

## COOPERATIVE RELATIONSHIP BETWEEN INTERFACIAL AND IMPREGNATION STATE ON CONTINUOUS FIBER REINFORCEMENT THERMOPLASTIC COMPOSITES

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### Abstract

*The interfacial properties of continuous carbon fiber reinforced thermoplastic composites can be characterized by the wetting ability and chemical interaction between fiber and matrix. Wetting ability would affect the impregnation quality while chemical interaction between fiber and matrix affects composite strength. The objective of this study is to improve the both impregnation state and interfacial shear strength by using surface treatment on carbon fiber. For the purpose, in this paper, the effects of the sizing agents on the interfacial adhesion and impregnation state were investigated.*

### 1 Introduction

Continuous fiber reinforced thermoplastic composites have a number of advantages such as high fracture toughness, recyclability and possibility to re-melt and reprocess compared with thermo-setting composites. Therefore, the continuous carbon fiber reinforced thermoplastics (CFRTP) is very keen under global environmental issue in which natural resources are effectively used.

However, there are two problems for the CFRTP. The first one is that thermoplastics as matrices generally have high melt viscosity so that it is difficult to impregnate resin into reinforcing fiber bundle. To overcome this problem, CF/PP micro-braided yarn was fabricated as an intermediate material by Japanese traditional braiding technique. Micro-braided yarn is fabricated by braiding resin fibers alongside reinforcement fiber. Since resin fibers are located close to reinforcement fiber bundle, impregnation performance of thermoplastics is excellent [1].

The other one is low interfacial properties between the fiber and the matrix. It is considered that interfacial properties in continuous fiber reinforced thermoplastic composites can be characterized by the wetting ability and chemical interaction between fiber and matrix. Wetting ability would affect resin impregnation state during molding while chemical reaction affects composite strength. Therefore, interface design of CFRTP is very important to obtain materials with improved processability and mechanical performance.

The objective of this study is to improve the both impregnation state and interfacial shear strength by using surface treatment on carbon fiber. For the purpose, in this paper, the effects of the sizing agents on the interfacial adhesion and impregnation state were investigated. Sizing agents are a chemical coating agent applied on the surface of the CF. The general purpose of the sizing is to protect the fiber from fuzzing and fragmenting during textile processing into a woven or braided preform. Sizing agent also affects the fiber/matrix adhesion. Carbon fiber with different amount of sizing agent was prepared and the interfacial shear strength was examined. Moreover, maleated PP was used to improve the interfacial adhesion of the CF to the PP matrix. To evaluate the impregnation state and mechanical properties, the CF/PP or MAPP composites was fabricated.

## 2 Methodology

### 2.1 Materials

Table 1 shows the specification of carbon fiber used in this study. 4 types of carbon fiber with different amount of sizing agents (TORAY Industries Inc.) were prepared. As matrix resin, PP fiber and PP with maleated PP; MAPP fiber were used (Daiwabo Polytec Co., ltd).

Grade	Type of sizing agents	Amount of sizing agents (%)	Filament diameter ( $\mu\text{m}$ )	Tensile modulus (GPa)	Tensile strength (MPa)
T700SC-12000-50C	5	1.1	6.9	230	4900
T700SC-12000-H0C	H	1.0			
T700SC-12000-31E	3	0.6			
T700SC-12000-60E	6	0.2			

**Table 1.** Specification of carbon fiber used in this study.

To evaluate interfacial shear strength of the CF/PP interface, the micro-droplet test was employed. The resin fiber was melted by using a hot plate at 220°C and a small droplet of resin was applied to a single fiber. Micro-droplet test machine HM410 (Tohei Sangyo Co., Ltd ) was used with a fiber pull-out speed of 0.03 mm/min. When the micro-droplet touches the knife edges, the interface is solicited in shear mode. The maximum load  $F$  measured before matrix detachment from the fiber is related to the fiber/matrix shear strength. The interfacial shear strength ( $\tau$ ) was calculated by the following equation,

$$\tau = \frac{F}{\pi dl}$$

where  $F$  is the maximum load,  $d$  is the fiber circumference, and  $l$  is the embedded fiber length. The values of the fiber circumference and embedded length were characterized using microscope images.

