RESIN IMPREGNATION BEHAVIOR IN CONTINUOUS CARBON FIBER REINFORCED THERMOPLASTIC POLYIMIDE COMPOSITES

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Abstract

In molding of carbon fiber reinforced thermoplastics (CFRTP), resin impregnation behavior to fiber yarns is very important because higher viscosity of molten thermoplastics inhibits resin impregnation to the interspace among fibers. Resultant resin un-impregnation causes lower mechanical properties of CFRTP. The purpose of this study was to clarify the relation among molding method, molding conditions and resin impregnation to fiber bundles experimentally. In this study, CFRTPs using continuous carbon fiber yarn as a reinforcement and a thermoplastic polyimide which is excellent in heat resistance as a matrix resin were produced by Micro-Braiding, Film Stacking and Powder method. As a result, as the molding time increased, the impregnation ratio increased. The resin impregnation saturated in a certain molding time and the time was shorter the larger molding pressure. In addition, as the resin impregnation ratio increased, the mechanical properties also tended to increase.

1. Introduction

Since carbon fiber reinforced plastics (CFRP) has excellent characteristics of specific strength and specific stiffness, it has been used in many fields including aerospace. At present, the use of thermoplastic resin is expanding in CFRP. This is because it is expected that the production cost can be reduced due to short molding time and high recyclability. However, they have inferior characteristics such as lower resin impregnation to reinforcing fiber yarns. The reason is higher melt viscosity of thermoplastic resin. Thus resin-unimpregnated region, referred as void, exists in the fiber yarns after molding, results in lower mechanical properties.

Therefore, various molding methods to achieve sufficient thermoplastic resin impregnation to fabrics consisted of continuous fiber yarns have been studied for the practical usage of the composites in structural members. For examples, Film Stacking (FS) method [1, 2] is the method to hot-compression molding with alternately laminated woven fabrics and film-like polymer sheets. Powder method [3, 4] uses fiber yarns attached with pulverizing thermoplastic resin. A Commingled Yarn (CY) method [5] uses commingled yarn with fiberized thermoplastic resin and reinforcing fiber. In Micro-Braiding (MB) method [6-9], a braided fibrous intermediate material (Micro-Braided Yarn) composed of reinforcing fibers at the center and thermoplastics fibers around them are prepared with traditional braiding technique. The resin fibers are assembled around the reinforcing fibers and are evenly adhered. Thus improvement in impregnation property is expected. Many experimental researches have been conducted to improve the impregnation property of the thermoplastic resin to the reinforcing fiber bundles.

In the previous study, many researches on resin impregnation behavior in FS, CY and Powder method could be confirmed. On the other hand, there are some researches about MB method [6-9] for unidirectional composites, whereas the resin impregnation behavior for continuous woven materials has been limited [10-12] because of their complexity. However, it is important to clarify the resin impregnation behavior for continuous woven material with superior in drape property compared to unidirectional material, considering actual usage of the composites.

In this research, we investigated the effect of molding method and molding conditions on resin impregnation to fiber yarns in CFRTP. Carbon woven fabric and thermoplastic polyimide (PI), which is a super engineering plastics with superior heat resistance, were used. CFRTPs were prepared with MB, FS and Powder methods, and the resin impregnation properties were evaluated experimentally.

2. Experimental method

2.1. Materials

In this study, carbon fiber yarns, T300B-3000 filaments (3K, Toray) were used as the reinforcements, and a thermoplastic polyimide (PI, AURUM PL 450 C, Mitsui Chemicals) was used as the base material resin. Table 1 shows properties of PI. In FS and Powder methods, pre-woven fabric (CO6343B, Toray) consisted of T300B described above were used. In MB methods, MBYs were fabricated with PI fiber yarns on a medium-class braider and plain woven fabrics were weaved with MBY on a hand looms. The fabrics were cut into sheets of square, a 75 mm, which include 40 yarns for 3K.

