TENSILE PROPERTY IMPROVEMENT BY CYCLIC LOADING TREATMENT FOR RAMIE/PP COMPOSITES

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1 Introduction

As a significant subject of the 21st century, construction of changing from the mass-production and mass-consumption society to the sustainable society is raised. In the field of composite materials, the difficulty in recycle for thermo-set resins and synthetic fiber reinforced plastics is pointed out. While these materials have superior mechanical properties, such properties make the disposal difficult. Therefore, most of these materials are ended by landfill disposal, but as easily known, the environmental impact increases due to their undegraded nature. From the above background, the research and development of biomass materials are much paid attention. Especially, plant-based natural fibers such as flax and ramie are expected as an alternative material of synthetic fibers. The natural fibers have excellent advantages, such as low cost, low density, high specific stiffness and their biodegradability. Moreover, it was reported that tensile strength and stiffness of natural fibers were improved by cyclic load application [1]. This is because cellulose microfibrils inside the fiber are rearranged along the direction of the fiber axis by cyclic loading. In this study, we aim to improve the tensile properties of a short fiber reinforced composite by cyclic tensile loading. First, ramie fibers and polypropylene were compounded and pelletized, and then injection-molded. Next, the composites obtained were tensile-tested through cyclic loading, and compared with the composite specimens without cyclic loading.

2 Experiments

2.1 Materials

Reinforcement used here was sliver of ramie fibers (TEIKOKU SEN-I Co., Ltd.). The fibers were preliminarily cut in 10mm length. In order to investigate the effect of the viscosity of matrix, two kind of polypropylenes, i.e. high polymer (PP-A; MFR=0.5) and low polymer (PP-B; MFR=13.0) were used. In addition, 2wt% maleic anhydride degeneration PP (MA-PP, Kayaku Akzo Co., Ltd.) was added into PP as a coupling agent to promote better surface interaction between the natural fibers and PP. Fiber content was 10wt%.

2.2 Injection molding

Ramie fibers were kneaded with PP-A or PP-B at 170^{o}C using a kneading machine (Labo-plastmill, Toyo Seiki Seisaku-syo Ltd.). In order to investigate the effect of kneading time on fiber length in composites, the kneading times were set to 7 and 10 minutes. After kneading, the obtained compound was pressed, and pelletized to the size of approximately 10x10x2mm^3. After that, the specimen was injection-molded into the die of a dumbbell shape using an injection molding (SHINKO SELLBIC Co., Ltd.). Shape and dimension of the specimen are shown in Fig.1.

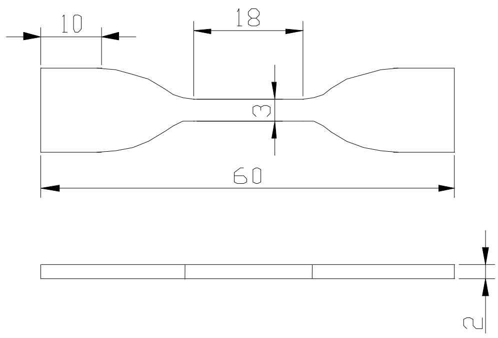

Fig.1 Shape and dimension of specimen by injection molding.

2.3 Tensile test and cyclic load treatment

Tensile tests and cyclic loading were conducted using a universal testing machine (Ritorusensuta small desktop tester, JT Tosi Co., Ltd.) at room temperature. The number of specimens was 5. Tensile speed was 10mm/min. The cross-sectional area was determined by measuring three locations along the longitudinal direction and then taking an average. Applied cyclic load was chosen as 70% level of fracture load. Cyclic loading times were twenty at 5mm/min.

2.4 Soxhlet extraction method

Fibers were shortened during kneading and injection-molding, and therefore the fiber length was investigated through Soxhlet extraction method. The fibers were observed by an optical microscope and measured in length. The fibers were extracted from one specimen, and the number of measured fiber lengths was 500.

3 Results and discussion

3.1 Tensile properties

Fig.2 shows the typical tensile stress-strain curves for 7 and 10 minutes of kneading times for ramie/PP-A composites. It was found that both curves look quite similar, but the average tensile strength for 7 minutes is improved 4.4% higher than that for 10 minutes in average strength. It was suggested that as the kneading time decreases, the fiber length increases.

