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Flexural modulus of the unidirectional and random composites made from biodegradable resin and bamboo and kenaf fibres

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Abstract

Bio-based polymer composites made from kenaf, bamboo and biodegradable resin, which was a corn starch base were fabricated with press forming. The relationship between fibre Young's modulus and flexural modulus in composites was investigated. The Young's modulus of each fibre was measured to predict the flexural modulus in composites. The flexural modulus in composites was predicted by Cox model, which incorporates the effect of the fibre compression. The flexural modulus increased with increasing the fibre content. In the case of kenaf, the flexural modulus in the experimental was in good agreement with the experimental. While in the case of bamboo, the difference between experimental and calculation was large. This is because Young's modulus in bamboo was estimated considerably lower than the actual modulus due to partial breakage of bamboo during single fibre tensile test. The flexural modulus in unidirectional fibre composite made a good agreement with the predicted. However, the flexural modulus in cross ply composite was considerably lower than the predicted. This is because less fibres movement during hot pressing resulted in the resin segregation and the movement made the fibres less wetting with resin.

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1. Introduction

Recently, the bio-based polymers made from natural resources such as starch, PCL, PLLA and PHB are paid attention as petroleum based polymer alternatives. These polymers could be used as structural materials like FRP. The combination bio-based polymer and natural fibres are expected to solve this problem.

Hence, many studies on natural fibre reinforced plastics have been reported [\[1–8\].](#page-6-0) Bamboo and kenaf have been already adopted as automobile parts due to lightweight and good mechanical properties. These composites has been studied in view of mechanical properties improvement by surface treatments and compared among natural fibres

Corresponding author. E-mail address: shibata@tec.u-ryukyu.ac.jp (S. Shibata). like hemp, bagasse, jute, oil palm, ramie, etc. However, there are few studies, which have focused on the relationship between the fibre Young's modulus and the flexural modulus in the composites.

On the other hand, flexural modulus in natural fibre composites and its prediction are essential to mechanical engineering designer. This study, therefore, aims to reveal the relationship between them in the biocomposites made from bamboo and kenaf. First, Young's modulus in each fibre was measured, and the prediction of flexural modulus in the composites by Cox's model [\[9\]](#page-6-0) was performed using fibre Young's modulus, fibre volume, fibre orientation factor and fibre dimension. Simultaneously, the effect of the fibre compression was incorporated into the model. Because the authors have found previously, the fibre can be compressive and condensed in the case of bagasse fibre. Hence, the fibre compressive of kenaf and bamboo was measured. The prediction value in Cox's model, which

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incorporates the effect of the fibre compression, was compared with the experimental.

Moreover, it is well-known that the fibre orientation distribution considerably effects on the flexural modulus in the composites. In the fact, there have been many reports to fabricate strong composites using unidirectional fibre to achieve full potential of fibre. However, no reports have described the relationship between flexural modulus and its fibre Young's modulus. The flexural modulus in those composites, which perform its fibre Young's modulus full potential, definitely need the well wetting between fibre and matrix. In this reason, we examined the unidirectional fibre composites and cross ply composites to compare the calculated with the model, which incorporates the effect of the fibre orientation distribution.

2. Experimental

Kenaf and bamboo were supplied from Toyota boshoku, Japan. The kenaf fibre diameter and length was 0.1 mm diameter and 70 mm long. The fibre had been extracted by retting and dried at room temperature. Bamboo, which was 0.27 mm diameter and 300–500 mm long, had been extracted by an alkali treatment and dried also. These fibres were used as received for this work. Both fibres were cut into 10 mm long before forming. The biodegradable resin, CP-300, was a corn-starch based resin, which was a blend of starch and PCL, supplied from Miyoshi Oil Fat, Japan (Tg: -60 °C, softening temperature 55– 62 °C). The resin was chopped into uniform pellets 1– 2 mm diameter. The mechanical properties in kenaf, bamboo and resin are shown in Table 1. All these values were measured by the authors. The composite specimen was fabricated by a cylindrical steel mould and the press forming was performed at 160° C, 10 MPa for 10 min. The fibre was put into the mould without any previous mixing.

In the case of fibre oriented composites, the fabrication steps were as follows. (1) Half of fibres were put into the mold through the clearances of the slit jig as shown in Fig. 1. At this time, the bottom of the cylindrical slit jig was contacted on the mold bottom in order to keep fibre orientation. (2) The slit jig was pulled out carefully and all resin, CP-300 was put into mold. (3) The slit jig was put on the resin again, and rest of fibres was put on the resin thorough the clearance of the slit jig. The clearances between the slits were 2, 5 and 7 mm, respectively. Wider

Table 1

Fig. 1. Slit jigs for aligned fibre composites.

slit jig would be expected to produce composites with wider fibre orientation distribution. The obtained specimens were disc shaped (30 mm diameter and 1.8–1.9 mm thickness) and the fibre orientation distribution was two-dimensional. Flexural test was conducted in accordance to ISO 178 specifications on five flexural specimens. The flexural specimens $(18 \times 15 \times 1.8$ –1.9 mm) were cut out of the original specimen. Fig. 2a and b shows the random and the fibre oriented composites in the flexural test. The span length and a cross-head speed were 18 mm and 1 mm/min, respectively.