### 2.2 Fabrication of unidirectional composites

CF as the reinforcement fibers was aligned and braided with PP fibers to yield CF/PP MBY by using the braiding technique. The fiber volume fraction ( $V_f$ ) of CF in MBY was adjusted to 40 %. The CF/PP MBY was wound 32 times onto a parallel metallic frame equipped with a spring mechanism to accept thermal shrinkage during molding, as shown in Fig. 1. The frame was then placed into a pre-heated mold before performing compression molding at 200°C with a molding pressure of 10 MPa. The molding time was varied at 5, 10, 20, and 40 min. The CF/PP composites were cut along the direction perpendicular to the fiber axis. The cross section of CF/PP composites was polished and observed by using an optical microscope. The regions that were un-impregnated with the resin could be determined from the cross-sectional

photographs. The image analysis software Photoshop CS2 (Adobe systems) was used to calculate the un-impregnation ratio. The un-impregnation ratio was calculated by normalizing the un-impregnated area by the fiber bundle area.

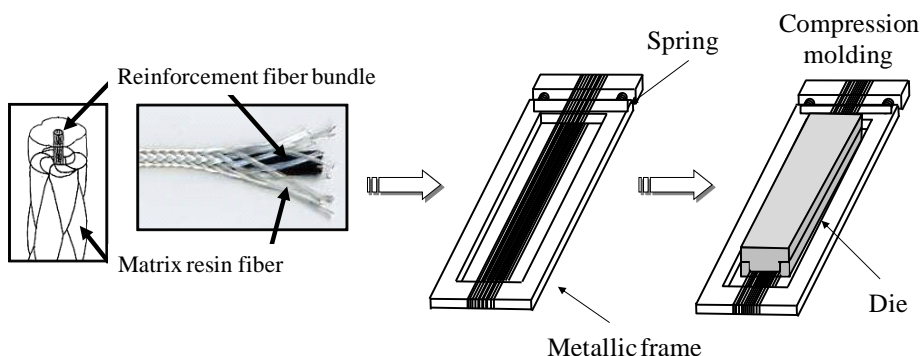


Figure 1. Fabrication method of unidirectional composites.

### 3 Results and discussion

Fig. 2 shows relationship between interfacial shear strength and sizing content. The interfacial shear strength was linearly decreased with increasing the sizing content as shown in Fig.2 (a). Sizing agents inhibited adhesion between CF and PP. Fig.2 (b) shows interfacial shear strength of CF/PA compared with CF/PP and CF/MAPP. By using MAPP, interfacial shear strength was greatly improved but CF/PA tended to have higher interfacial shear strength compared with CF/PP and CF/MAPP. However, in the case of CF/MAPP and CF/PA, interfacial shear strength was also decreased with increasing the sizing content.

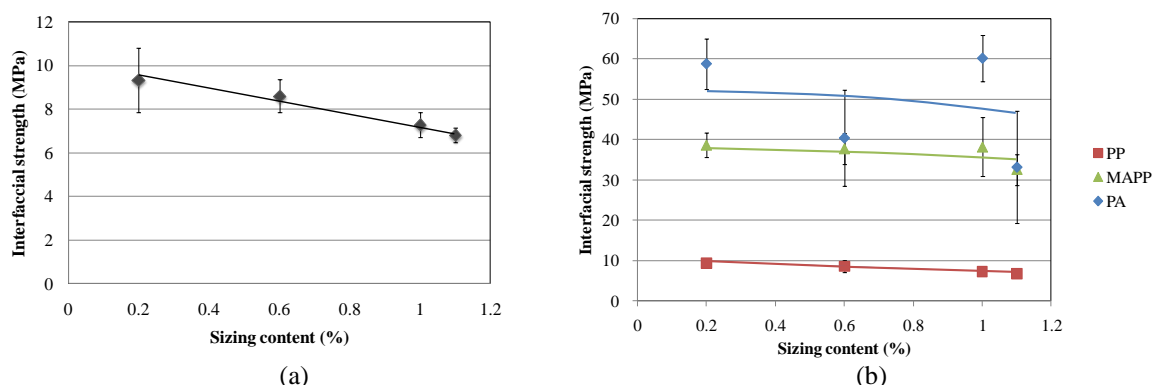


Figure 2. Relationship between interfacial shear strength and sizing content.

