

Design of Carbon Fiber Mat Reinforced Thermoplastics by Controlling Fiber/Matrix Adhesion and Volume Fraction of Fiber

Wataru Nagatsuka^{a*}, Noriyuki Hirano^b, Hidetaka Muramatsu^b,
Yoshiki Takebe^b, Atsuki Tsuchiya^b, Jun Takahashi^a

^aThe University of Tokyo

^bComposite Material Research Laboratories, Toray Industries, Inc.

*nagatsuka-wataru@cfrtp.t.u-tokyo.ac.jp

Keywords: Thermoplastic, Interfacial shear strength, Mechanical properties

Abstract

Polypropylene (PP) -used discontinuous carbon fiber reinforced thermoplastic (CFRTP) often exhibits insufficient mechanical properties, because PP is known to show poor adhesion to carbon fibers (CF). Additionally, a V_f limitation of discontinuous CFRTP also decrease the composite strength. In this study, the relationship among adhesion, V_f and mechanical properties in newly developed PP-used CFRTP sheet were evaluated. Interfacial shear strength (IFSS) between CF and PP can be quantitatively controlled by changing the modified PP content in matrix resin. At Higher IFSS, saturated flexural strength indicates that IFSS was no longer dominant the failure mode. The flexural strength was also saturated in higher V_f . It is probably due to increasing the contact points in the dense CF network that reduces the utilization of strength in carbon fiber. In this study, the relationship among adhesion, V_f and mechanical properties in newly developed PP-used CFRTP sheet were evaluated. At Higher IFSS or V_f , saturated flexural strength.

1. Introduction

The Carbon fiber reinforced plastics (CFRP) has high specific mechanical properties, so it is attracted attention as light-weight structure. Therefore CFRP is widely expanding application in structures of aircraft and cars in order to reduce CO₂ and environmental load. Specifically, reducing weight has been important mission for automotive industries and others [1]. In aerospace industries, CFRP is already accepted to the primary structure and bodies, and the effect is widely known. Some industries for mass-production need the CFRP that can be produced by high cycle process. But traditional CFRP using thermosetting resin matrix don't suit the general process because of low moldability and long curing time.

For their requirement, a new isotropic CFRP perform using discontinuous CFs (CMT: carbon fiber mat reinforced thermoplastics) has been developed by TORAY industries in Japanese METI-NEDO project (2008-2012) (Figure 1). CMT can be applied the Polypropylene (PP) for matrix resin, because it is lightweight, water-proof, chemical-resistance, and inexpensive. Owing to the controlled fiber length and individual fiber dispersion, this sheet shows excellent weight reduction effect [2-3].

However, in generally PP-used carbon fiber reinforced thermoplastic (CFRTP) often exhibits insufficient mechanical properties because homo or block-PP shows poor adhesion to CFs. So it is known that interfacial adhesion is improved by used acid-modified PP [4]. Additionally, the limitation in the volume fraction of fiber (V_f) of discontinuous CFRTP also prevent to increase the composite strength. And so, CMT appear the interesting mechanical properties.

In this study, the relationship among adhesion/ V_f and mechanical properties in newly CMT was evaluated. As a measurement of adhesion for the single-fiber composite (SFC) method for thermoplastic was developed by TORAY, which can be used to quantitatively discuss about the relationship between interfacial shear strength (IFSS) and mechanical properties of CFRP. So at first, the IFSS between CF and PP controlled by changing the modified PP content, and the mechanical properties of CMT were tried to clarify. And the next, the effect of V_f on the mechanical properties of CMT was investigated. From these result, optimal design of CMT is discussed.

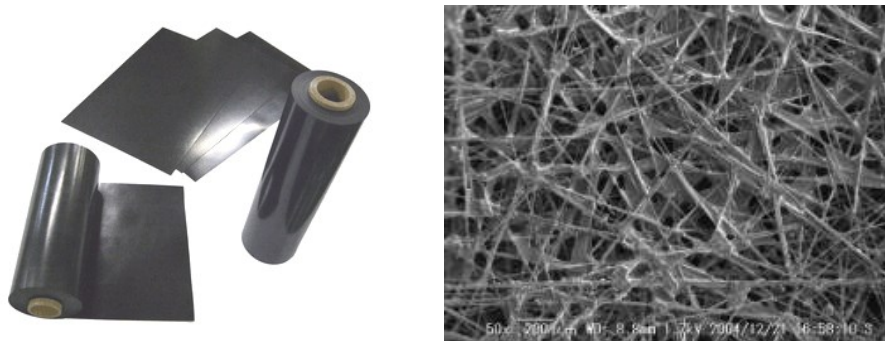


Figure 1. Image of left side is CMT sheet, and right is CF mat dispersed in isotropic.