To determine the Young's modulus of kenaf and bamboo, 72 fibres were tested. The span length and cross head speed were 15 mm and 1 mm/min. Randomly chosen fibres were mounted on a paper tab with both ends of the fibre glued. The diameters were measured on three points of

Fig. 2. Appearance of flexural specimens, (a) kenaf, random (b) kenaf, fiber oriented with slit size 2 mm.

the fibre by the optical projector, V16-D Nikon (Japan) with assuming that the fibre was a cylindrical.

3. Calculation

It is well-known [\[9\]](#page-6-0) that Young's modulus of a short fibre reinforced composite is determined by

$$
E_{\text{comp}} = \eta_{\vartheta} \eta_{\text{f}} V_{\text{f}} \cdot E_{\text{f}} + (1 - V_{\text{f}}) \cdot E_{\text{m}}
$$
 (1)

where E_f , E_m , and V_f denote the Young's modulus of the fibre, the matrix, the volume fraction of the fibre in the composite. η_f , η_θ denote efficient factors of fibre length and orientation. η_f is given [\[10,11\]](#page-6-0) by

$$
\eta_{\rm f} = 1 - \left(\tanh\frac{1}{2}\beta L\right) / \frac{1}{2}\beta L\tag{2}
$$

$$
\beta = \left(\frac{2G_{\rm m}}{E_{\rm f}r_{\rm f}^2\ln(R/r_{\rm f})}\right)^{\frac{1}{2}}\tag{3}
$$

Eq. (2) means the Young's modulus of the composite decreases with decreasing the fibre length *l*, where r_f and R denotes the radius of the fibre and the interval among fibres. If the distribution of the fibres is homogeneous in an ideal packing square composite, R is given by

$$
R = \frac{r_{\rm f}}{2} \sqrt{\frac{\pi}{V_{\rm f}}} \tag{4}
$$

Shear modulus G_m with assuming that the composite is an isotropy given by

$$
G_{\rm m} = \frac{E_{\rm m}}{2(1 + v_{\rm m})} \tag{5}
$$

where r_f and v_m denote the radius of the fibre and Poisson's ratio of the matrix that assumed v_m is 0.3.

On the other hand, the efficiency factor η_{θ} has been analyzed by Fukuda and Chou [\[12\]](#page-6-0) using a probabilistic theory and assumed orientation distribution functions. The distribution functions are categorized by rectangular, sinusoidal and triangular distribution, respectively. Judging from the experimental result as shown later in [Fig. 12](#page-5-0), we assumed the triangular distribution as an orientation distribution function in the specimen shown in [Fig. 2](#page-1-0)b. This function is given by

$$
g(\theta) = \begin{cases} -2\theta/\alpha^2 + 2/\alpha & (0 \le \theta \le \alpha) \\ 0 & (\alpha < \theta) \end{cases}
$$
 (6)

where α denotes the limit angle of fibre orientation. If we experimentally determine the angle α , the orientation efficiency factor is given by

$$
\eta_{\vartheta} = 4 \cdot \frac{1 - \cos \alpha}{\alpha^2} \left(\frac{3 - \nu}{4} \cdot \frac{1 - \cos \alpha}{\alpha^2} + \frac{1 + \nu}{4} \cdot \frac{1 - \cos 3\alpha}{9\alpha^2} \right) \tag{7}
$$

Fig. 3. Relationship between orientation efficiency factor and limit angle.

Table 2 Parameters used for the flexural modulus prediction in the fibre oriented composites

	Random	Slit 7 mm	Slit 5 mm	Slit 2 mm
α	$\overline{}$	25	29	45
η_{θ}	0.27	0.61	0.75	0.82
η_1	0.91	0.91	0.91	0.91
$K_{\rm kf}$	1.35	1.35	1.35	1.35
$K_{\rm bf}$	1.05	1.05	1.05	1.05

This distribution function assumes that the fibre distribution is two-dimensional. Fig. 3 shows the relationship between η_{θ} and limit angel α . Efficiency factor η_{θ} is 1.0 when α equal to 0° (unidirectional fibre distribution), while η_θ is 0.47 at 90°.