PI films with thickness 50 μm were used in FS methods. For Powder methods, PI powder was prepared with PI pellets using a pulverizer (SM-1, Hisiangtai). PI resin powder with diameter 50 to 200 μm obtained with a test sieve (mesh opening 425 μm) was used. In the manufacturing process of MBY, a reinforcing fiber yarn was located at the center of the braider and matrix resin fiber yarns were braided around the reinforcing fiber yarn. In the present study, fiber volume fraction was 38.4% in all methods.

2.2. CFRTP fabric textile molding

CFRTP textile composites were compression-molded with a hot press system (IMC-1837, Imoto Machinery). Molding conditions are shown in Table 2. Fabrics and matrix resin, or MBY fabrics were placed in a mold at room temperature, and the mold was placed on the lower platen of a hot press machine preheated to 350 °C and heated to a molding temperature. When mold temperature reached the test temperature, pressure was applied to the mold. The time at the beginning of pressurizing was defined as 0 s of molding time. After maintaining the pressure for a certain period of the molding time, the pressure was relieved and the mold was air-cooled until 50 °C. The mold temperature and the pressure loaded on the specimen were defined as molding temperature and molding pressure.

Table 1. Properties of PI matrix.

AURUM-PL450C	
Viscosity [Pa·s]	600 (at 410 °C)
Melting Point [°C]	390
Density [g/cm³]	1.33
Tex for PI yarn [g/1000m]	60

Table 2. Molding condition.

Method	MB, FS, Powder
Molding Temperature [°C]	410
Molding Pressure [MPa]	2, 4
Molding Time [s]	0 ~ 300
Number of Layers	4
Fiber Volume Content [%]	38.4
Specimen Size [mm]	75×75

2.3. Cross-sectional observation

In order to measure the impregnation ratio as a function of molding conditions, cross sectional observation was performed at the center of the specimen molded under each condition. After the molded specimen was embedded in epoxy resin, the cross-section was polished using # 180 - 2000 emery papers, and the section was buffed with alumina slurry (0.3 μm, Maruto). The polished surface was observed using a digital-microscope (VH-Z100R, Keyence) having a zoom lens which enables to confirm a cross-section of a single carbon fiber. The digital image obtained was converted into a binary bitmap image using software (GIMP 2). The resin impregnation ratio was calculated as the ratio of the number of pixels in the impregnated region including the cross-sectional area of the fiber yarn to that of pixels in the whole cross section of the yarn. Since a moderate scatter was observed, the average value was shown as the result.

2.4. Tensile test

Tensile tests were performed for the prepared specimens in order to evaluate the influence of resin impregnation on mechanical properties. The dimensions of the specimen were 75 mm × 10 mm, and three pieces were cut out from near the center of the molded CFRTP textile. For testing, a universal testing machine (AUTOGRAPH AGS-X, Shimadzu) equipped with a 50 kN load cell was used and the crosshead speed was set to 1 mm / min. In order to measure the elastic modulus in each specimen, a strain gauge (KFG-5, Kyowa Electronic Instruments) was bonded at the center of the parallel portion. Three test specimens were prepared for each condition, and the tensile strength and the elastic modulus were averaged over the measured values. The test was conducted until the specimen broke.

3. Result and discussion

3.1. Effect of molding time and molding method (3K-4 plies-MB, FS and Powder)

Figures 1 and 2 show the effect of molding time on the resin impregnation ratio for each molding pressure (2, 4 MPa) and molding method. Molding pressure affects on the resin impregnation behavior; the promotion of resin impregnation caused by increasing flow rate of molten resin. The resin impregnation was promoted with increasing molding pressure, which result suggest that the molding pressure acted as a driving force for resin impregnation.

In addition, as the molding time increased, the impregnation ratio increased. Furthermore, under a condition exceeding a certain molding time, there was no remarkable change in the impregnation ratio, and it became a substantially constant value. For example, under the molding pressure of 2 MPa, the impregnation ratio became constant at molding time of approximately 120 s or more. This is because resin impregnation became saturated at a certain molding time. Also, under the molding pressure of 4

MPa, the saturation time of this impregnation became approximately 60 s, so the saturation time of impregnation was shown to be shorter the larger molding pressure. Additionally, since the aspect ratio of the carbon fiber bundle used in MBY was larger than that of the pre-woven fabric used in the FS and the Powder method, it is considered that the impregnation property was low in the MB method; the impregnation distance increased.