2. Materials and Test Method

2.1. Material

CMT sheet was composed from CF mat made from discontinuous CF and PP as the matrix resin. CF mat was made by TORAY industries, Inc with T700 (tensile modulus 230 GPa; tensile strength; 4900 MPa). CF length can be designed and CF dispersion is isotropic in-plane. On the other hand, PP matrix was made from homo PP, and acid-modified PP and obtained by using a twin-screw extruder in melting-mixed. The content of homo PP and acid-modified PP was changed in order to control various IFSS. CMT were obtained by melt-pressing 200 g/m² of the CF mat and PP films. CF mats and PP films were laid up against each other and PP films were melt-pressured at 220 degree Celsius for 2 min under 4 MPa of pressure. The sheet was cooled under the same pressure until the material temperature reached 50 degree Celsius, the CFRP sheet was obtained. V_f was controlled by number of PP films.

2.2. Measurement of Interfacial Shear Strength and Mechanical Properties

The next, the IFSS between CF and PP was evaluated by the SFC method. IFSS test specimens were prepared as follows. A CF monofilament was embedded on the PP film used CMT sheets. In this time, pressing condition was at 220 degree Celsius for 2 min. Then they

were cooled to 50 degree Celsius and a CF embedded sheet was obtained. A dumbbell-shape specimen was cut from the sheet. After tensile test, the fragmented fiber length (L_f) was measured using a digital microscope. The average fiber length (L_{af}) was calculated from L_f , and the number of fragments (L_n) was calculated by equation (1). Then, the critical fiber length (L_c) and IFSS (τ) were calculated respectively by equations (2) and (3).

$$L_{af} = \frac{\sum L_f}{L_n} \quad (1)$$

$$L_c = \frac{4}{3} L_{af} \quad (2)$$

$$\tau = \frac{\sigma_f \cdot d}{2L_c} \quad (3)$$

Then three point flexural test was conducted according to ASTM D790 (crosshead speed: 1.4 mm/min; specimen width: 25 mm; length: 50 mm; thickness: 1.6 mm). The Izod impact test was conducted according to ASTM D256 Method A. A notched test piece was impacted in the edgewise direction. The fracture surface was observed by SEM.

3. Results and Discussion

3.1. Interfacial Shear Strength (IFSS) and PP Modification.

The relationship among IFSS, flexural strength of matrix resin and the acid-modified PP content were shown in figure 2. The IFSS becomes double by the addition of a small amount of acid-modified PP in 10%. However, only a gradual increase was obtained in case of 100% of acid-modified PP. It looks like IFSS is saturated over 10%. This results means that IFSS is related quantitatively by content of acid-modified PP. These data shows that the IFSS can be quantitatively controlled simply by changing the amount of acid-modified PP in the matrix resin, and then a little acid-modified PP can effect to interfacial adhesion at CF/matrix resin. On the other hand, flexural strength of only matrix is constant value though acid-modified PP content was increased. Adding the acid-modified PP in matrix resin has little effect to flexural strength of PP. It is expected that adding the acid-modified PP contribute to enhancing the only CF/PP interfacial adhesion, and matrix strength is remained.

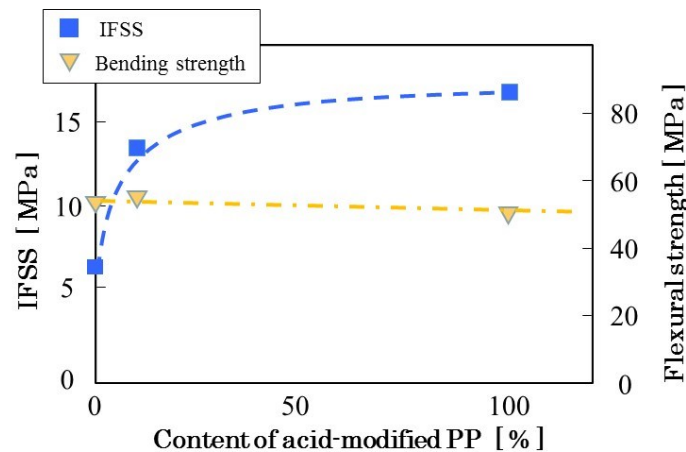


Figure 2. Relationship among interfacial shear strength/ flexural strength and content of acid-modified PP.

