ESTABLISHMENT OF CFRP RECYCLING METHOD PROCESSABLE UNDER ATMOSPHERIC PRESSURE

M. Sakuma[1]*, M. Koyama[2], H. Fukuda[2],
[1] Graduate Student, Tokyo University of Science, [2] Tokyo University of Science
* (sakusakuminoru@hotmail.co.jp)

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1 Introduction

Carbon fiber reinforced plastics (CFRPs) have high-specific strength and stiffness. Therefore, CFRPs are used in a wide range of industrial fields such as automotive and construction. However, recycle methods for CFRPs have not been established yet and this is a big problem from the viewpoint of environmental consciousness.

Recycling processes for thermoset composites can be classified into mechanical recycling and chemical recycling. In the mechanical recycling, CFRPs are crushed into small pieces and they are reused as, for example, a part of raw materials of cement. In the conventional chemical recycling, CFRPs are also crushed into small pieces and carbon fibers are recycled by pyrolysis processes or chemical treatment.

Thus, these methods have a shortcoming that the fiber length becomes short by the crush processing.

Then in the present study, a trial of collecting continuous fibers from used CFRPs is carried out where depolymerization of thermoset matrix is conducted under atmospheric pressure.

2 Materials and experimental procedure

2.1 Material

Carbon fiber used in the present study is Toray T-300B 12000. For the matrix, epoxy resin of Mitsubishi Chemical 801PN together with curing agent, LV11 was used. The bundles of fibers were arrayed unidirectionally by hand and vacuum assisted resin transfer molding (VaRTM) was used to fabricate CFRPs, although it was laboratory-level miniature equipment.

2.2 Decomposition of matrix resin

Benzyl alcohol and tripotassium phosphate were used for the solvent and catalyst, respectively. The CFRPs for decomposition were put in a glass vessel in which above solution was filled, and the vessel was covered with aluminum foil for loose seal. Then the glass vessel was put in an oil bath, for 14 hours at 190℃.

To examine the relation between the fiber orientation angle and CFRP tensile strength, we prepared two kinds of CFRP plates for dissolution. The one is that the CFRP was fixed with picture-frame shape aluminum jig to keep the fibers straight during dissolution and the other, not fixed.

After the matrix of CFRP specimen was completely decomposed, carbon fibers remained in the solution were collected and rinsed with acetone. Using these fibers, recycle CFRPs were again made by the above-mentioned VaRTM method.

Hereafter, recycle CFRP with fixture is named as RF-CFRP and recycle CFRP without fixture, as R-CFRP.

2.3 CFRP tensile test and monofilament tensile test

CFRP, R-CFRP, and RF-CFRP were prepared for the tensile test. The specimen configuration is shown in Fig. 1. The tensile tests were performed at a crosshead speed of 0.5 mm/min.

In addition, a monofilament tensile test was also conducted to examine the influence of the dissolution on the strength of the fiber. The dimension of the specimen is shown in Fig. 2. In this case, the gage length was 25mm and the crosshead speed was 0.1 mm/min.

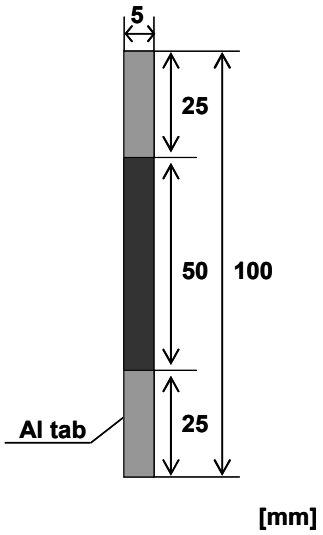


Fig. 1 Specimen configuration for CFRP tensile test.

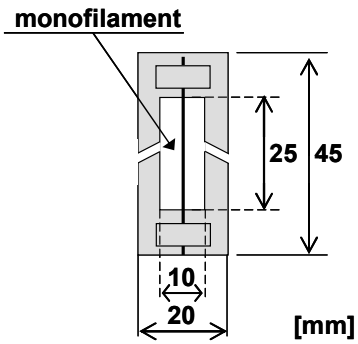


Fig. 2 Specimen configuration for monofilament tensile test.

2.4 Measurement of fiber volume fraction

The cross-section of each CFRP was observed with an optical microscope to calculate fiber volume fraction. The fiber volume fraction was calculated by line method [1].

2.5 Fiber orientation angle distribution

Because the fibers were arrayed by hand, the orientation of fibers was not necessarily unidirectional and this may cause the reduction of strength of CFRPs. Then the surface of each CFRP was observed with an optical microscope and the angle x between the direction of each fiber and tensile direction was measured. Normal distribution was assumed concerning the distribution of orientation angle x. The probability density function $f(x)$ and variance σ^2 are given by the following equations.