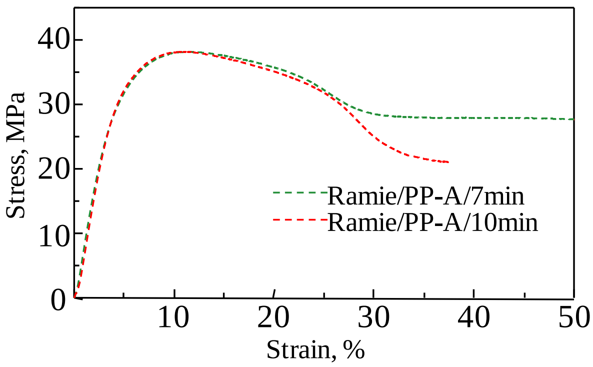

Fig.2 Stress-strain curves of PP-A for 7 and 10 minutes of kneading times.

Fig.3 shows the typical tensile stress-strain curves before and after cyclic loading for neat PP-A and ramie/PP-A composites for 10 minutes of kneading time. At first, to add the 10wt% ramie fibers into PPs, the Young's modulus of composites is improved 38% higher than that of neat resin. The tensile strength is not improved. Secondly, by applying the cyclic loading treatment, 8.5% in Young's modulus and 3.2% in tensile strength are improved.

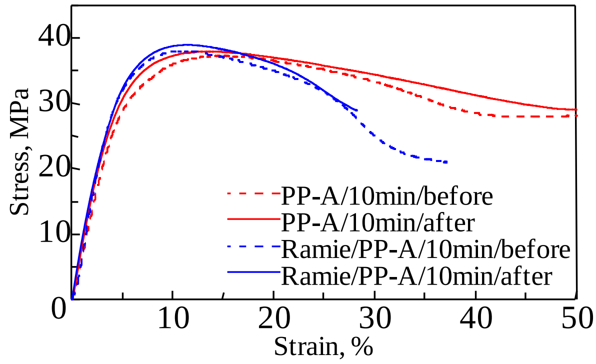

Fig.3 Stress-strain curves before and after cyclic loading for PP-A and ramie/PP-A composites.

Fig.4 shows the typical tensile stress-strain curves before and after cyclic loading for neat PP-B and ramie/PP-B composites for 10 minutes. By adding the 10wt% ramie fibers into polypropylenes, 19% in Young's modulus and 7.8% in tensile strength are improved. When applying the cyclic loading treatment, 16% in Young's modulus and 14% in tensile strength are furthermore improved.

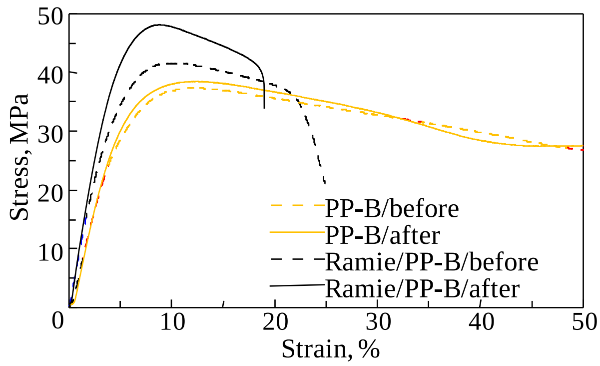

Fig.4 Stress-strain curves before and after cyclic loading for PP-B and ramie/PP-B composites.

3.2 Fiber length distributions

Fig.5 shows the extracted fiber length distributions from ramie/PP-A-10min, -7min and ramie/PP-B-10min composites. It was found that the fibers were extremely shortened through kneading and injection molding from the initial length of 10mm. Such decrease in length would be a reason why the strength was not improved so largely. In comparison between kneading time of 7 and 10 minutes, PP-A-10min decrease in average fiber length by 33%, as compared to PP-A-7min. On the other hand, 73% in average fiber length is improved to change the low viscosity PP (PP-B). Such condition against decrease in length would be a reason why the tensile properties was improved largely as shown in Fig. 4.