Table 1. Measurement result of IFSS and flexural strength of only matrix resin.

Content of acid PP [%]	IFSS [MPa]	Flexural strength [MPa]
0	6.21	57
10	13.31	58
100	16.60	53

3.2. Relationship between Matrix Resin Modification and Flexural Strength of CMT

The relationship between IFSS and flexural strength of CMT which V_f is 20% was shown in figure 3. This picture indicates that flexural strength was increased with increasing IFSS until adding 10% acid-modified PP. This result means that increased IFSS contribute to improve the composite strength. On the other hand, composite strength was saturated over the 10% added acid-modified PP. This result means that fracture mode, in high IFSS range, was changed to another fracture mode that is no longer related the IFSS. Specifically, fracture mode changed to fiber breakage over the 10% content of acid-modified PP from interfacial failure in lower content of acid-modified PP than 10%, nevertheless it is said PP matrix resin is poor adhesion. These results suggest that fracture mode of CMT can be transferred from interfacial failure to fiber breakage by design of matrix resin, even in case of CMT applied PP. CMT obtain the sufficient utilization of CF reinforced effect.

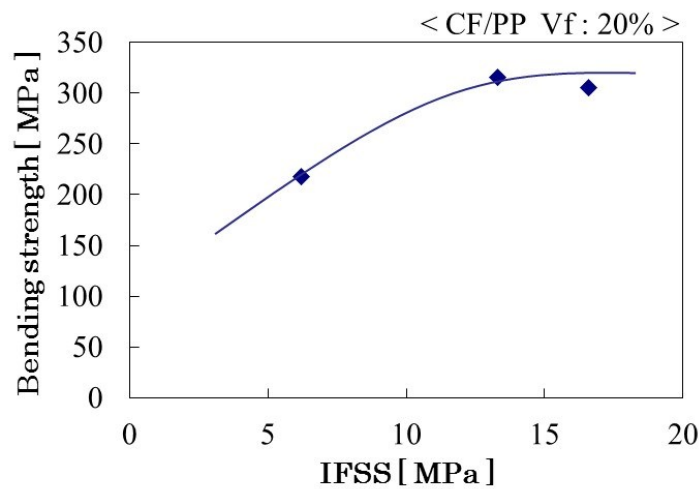


Figure 3. Relationship between flexural strength of CMT and IFSS.

3.3. Relationship between V_f and Mechanical Properties of CMT

The relationship between V_f and mechanical properties of CMT was examined. Figure 4 is shown the results that flexural strength and flexural modulus of CMT was measured at various V_f . Flexural modulus and flexural strength of CMT was improved with increasing V_f . Relationship between V_f and flexural modulus is explained by rule of mixture formula. But result of flexural strength indicated interesting data that it was improved with increasing until about 35 vol%, and it was saturated over about 35 vol%. Flexural strength is not exactly

follow the rule of mixture formula in higher V_f than about 35 vol%. In other words, these result reveal that it is difficult for CMT made from CF mat to improve the flexural strength over 35 vol% by controlling V_f .

It is assumed that saturation of flexural strength is caused by the filling shape of CF. The increasing V_f lead to augment the amount of contact points among each other CFs due to becoming the high dense structure of CF-network in mat. Image of relationship between V_f and number of contact points is illustrated in figure 5. Since stress transmission through the matrix resin is not enough possible at contact points, so contact points are apparent as failure section on fiber at high V_f mat. As a result, reasonable strength didn't appear in the range of high V_f . Additionally it was presumed that increasing the contact points each other CFs reduce the utilization of CF strength of in-plane direction. Because CF is bent to out-of-plane direction at contact points of each other CF. Thus added CF cannot contribute to improving the strength in-plane direction over high V_f range. For this reasons saturation of flexural strength is shown in figure 4.

Therefore, for effective design of mechanical properties with the isotropic dispersion mat, these results were suggested to need controlling the optimization range of V_f with purpose. It is suggested that optimum V_f is between from 20vol% to 30vol%.