Here, $\bar{x}$ is an arithmetic average of measured fiber angle orientation.

$$f(x) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{x^2}{2\sigma^2}\right) \tag{1}$$

$$\sigma^2 = \frac{1}{n}\sum_{i=1}^{n}\left(x_i - \bar{x}\right)^2 \tag{2}$$

2.6 Prediction of tensile strength based on fiber orientation angle distribution

As was discussed in Section 2.5, some distribution of fiber orientation was observed. Then, we regarded the fabricated CFRPs as laminated plates of unidirectional laminate, where the thickness (fraction) of each lamina was proportional to the measured orientation angle distribution. We assumed successive failure of laminate, such as first-ply-failure, second-ply-failure, etc. Tsai-Hill criterion [2] for the ply strength was adopted here. This idea is also close to Coleman's dry bundle theory [3] and the maximum stress during the failure process was defined as predicted strength.

3 Results and discussion

3.1 CFRP tensile test

Figure 3 shows the result of CFRP tensile test. The decrease of strength by the recycling was recognized. It was also made clear that the rate of strength reduction of RF-CFRP was smaller than R-CFRP's. Strength reduction of recycled CFRP may be attributed to either the strength reduction of carbon monofilaments during matrix dissolution or linearity loss of the fiber bundles supplied for R-CFRP or RF-CFRP. Therefore, these factors were examined one by one in the following sections.

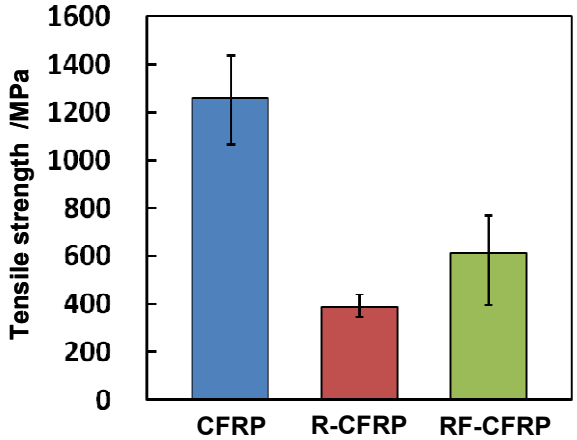


Fig. 3 Tensile strength of original and recycled CFRPs.

3.2 Monofilament tensile test

Figure 4 shows the result of monofilament tensile test. From this figure, it may be concluded that the strength of carbon monofilament itself is not affected by the dissolution process.

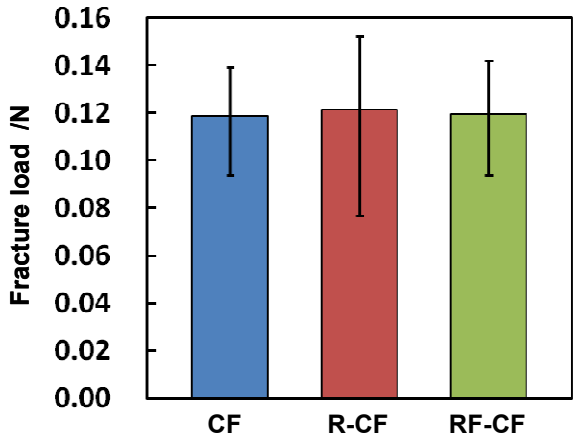


Fig. 4 Strength of monofilaments.

3.3 Fiber volume fraction

Figure 5 shows the fiber volume fraction of original and recycled CFRPs. The decrease of fiber volume fraction by the recycling was recognized. It was also made clear that the rate of fiber volume fraction reduction of RF-CFRP was smaller than R-CFRP's. Furthermore recycle CFRP's fiber volume fraction were had a large margin of error.

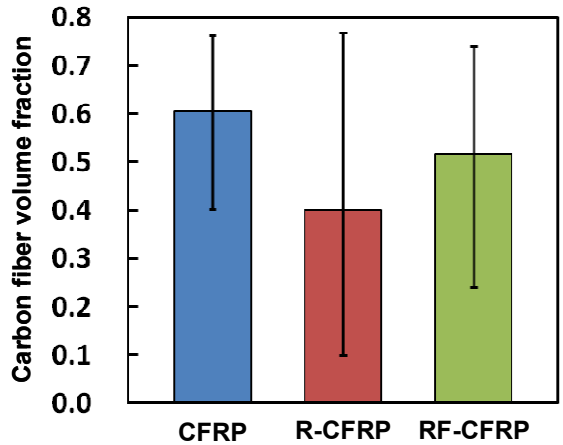
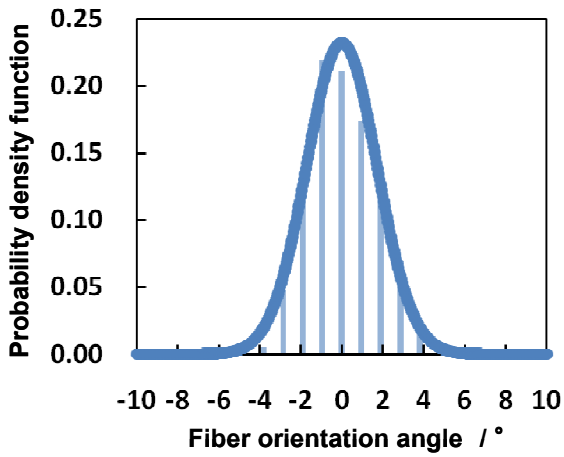


