

A STUDY ON THE PROCESSING ABILITY OF CARBON FIBER REINFORCED THERMOPLASTIC EPOXY RESIN COMPOSITES BY USING PULTRUSION TECHNIQUE

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Keywords: Pultrusion, Thermoplastic epoxy resin, Braided yarn, Carbon fiber, In-situ polymerization

ABSTRACT

In this research is to studies processing of carbon fiber reinforced thermoplastic epoxy resin composites by using pultrusion technology to produce the hollow shape composite. The optimum condition of thermoplastic epoxy resin temperature reaches, molding condition for pultrusion process and an effect of thermoplastic epoxy resin on the pultrusion processing of the composites was evaluated. The result of this study, the thermoplastic epoxy resin temperature reaches 105°C then decreased to 70°C after that added the accelerator to the resin and mixing, and the molding temperature at exit die for pultrusion process is 200°C are the optimum condition for this study. It could be improve the impregnation and mechanical properties of composites.

1 INTRODUCTION

Pultrusion is a processing to make highly aligned fiber reinforced polymer composites. Thermoset pultrusion has been well established and mainly used in the composites industry. Thermoplastic pultrusion has not been well established because thermoplastic resins are difficult to process due to their high viscosity and limited work. However, thermoplastic resins offer distinct advantages that make thermoplastic pultrusion worth exploring. [1]

Thermoplastic hold several important advantages over their thermoset counterparts for using in composites. Because of higher toughness, higher impact resistance, faster and more flexible manufacturing. Manufacturing cycle times, which include melting the resin, shaping, and consolidation by cooling are significantly shorter than thermoset counterparts. These require a time consuming curing step. As an alternative, reactive processing of textile fiber reinforced thermoplastics is a very attractive theme: low viscosity mono- or oligomeric precursors are used to impregnate the fibers, followed by in situ polymerization. In addition, Thermoplastic composites can be welded and recycled [2].

Braided yarn (BY) has been proposed as a means of overcoming the difficulty of impregnation of thermoplastic resin into fiber bundle in manufacturing long fiber reinforced thermoplastic composites [3]. BY 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 [4].

In our previous research, it has been clarified that the CF/thermoplastic resin fiber composites had not been good impregnation and surface for pultrusion process. The main problem during the processing of thermoplastic resin composites is the impregnation of the fiber reinforcement with highly viscous resin. The objective of this study is to improve the both impregnation state and surface by using the low viscosity of thermoplastic resin. To achieve this objective, thermoplastic epoxy resin were used instead of thermoplastic resin fiber.

This research is to studies processing ability of carbon fiber reinforced thermoplastic epoxy resin composites by using pultrusion technique to produce the hollow shape composite. The optimum condition and an effect of thermoplastic epoxy resin on the pultrusion processing of the composites was evaluated.

2 MATERIALS AND METHODS

The materials used in this research were carbon fiber as the reinforcement (T700SC-12000, Toray Co., Ltd.) and thermoplastic epoxy resin (XNR6850A) and the accelerator (XNH6850B) as matrix resin. Thermoplastic and accelerator was supplied by Nagase Chemtex Corp. The intrinsic properties of Thermoplastic epoxy resin and accelerator (the values given in product information supplied by the company) are shown in Table 1.

		DENATITE XNR6850A	ACCELERATOR XNH6850B
Chemical classification		Formulated epoxy resin	Aromatic phosphoric acid ester
Aspect		White paste	White powder
Viscosity	at 25°C	220 Pa·s	Solid
Specific Gravity	at 25°C	1.17	1.10

Table 1 Thermoplastic epoxy resin and Accelerator data

• In-situ Polymerization Process

- 1) The thermoplastic epoxy resin part, 'XNR6850A', is heated with stirring by using magnetic stirring hot plate which is set to 170°C.
- 2) The inside temperature of the resin reaches 105°C, or 105°C decreased to 70°C, or 70°C respectively of reaches temperature of resin. The resin will become clear and homogeneous.
- 3) After that, 'XNH6850B', is added to the resin part quickly with stirring.
- 4) While heating by hot plate is stopped, stirring is continued until the resin mixture become completely homogeneous.