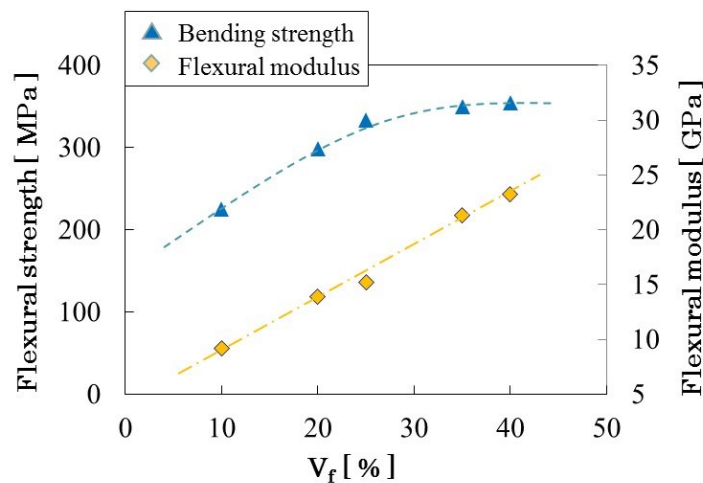


Figure 4. Comparison mechanical properties of CMT and various V_f .

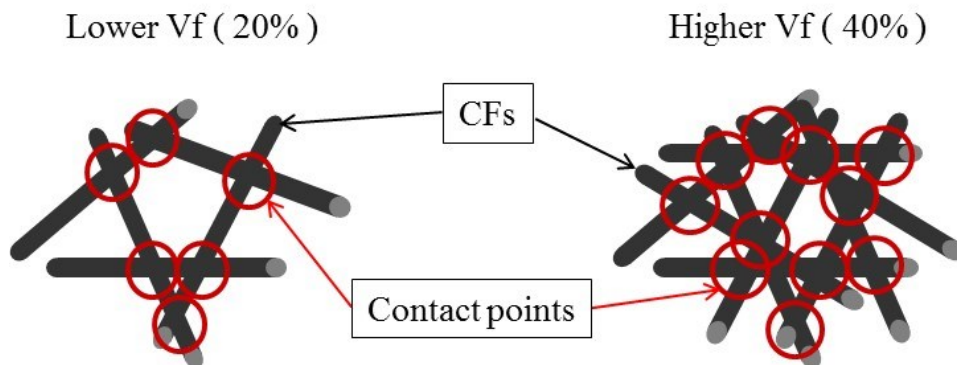


Figure 5. Image of contact points in CF mat.

4. Conclusion

In this study, the effect of resin formulation or V_f on mechanical properties of CFRTP with PP and discontinuous CF was examined in order to design the optimum CMT. The relationship among CF/PP interfacial adhesion or V_f and mechanical properties was illustrated with quantitatively about CMT. And following conclusions were obtained.

- Interfacial shear strength (IFSS) was controlled by changing the a little proportion of acid-modified PP in the matrix resin. As a result, even in case of applied the PP for matrix resin, achieving fracture mode with fiber breakage was revealed over the IFSS with added 10% acid-modified PP. CMT utilized the CF strength can be designed by consideration of IFSS.
- For this experiments appear that flexural modulus of CMT was improved by enhancing the volume fraction of CF. On the other it was found there was limit to the design to improve flexural strength by V_f . It was resumed that this result is due to filling shape of CF mat with higher V_f .

5. Acknowledgement

This work is a part of the Japanese METI-NEDO project “The development of sustainable hyper composite technology” (2008-2012fy).

Reference

- [1] K. Uzawa and J. Takahashi, Introduction of FRP composition materials (Chapter 5: Recent trend: recycling, nano-fibers reinforced composites, and new molding method - Recycling of FRP -), *Journal of Japan Society for Composite Material*, Vol. 34, pp. 245-250, 2008.
- [2] M. Honma, A. Tsuchiya, N. Hirano and T. Hashimoto, “Novel carbon-fiber reinforced stampable thermoplastic sheet with high strength”, *Proceedings of the 18th International Conference on Composite Materials*, 2011.
- [3] N. Hirano, A. Tsuchiya, T. Okabe, M. Hashimoto and M. Honma, “New stamping CFRTP sheet with excellent mechanical properties”, *Proceedings of the 12th Japan International SAMPE Symposium & Exhibition*, 2011.
- [4] H. Zushi, M. Tamaru, I. Ohsawa, K. Uzawa, J. Takahashi and H. Yasuda, “Evaluation on Mechanical Properties of Carbon Fiber Reinforced Polypropylene”, *Journal of Japan Society for Composite Material*, Vol. 32, pp. 153-162, 2006.