Fig. 3 shows the cross-sectional photographs of CF/PP for each molding time with different sizing content. In this figure, the clear circles are the CF bundles surrounded by grey matrix rich regions. Black regions, attributed to un-impregnated areas, were observed inside the fiber bundles. In the case of 1.1%, for the molding time of 5 min, a un-impregnation region in the fiber bundle was observed. The un-impregnation region was decreased with the increasing molding time. However, the un-impregnation region did not disappear after molding time of 40 min. In the case of 0.2 %, for the molding time of 5 min, un-impregnation regions in fiber bundle were observed but disappeared in the 10 min molding time. Therefore, the impregnation process was achieved in 10 min. Un-impregnation ratio was decreased with increasing sizing content and molding time. Therefore, it was clarified that sizing condition affected the impregnation process of CF/PP.

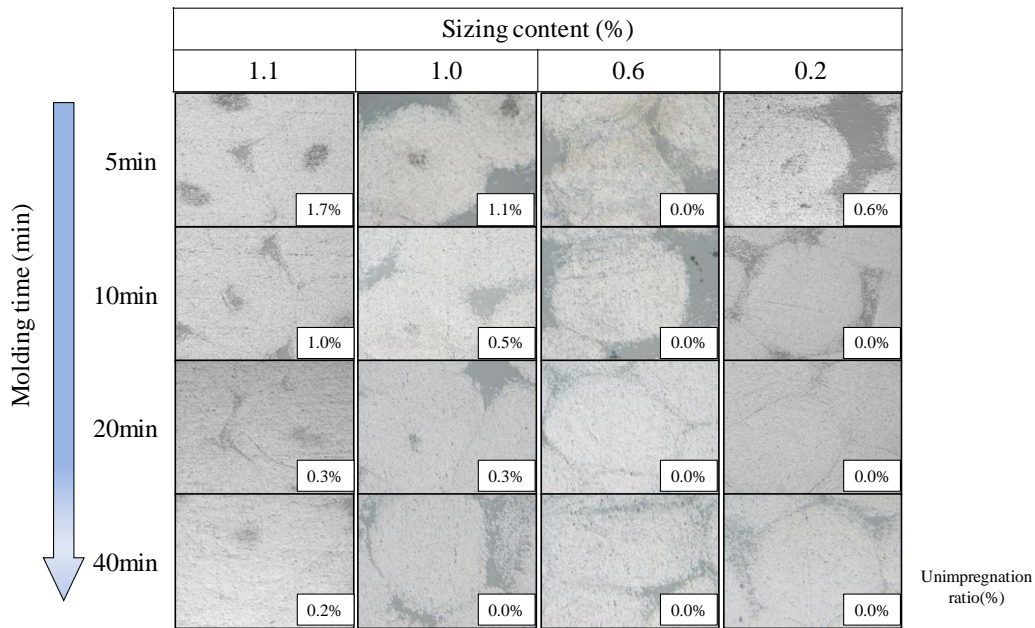


Figure 3. Cross-sectional photographs of CF/PP.

Next, effect of Maleated PP on the impregnation state was investigated. Fig. 4 shows the cross-sectional photographs of CF/PP and CF/MAPP for each molding time. In the case of CF/MAPP, larger un-impregnated regions were observed compared with CF/PP. Therefore, it was clarified that CF/MAPP had higher interfacial shear strength but matrix resin was not impregnated into the fiber bundles. Then, to clarify the wetting ability, contact angle was investigated as shown in Fig. 5. The contact angle of CF/MAPP was higher than that of CF/PP. It indicated that the wetting ability became lower by using maleated PP, so that CF/MAPP had poor impregnation state.