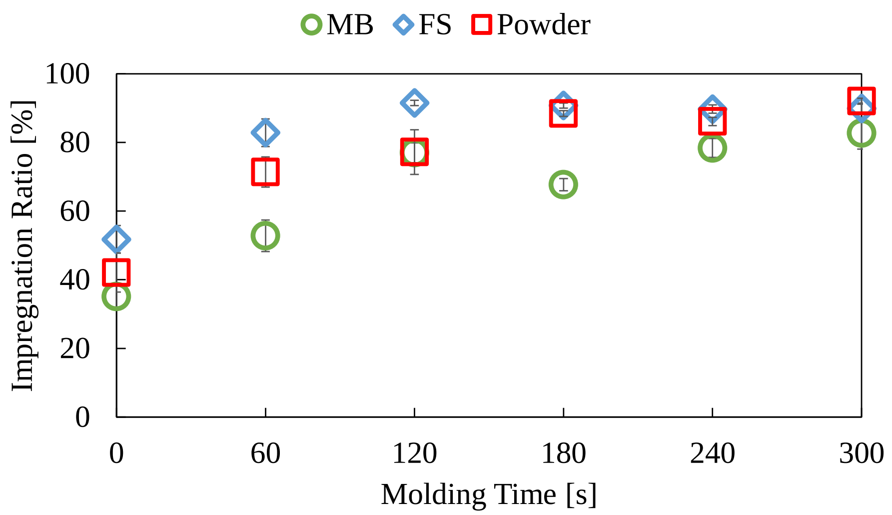

Figure 1. Effect of molding time on the resin impregnation behavior (Molding pressure: 2 MPa).

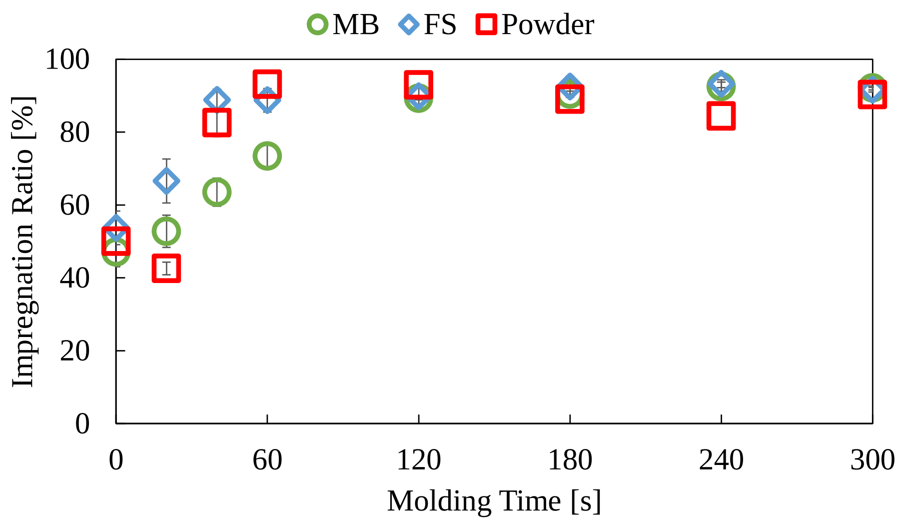

Figure 2. Effect of molding time on the resin impregnation behavior (Molding pressure: 4 MPa).

3.2 Relationship between resin impregnation ratio and mechanical properties

In order to clarify the effect of resin impregnation on mechanical properties, tensile tests were performed for the prepared specimens. Figures 3 and 4 show the relationship between elastic modulus, tensile strength and impregnation ratio of the specimens of each molding method. In the all molding method, both elastic modulus and tensile strength showed a tendency to increase as the impregnation ratio increased. However, the tensile strength of the MB method specimen was lower than that of other molding methods even at a high impregnation ratio. It is thought that this was caused by the influence of the waviness of carbon fiber bundle of occurring in MBY. This undulation occurs during the process of combining carbon fiber bundles and resin fiber bundles with a medium-class braider for manufacturing MBY (Fig. 5). In addition, the tensile strength of the FS method specimen was high. This is because the resin shape is a film in the FS method, so it is considered that the molten resin is uniformly dispersed entire pre-woven fabric as compared with other molding methods. Further, since the molding pressure becomes the driving force for impregnating the resin, it is an important factor of promoting resin impregnation.