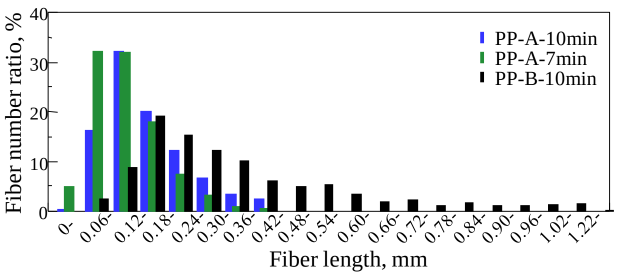

Fig.5 Fiber length distributions for PP-A-10min., -7min. and PP-B-10min.

3.3 Strength analysis of a ramie/PP composite

In this section, the change in tensile strength of a ramie/PP specimen by cyclic loading was predicted through a conventional mechanical model of a short fiber reinforced composite. At first, the fiber critical length, l_c, the least fiber length for exhibiting its strength in a matrix [2], was calculated as:

$$l_c = \frac{d}{2\tau_{my}} \sigma_f^*, \ \tau_{my} = \frac{\sigma_m}{\sqrt{3}} \tag{1}$$

Where, d is the fiber diameter, τ_{my} is the shear yield stress of the resin, σ_f* is the average strength of ramie fibers, and σ_m is the maximum stress of the resin. In this study, τ_{my} was estimated from σ_m based on the von Mises criterion. The above values are given as:

d=0.03mm, σ_m=37.3 and 38.7 MPa for the resin PPA and PPB, respectively and σ_f^*=610MPa tested at 10mm gage length [1]

In general, reinforcing fibers vary in strength because of size effect. As the average fiber strength in the above was obtained at 10mm length, a shorter length leads to larger strength for the fiber. To obtain an exact critical length, Weibull statistics is applied for its estimation as follows;

$$l_c = \frac{d}{2\tau_{my}} \sigma_f^* \left(\frac{l_c}{l_0}\right)^{-\frac{1}{m}} \tag{2}$$

Where, m is the shape parameter of Weibull distribution. In general, m of natural fibers varies from 2.5 to 5.0 [3-5]. In this study, therefore, m was given as 4.0. As a result, the obtained critical lengths for PP-A and PP-B were 0.82 and 0.77mm, respectively. Average fiber stress level σ_f at fiber failure is calculated as [6]:

$$\begin{aligned} \sigma_f &= \sigma_f^* \left(1 - \frac{l_c}{2l}\right) \quad (l_c \le l) \\ \sigma_f &= \sigma_f^* \frac{l}{2l_c} \qquad (l_c > l) \end{aligned} \tag{3}$$

Where, l is the fiber length. As l distributes as shown in Fig.4, its relative frequency has to be reflected on the eq.(3) [6]. That is:

$$\begin{aligned} \sigma_f &= f_1 \sigma_f^* \frac{l_1}{2l_c} + \cdots + f_{n'} \sigma_f^* \frac{l_{n'}}{2l_c} \\ &+ f_{n'+1} \sigma_f^* \left(1 - \frac{l_c}{2l_{n'+1}}\right) + \cdots + f_n \sigma_f^* \left(1 - \frac{l_c}{2l_n}\right) \end{aligned} \tag{4}$$

Where, f_1 to $f_{n'}$ are relative frequencies of the fiber lengths, l_1 to $l_{n'}$, less than l_c, and $f_{n'+1}$ to f_n are relative frequencies of the fiber lengths, $l_{n'+1}$ to l_n, more than l_c. In addition, the effect of fiber orientation has to be incorporated into eq. (4). In this study, the fiber orientation was defined as a normal line to a cross-sectional area, as shown in Fig 6. In the case of two-dimensional array, the fiber axial direction is oriented randomly in a plane A of Fig 6. That is, the orientation angle θ is distributed uniformly at $-\pi/2$ to $\pi/2$. In the case of three-dimensional array, the angle ϕ from a plane perpendicular to the two-dimensional plane has to be added. And the orientation angle ϕ is similarly distributed uniformly at $-\pi/2$ to $\pi/2$. Then the coefficients of fiber orientation, C_{2D} and C_{3D}, are respectively given for 2D and 3D arrays as:

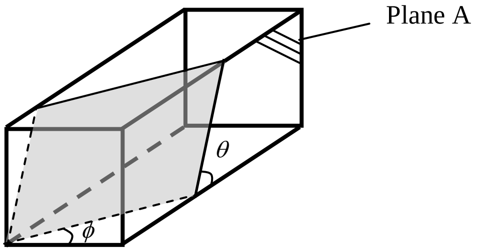

Fig.6 Schematic of fiber orientation angles θ and ϕ to the longitudinal direction

$$C_{2D} = \frac{1}{\pi^2}\int_{-\pi/2}^{\pi/2}\cos^2\theta d\theta \tag{5}$$

$$C_{3D} = \frac{1}{\pi^2}\int_{-\pi/2}^{\pi/2}\int_{-\pi/2}^{\pi/2}\frac{\cos^2\theta\cos^2\phi}{\sqrt{1-\sin^2\theta\sin^2\phi}}d\theta d\phi \tag{6}$$

Using these coefficients, the fiber stress level is rewritten as:

$$\sigma_f = C_{iD} f_1 \sigma_f{}^* \frac{l_1}{2l_c} + \cdots + C_{iD} f_n \sigma_f{}^* \left(1 - \frac{l_c}{2l_n}\right) \tag{7}$$

Where, C_{iD} is the coefficient of fiber orientation, and i is equal to 2 and 3 for 2D and 3D arrays. In the present study, C_{2D} and C_{3D} were calculated as 0.5 and 0.291, respectively. Tensile strength of the composite was then calculated by using the rule of mixture as follows:

$$\sigma_c = V_f \sigma_f + (1 - V_f)\sigma_m{}^* \tag{8}$$

Where, V_f is the fiber volume fraction, and σ_m* is the matrix stress level at the strain where the composite indicates the maximum stress. In this study, the fiber volume fraction was given as 9%.

Tensile strength of ramie fibers is known to increase with an increase in the number of cyclic loading [1]. According to the reference [1], the strengths increase up to 11% and 48% at five and twenty times, respectively, as compared to the original strength. Therefore, the fiber strength σ_f* was estimated as:

$$\sigma_f{}^* = \left(\frac{l_c}{l_0}\right)^{-\frac{1}{m}}(14.7N + 607) \tag{9}$$

Where, N is the number of cyclic loading. Table 1 shows change in tensile strength before and after cyclic loading for various ramie/PP composites. The analyzed strength increases by ~11% in the case of 2D array after cyclic loading, while the experimental value increases by ~14%. In Table 1, analyzed values using a coefficient of random fiber orientation of Fukuda & Chou [6] are also shown. The increasing rates of 3D array and Fukuda & Chou are given as ~8%. These increasing rates are less than that of the experiments. It is guessed from these analyses that the distribution of actual fiber orientation in present specimens provided from injection molding is similar to 2D array. More detailed analysis to explain such phenomena is our future subject.

Table 1 Tensile strength analysis using various coefficients of fiber orientation.

Composites Ramie-	Exp. [MPa]	Analyses, [MPa]		
		2D	3D	F & C
PPA/7/bef.	38.1	40.1	37.0	36.7
PPA/7/aft.	42.3	43.6	39.1	38.6
PPA/10/bef.	37.8	37.7	35.6	35.4
PPA/10/aft.	39.0	40.0	37.0	36.7
PPB/10/bef.	41.7	48.0	42.6	42.1
PPB/10/aft.	47.6	53.4	45.8	45.0

4. CONCLUSIONS

The present study tried to improve the mechanical properties of Ramie/PP green composites by cyclic tensile loading. The results obtained are as follows:

1) Ramie/PP-A increase in Young's modulus, while not improved in tensile strength. Tensile strength of ramie/PP-A kneaded for 7min was higher than that for 10 min. The cause is attributed to shortening of fiber length.

2) 19% in Young's modulus and 7.8% in tensile strength are improved for ramie/PP-B due to loading of 10wt% ramie fibers. Then, to apply the cyclic loading treatment, 16% in Young's modulus and 14% in tensile strength are improved. This is because the reinforcing fibers were improved in strength by cyclic load application.

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