The compression ratio is defined by the ratio of the original fibre volume to the fibre volume in the composite. Hence, the compression ratio, K , can be calculated by

$$
K = V'_{\rm f} / \left[V - \left(\frac{W - W_{\rm f}}{\rho_{\rm m}} \right) \right]
$$
 (8)

where V'_f , V , W , W_f and ρ_m denote original fibre volume, volume of the composite, weight of the composite, weight of the fibre and density of the matrix, respectively. Therefore, the final equation that predicts flexural modulus of the composite is

$$
E_{\text{comp}} = K \eta_{\vartheta} \eta_{\text{f}} V_{\text{f}} \cdot E_{\text{f}} + (1 - V_{\text{f}}) \cdot E_{\text{m}} \tag{9}
$$

Table 2 shows these parameters determined by the experiments mentioned above.

4. Results and discussion

4.1. The measurement of Young's modulus in kenaf and bamboo

[Fig. 4](#page-3-0) shows typical stress–strain curves for the bamboo and kenaf. The curve in the kenaf was clearly linear, while the curve in the bamboo was exponential and had some broken points, which were due to partial breakage of the

Fig. 4. Typical stress–strain curves of bamboo and kenaf in single fibre tensile test.

fibre with naked eye observations. In the case of bamboo, the initial slope was rather bland. In the initial stage, the bamboo was not tensiled uniformly due to the separated bundle structure shown in [Fig. 7b](#page-4-0). This is the reason that the final slope was selected to measure Young's modulus.

The fibre Young's modulus was determined to measure the inclination of the final linear part in the stress–strain

Fig. 5. SEM photographs of (a) bamboo and (b) kenaf after tensile test.

curve. Fig. 5a and b shows SEM micrographs of the kenaf and the bamboo after the tensile test. It was found that bundles of the bamboo were broken partially and broaden, while the kenaf was simply broken at the end of the fibre. [Fig. 6](#page-4-0)a–d shows the surfaces and the cross sections in the kenaf and the bamboo. The surface was observed before the tensile test, and the cross section was done after the test. The bamboo surface was obviously rough and there was the partial breakage in bundles. Moreover, the kenaf had clearly porous structure in the cross section, while the bamboo had solid-like structure.

[Fig. 7](#page-4-0) shows the relationship between fibre Young's modulus and fibre diameter. The Young's modulus in kenaf was rather higher than that in bamboo. The average fibre diameter, 0.10 mm in kenaf was smaller than that, 0.27 mm in bamboo. The average Young's modulus was 22,000 MPa (S.D. 6900 MPa) for kenaf and 18,500 MPa (S.D. 5300 MPa) for bamboo.

4.2. Effect of fibre volume fraction on flexural modulus

[Fig. 8](#page-4-0) shows the relationship between flexural modulus and fibre volume fraction in flexural specimens. The transformation of weight fraction into the fibre volume fraction was calculated by

$$
V_{\rm f} = \left[V - \left(\frac{W - W_{\rm f}}{\rho_{\rm m}} \right) \right] / V \tag{10}
$$

where W_f , ρ_m , V and V_f denote the fibre weight, the matrix density, the specimen volume and the fibre volume fraction, respectively. The flexural modulus in both kenaf and bamboo was almost same level despite the Young's modulus, 22,000 MPa for kenaf and 18,500 MPa for bamboo. The maximum flexural modulus was observed at 4800 MPa, 60% for kenaf and 5400 MPa, 72% for bamboo. The diameter in bamboo fibre was apparently larger, 0.27 mm than that in kenaf, 0.10 mm. Hence, the total surface area in bamboo was smaller than that in kenaf and this would enable the better wetting between bamboo fibre in less resin. However, in the above volume fractions, flexural modulus decreased. This is due to insufficient resin because it was found that many fibres on composite surfaces were not wetted by the resin sufficiently.

[Fig. 9](#page-5-0) shows relationship between compression ratio and fibre volume fraction. The compression ratio decreased with increasing fibre volume fraction. This is might be owing to that lower fibre volume fraction makes fibres moving into optimum positions. The compression ratio was 1.6–1.35 for kenaf and 1.1–1.0 for bamboo. These results are related to the cross section structures shown in [Fig. 6](#page-4-0)c and d. Hence, kenaf porous structure reflected the higher compression ratio. The decrease at 80% volume fraction may be attributed to void between fibres without resin.

With these compression ratios, the prediction lines of flexural modulus in kenaf and bamboo composites were calculated by Eq. [\(9\)](#page-2-0). As shown in [Fig. 8](#page-4-0), the solid line in kenaf prediction was in good agreement with the exper-

Fig. 6. SEM microphotographs of surfaces of (a) kenaf, (b) bamboo and cross sectional view, (c) kenaf, (d) bamboo after tensile test.

Fig. 7. Relationship between Young's modulus of fibre and its diameters.