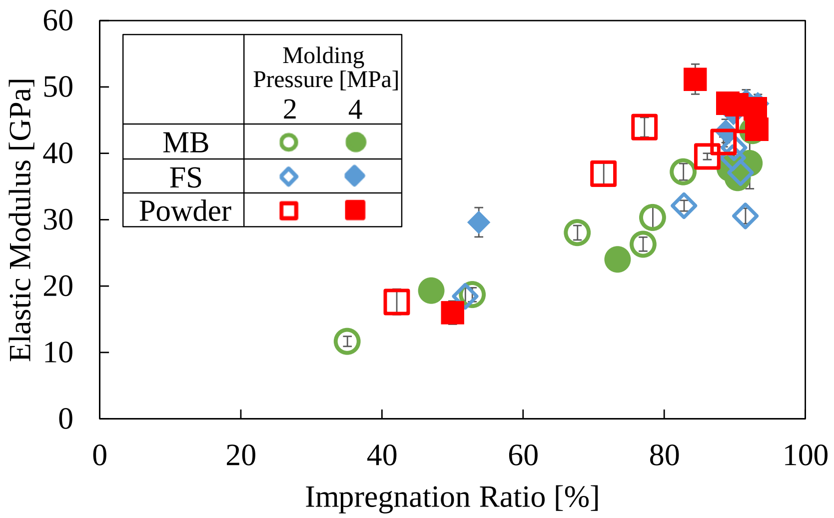

Figure 3. Elastic modulus as impregnation ratio (MB, FS, Powder - 2, 4 MPa).

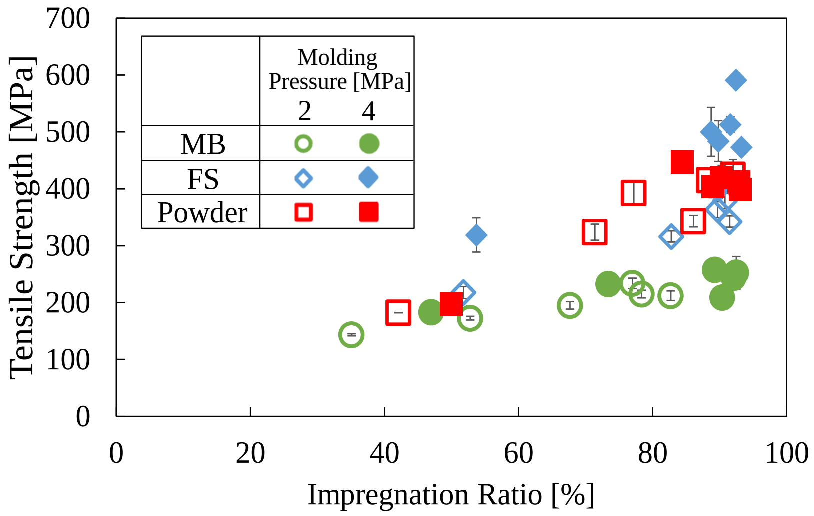

Figure 4. Tensile strength as impregnation ratio (MB, FS, Powder - 2, 4 MPa).

Figure 5. Waviness of carbon fiber bundle in MBY.

4. Conclusion

In this study, resin impregnation behavior for continuous carbon fiber reinforced polyimide composites was investigated experimentally. In order to investigate the effect of molding method on the resin impregnation to carbon fiber yarn, specimens were prepared by MB, FS and Powder method. The influence of the resin impregnation process on mechanical properties was also discussed. The conclusions obtained were as follows.

1. As the molding time increased, the impregnation ratio increased. The resin impregnation saturated in a certain molding time and the saturation time of impregnation was shown to be shorter the larger molding pressure.
2. The elastic modulus and the tensile strength showed a tendency to increase as the impregnation ratio increased in the MB, FS and Powder method.

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