57	79.69	73.83
	25	75.76	77.06	73.61
180	5			
	10	73.80	74.97	72.33
	15	74.00	76.74	69.94
	20	81.68	82.25	80.29
	25	75.79	78.96	73.76
185	5			
	10	74.22	76.12	71.57
	15	82.28	83.04	81.49
	20	67.19	70.70	65.32
	25			
190	5	72.80	76.13	71.61
	10	80.72	84.12	77.59
	15	71.63	72.83	70.28
	20			
	25			

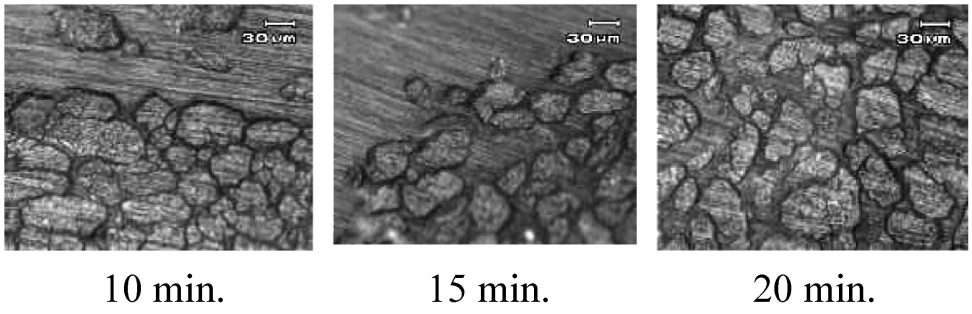

Figure 15. Comparison of cross-sections at melting temperature of 185°C.

5.2. Impregnation time

After the melting process of PLA, the impregnation process followed for a few seconds under the pressure of 10 MPa and the temperature at 185°C as shown in Table 1.

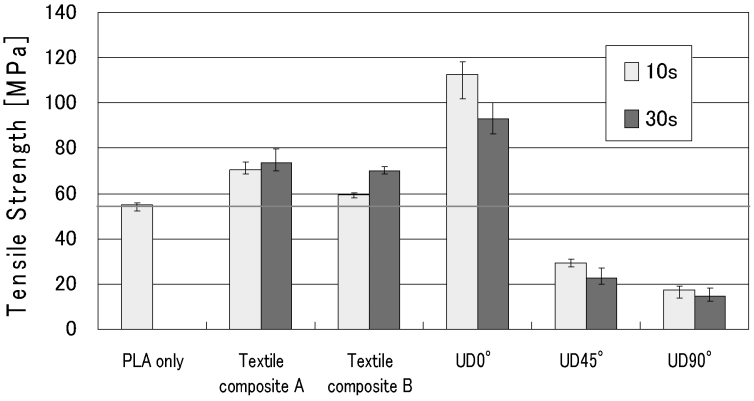

Figure 16. Difference of impregnation time for tensile strength.

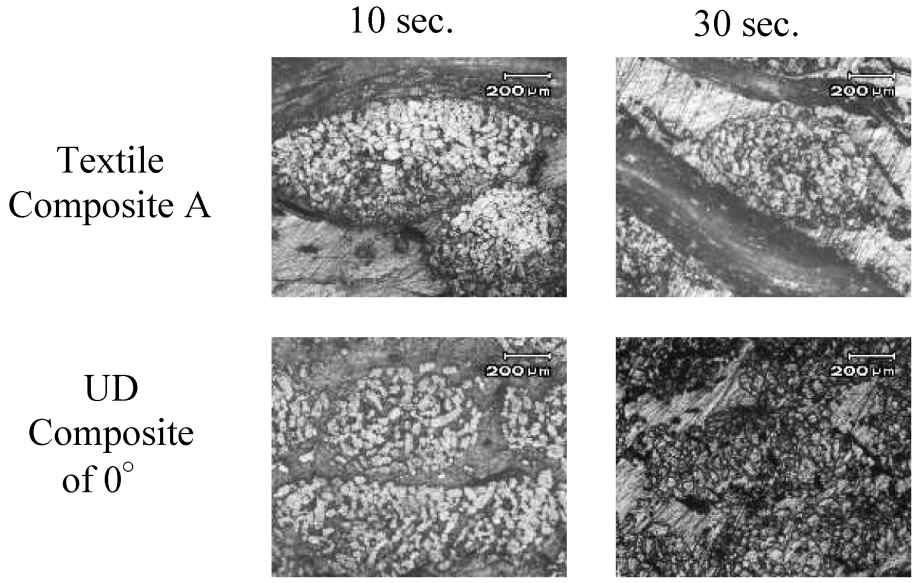

Figure 17. Comparison of specimens for impregnation time for 10 s and 30 s.