Fig. 8. Relationship between flexural modulus in the composites and fiber volume fraction.

imental, while the broken line in bamboo was obviously lower than the experimental. It is considered that the Young's modulus in bamboo was estimated considerably lower due to the partial breakage during tensile test. If we fit the predicted line to the bamboo experimental result, the Young's modulus in bamboo would be 31,000 MPa. Hence, flexural modulus in kenaf and bamboo composites made no difference despite the fibre Young's modulus and the compression ratio in kenaf was higher than that in bamboo.

4.3. Effect of fibre orientation distribution on flexural modulus

[Fig. 10](#page-5-0) shows the fibre orientation distributions in kenaf composites by slit jigs. The orientation angle was defined as the deviation angle between the horizontal direction of

Fig. 9. Relationship between compression ratio and fibre volume fraction in the composites.

Fig. 10. Fibre orientation distribution of kenaf fiber in fibre oriented composites with various slit clearance.

the flexural specimen in [Fig. 2](#page-1-0)b and the longitudinal direction of the fibre on the composites surface. Thus, if the deviation angles of all fibres are zero degree, the specimen would be unidirectional. In each composite, 600 fibres randomly chosen were measured. Frequency on the y-axis in Fig. 10 means fibre existing possibility in every 10° range. The fibre orientation distributions were widened with increasing the slit clearance from 2 to 7 mm. In all composites, the fibre frequencies were highest in $0-10^{\circ}$, and decreased linearly with increasing fibre orientation angle. Hence, it was found that the fibre orientation distribution function in the slit jig composite was triangular as shown in [Fig. 3](#page-2-0).

Fig. 11 shows the relationship between flexural modulus in kenaf composites and slit clearances. All the fibre volume fraction was 60%. The black and the white plots represent experimental in kenaf and bamboo, while the solid and the broken lines do calculated. The flexural modulus increased considerably with decreasing slit clearance and reached 14,900 MPa for kenaf and 14,400 MPa for bamboo. These values were 3.1 and 2.7 times higher than those

Fig. 11. Relationship between flexural modulus in fibre oriented composites, bamboo and kenaf.

in kenaf and bamboo random composites. It was clear that the calculated flexural modulus in bamboo is not close to the experimental, while the flexural modulus in kenaf is close. This is same reason described in the previous paragraph referring to random bamboo fibre composites.

Fig. 12 shows the relationship between flexural modulus in kenaf composites and slit clearance in the case of the cross ply composites. This cross ply means that the fibre distribution in the lower half composites was orthogonal to that in the upper half layer. In the cross ply composites, the resin was put between the upper and lower fibre layer. The flexural modulus in the cross ply experimental was apparently lower than that in the calculated and even random composite. [Fig. 13](#page-6-0)a and b are the microphotographs of the fibre oriented kenaf composite with 2 mm slit and cross ply kenaf composite. The top and the bottom in the figures are surfaces, respectively. It was found that the resin was clearly segregated in the middle as indicated by the arrow. The reason is not cleat at this point, however, in the case of cross ply composites, the fibres did not mix each other due to the upper and lower layer not mixing each

Fig. 12. Relationship between flexural modulus in kenaf cross ply composites.

Fig. 13. Optical microphotographs of (a) kenaf fibre oriented composite and (b) kenaf cross ply composite in the cross-section.

other. On the other hand, the fibre in the fibre oriented composites mixed well and no segregation was found. This may be the reason why the flexural modulus was considerably lower than that in the calculated.

From these result, we can conclude Cox model that incorporates the effect of fibre compression can predict well in both for random and aligned fibre composites under the good fibre–matrix wetting condition.

5. Conclusions

Fibres Young's modulus were found to be 22,000 MPa for kenaf and 18,500 MPa for bamboo. In addition, kenaf was found to be more compressible than bamboo due to the porous structure. The compression ratios were 1.6–1.3 for kenaf and 1.1–1.0 for bamboo. However, the flexural modulus in bamboo composites was as same level as that in kenaf composites. This is because the Young's modulus in the bamboo was measured lower than actual modulus due to the partial breakage behavior during testing.

The flexural modulus increased with increasing fibre volume fraction up to 60% for kenaf and 72% for bamboo, and decreased above volume fraction. This decreased was owing to insufficient resin. The calculated flexural modulus by Cox's model that incorporates the effect of fibre compression were in good agreement with experimental results.

The flexural modulus in the fibre oriented composites made by slit jigs was 2.7–3.1 times higher than that in random composites. These were also well predicted by the Cox's model. However, the flexural modulus in cross ply composites was considerably lower than the predicted. The microstructure showed that the resin in cross ply composites was apparently segregated in the middle of the composites while the resin was homogeneous in the fibre oriented composite. This was due that the fibres did not mixed well each other in the cross ply composites. To predict flexural modulus, well and homogeneous wetting is needed between fibre and matrix.

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