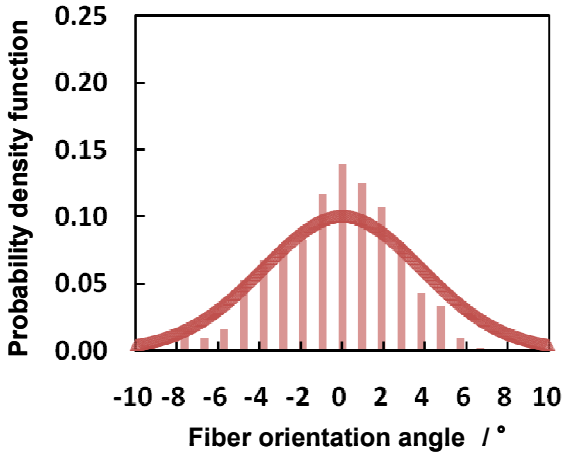
Fig. 5 Carbon fiber volume fraction.

3.4 Fiber orientation angle distribution

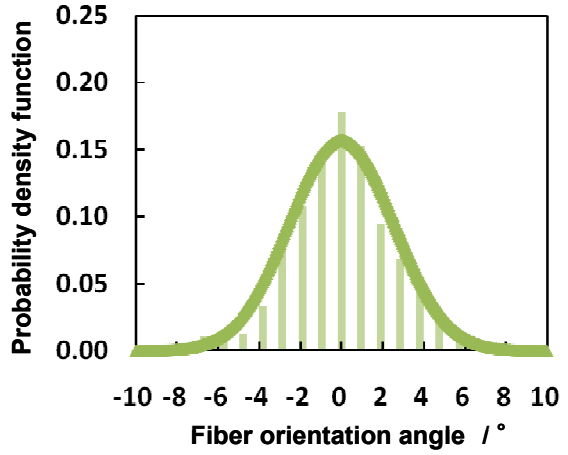
Figure 6 shows the fiber orientation angle distribution. By reproducing CFRPs using recycled fibers, the distribution became broader which suggests the reduction of tensile strength. Comparing with R-CFRPs, the orientation angle distribution of RF-CFRP is within narrower region, which again suggests the validity of Fig. 3. Therefore, it may be concluded that the main factor of the decrease of strength by recycling is due to the increase of scattering of the fiber orientation.



(a) CFRP



(b) R-CFRP



(c) RF-CFRP

Fig. 6 Fiber orientation angle distribution of three kinds of CFRPs.

3.5 Prediction of tensile strength based on fiber orientation angle distribution

Figure 7 shows the predicted and experimental tensile strength. The left bars of each group is theoretical tensile strength of unidirectional composite calculated with the rule of mixtures where the catalog value of fiber strength and the average volume fraction shown in Fig.5 are employed. The center striped bars are the predicted tensile strengths considering the fiber orientation angle distribution of Fig.6. In the case of original CFRPs, the predicted value is close to the experimental value. However, for R-CFRPs and RF-CFRPs, the experimental values are much smaller than predicted strength, although the tendency is reasonable.

The above result shows that large scatter of fiber orientation distribution during the recycling process is one reason of strength decrease. However, the results also suggests that other unknown factors have undesirable effects on the strength of recycled CFRPs; this point is left for future research.

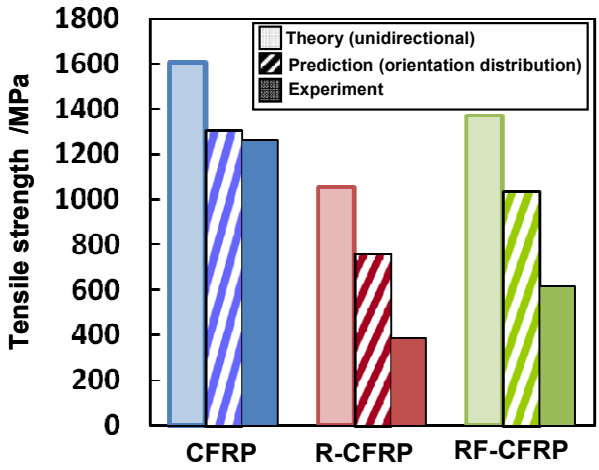


Fig. 7 Prediction of tensile strength based on fiber orientation angle distribution.

4 Conclusions

・The strength reduction of carbon fiber itself was not recognized by the dissolution treatment.

・Wide distribution of fiber orientation was observed for recycled CFRPs, which may lead the reduction of tensile strength.

・The strength of recycled CFRP can be improved by controlling the fiber orientation during recycling, although it was also recognized other unknown factors may also affect the strength.

References

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