Figure 16 compares the result for 10 and 30 s of the impregnation time. The number of specimens was five for the all the cases. Their average values are shown with the bar graphs and the vertical thin line indicates the highest and the lowest values of strength. In the Textile Composites of A and B, the tensile strength for the 30 s was somewhat higher than that for the 10 s. Otherwise, the tensile strength for the 10 s was larger than that for the 30 s in the UD composites of 0°, 45° and 90°. Figure 17 shows the comparison of the cross-sections of Textile Composite A and the UD composite of 0° for impregnation time of 10 and 30 s.

In Textile Composite A, sufficient impregnation was obtained with 30 s but not with 10 s. On the other hand, 10 min were sufficient for impregnation in the UD

composite of 0° and the color of fibers changed in 30 min due to searing of the fibers. As a result, the strength due to the difference of the impregnation time between 10 and 30 s was not clear under the pressure of 10 MPa and the temperature at 185°C. However, the shorter the time, the better the impregnation.

6. TENSILE STRENGTH AT THE HIGH TEMPERATURE

Since the glass transition temperature of PLA was less than 60°C, another biodegradable resin, PBS, was used as the matrix of the green composite. This resin showed the same mechanical properties until 97°C as those of room temperature. The PBS sheet with the 0.08 mm thickness was used for fabricating the green composites. Since the temperature of melting point of PBS was 114°C, the temperature of molding was determined to be 140°C.

Figures 18 and 19 show the relation of tensile strength to strain for the UD composite of 0°, PLA and PBS alone, respectively, under the various levels of temperatures. Table 5 shows the tensile strengths of PLA, PBS alone and their UD composite of 0° at the temperatures of 25, 35, 50 and 100°C. Although it was not possible to obtain the strength of PLA at 50°C, the strength of the UD composite of 0° was kept over 70% of the value at 25°C. This implies that the kenaf fibers could reinforce PLA at 50°C. At 100°C, the strengths of PLA and the UD composite of 0° could not be obtained and the kenaf fibers could not reinforce PLA any more at 100°C, because of the higher glass transition temperature of PLA.

For the case of PBS resin, the absolute values of the strength and the Young's modulus (Table 6) for the UD0° were smaller than those of PLA/Kenaf UD0° at 25°C but at 50°, they were larger than those of PLA. This property is very important for applying green composites to automotive secondary structures.

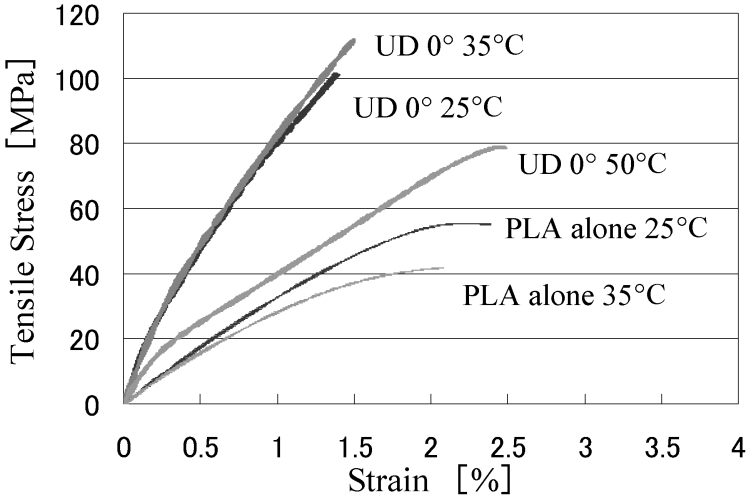

Figure 18. Relation of tensile stress to strain for PLA and its composite.