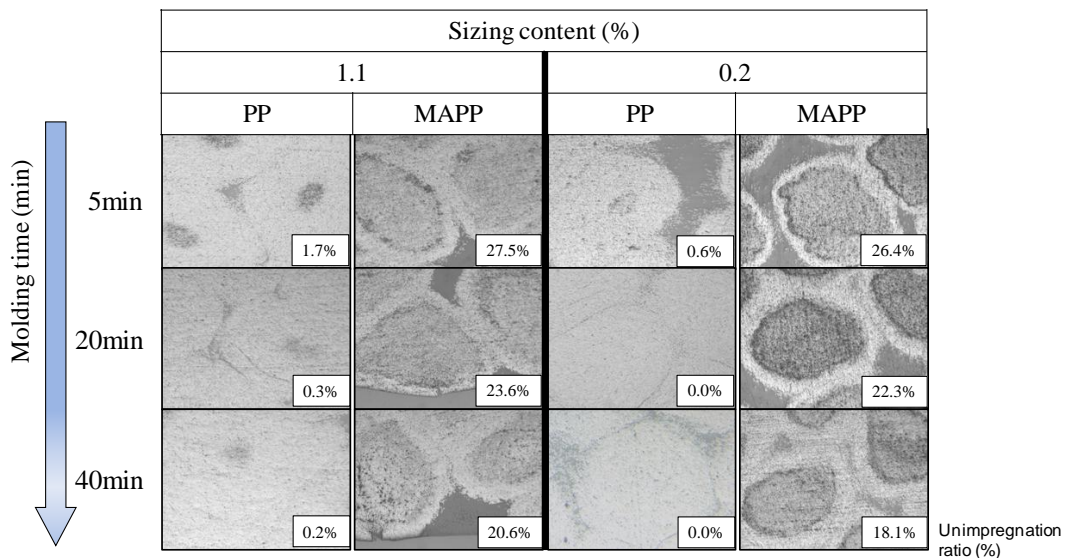


Figure 4. Cross-sectional photographs of CF/PP and CF/MAPP.

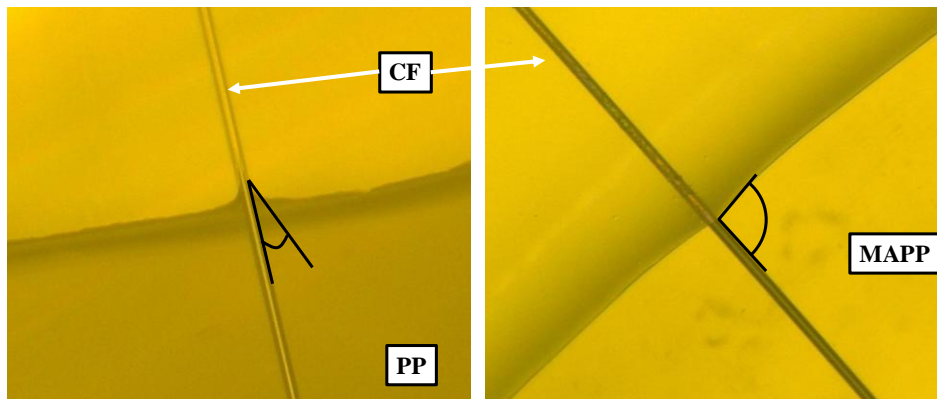


Figure 5. Photographs of contact angle.

#### 4 Conclusions

In this study, the effect of the sizing agents on the interfacial adhesion and impregnation state was investigated. Moreover, maleated PP was used to improve the interfacial adhesion of the CF to the PP matrix.

It was clarified that interfacial shear strength of CF/PP was decreased with increasing amount of sizing agents because sizing agents inhibited adhesion of CF/PP. Moreover unimpregnation region was increased with increasing amount of sizing agents. By using MAPP, interfacial adhesion was improved, but CF/MAPP had poor impregnation state compared with CF/PP because of poor wetting ability.

#### References

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