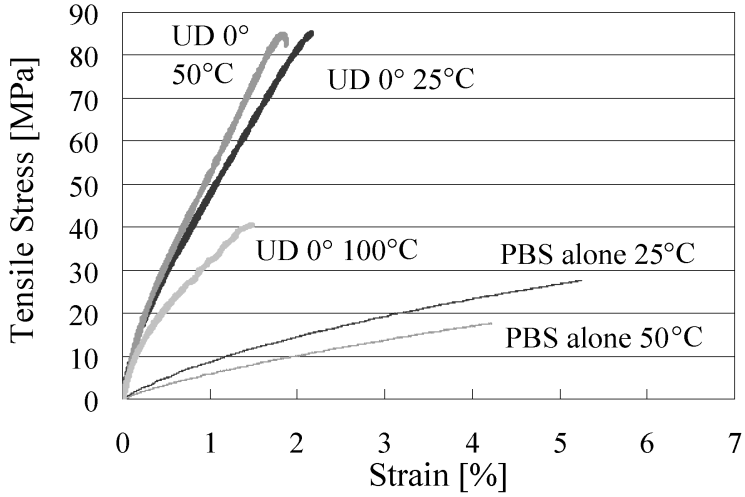

Figure 19. Relation of tensile stress to strain for PBS and its composite.

Table 5.
Tensile strengths at high temperature

Test	Tensile strength [MPa]			
temperature [°C]	PLA alone	PLA/Kenaf UD0°	PBS alone	PBS/Kenaf UD0°
25	54.9	112.3	30.4	86.2
35	42.4	117.5		
50	N.A.	81.7	23.4	85.7
100	N.A.	N.A.	N.A.	50.4

Table 6.
Young's moduli at high temperature

Test	Young's modulus [GPa]			
temperature [°C]	PLA alone	PLA/Kenaf UD0°	PBS alone	PBS/Kenaf UD0°
25	3.4	11.8	0.8	8.8
35	3.3	11.6		
50	N.A.	2.9	0.5	8.7
100	N.A.	N.A.	N.A.	7.0

7. ESTIMATION OF TENSILE YOUNG'S MODULUS OF KENAF FIBER BUNDLES

Since the kenaf fiber yarn bundles were composed of interlaced short kenaf fibers, it was hard to obtain the Young's modulus of kenaf fiber yarn bundles alone, because

of the pullout of fibers in the conventional tensile test. From the experimental tensile data of resin alone and the UD composite of 0° under the same temperatures, the tensile Young's modulus of kenaf fiber yarn bundles can be estimated by use of the rule of mixture and the fiber volume fraction.

In Table 6, the experimental tensile Young's modulus for PLA alone, PBS alone and their composites are listed. The tensile Young's modulus of PLA fiber yarn bundles was estimated to range from 22 GPa for the case of the PBS composite to 25 GPA for the PLA one. Since the kenaf fiber yarn bundles are made of natural fibers, their Young's moduli are not definite values [6], such as for glass or carbon fibers, and therefore a certain range of values must be expected.

8. CONCLUSIONS

1. The tensile Young's modulus of all of the green composites tested here was larger than that of PLA alone and this result showed the effect of the reinforcement of PLA tensile modulus by kenaf fibers.
2. The tensile strength of the composite A and the UD composite of 0° was larger than that of PLA alone. In particular, the value of UD composite of 0°showed double the tensile strength of PLA alone. However, the tensile strength of the composite B was a little smaller than that of PLA alone.
3. The boiling treatment for 30 min and the solvent washing by ethanol improved the adhesion between the kenaf fiber bundles and the PLA resin.
4. The combination of temperature at 185°C and its duration for 15 min gave the highest tensile strength for Textile Composite A in the melting process. The difference of strength due to the impregnation duration of 10 and 30 s could not be made clear under the pressure of 10 MPa and the temperature at 185°C.
5. The strength of UD composite of 0° at 50°C was kept over 70% of the value at 25°C although PLA could not sustain any tensile load at this temperature.
6. For the case of PBS resin, the absolute values of strength and Young's modulus of UD composite of 0° at 25°C were smaller but larger than those of PLA over 50°C.
7. The tensile Young's modulus of kenaf fiber yarn bundle was estimated between 22 GPa and 25 GPa.

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