In this process will various the temperature reaches of resin and the gelation time of each temperature of thermoplastic epoxy resin was evaluated.

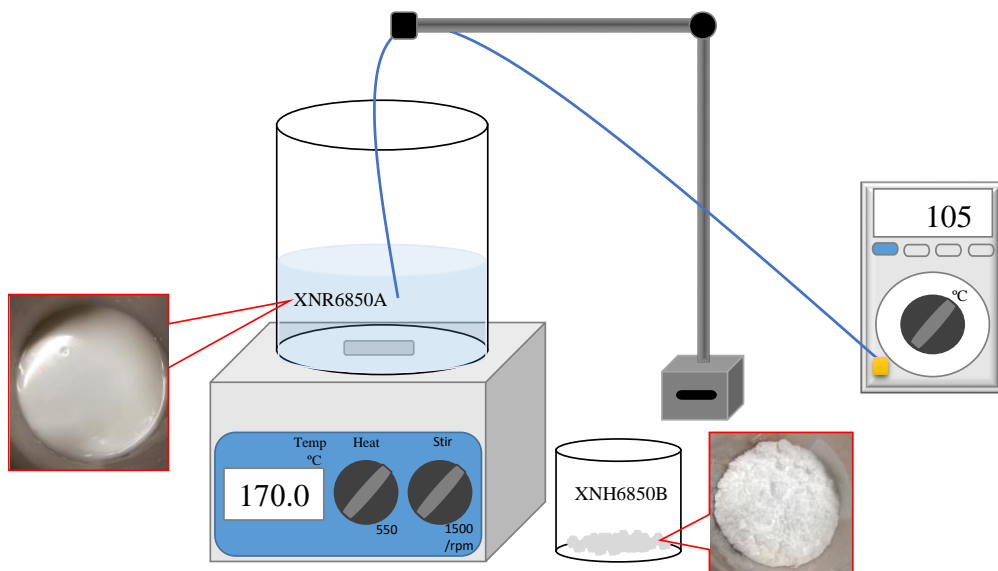


Figure 1 In-situ Polymerization Process

No.	Temperature reaches of resin before mixing (°C)
1	105
2	105»70
3	70

Table 2 The various temperature reaches of resin before mixing with the accelerator (XNH6850B, the particles are dissolved completely at resin temperature 105°C)

- **Forming the CF/Thermoplastic epoxy resin composites into square plate**

For CF/Thermoplastic epoxy resin composites, 4 ply of CF fabric plain pattern was put in the square box. The put the square box on the hot plate which is set the temperature to 200°C. After that pore the completely homogeneous of thermoplastics resin into a square box and wait until the polymerization is complete. The impregnation and mechanical properties was evaluated.

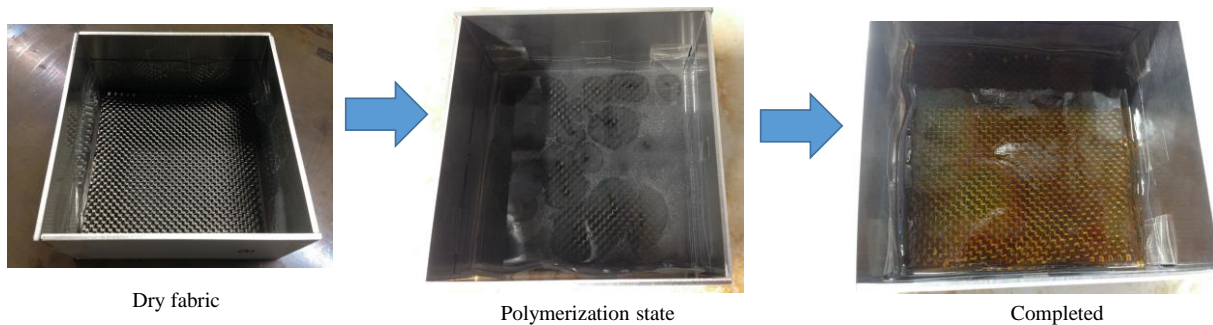


Figure 2 CF/Thermoplastic epoxy resin composites into square plate process

- **Braiding and Pultrusion process**

The fabrication of tubular CF/Thermoplastic epoxy resin braided composite by presented pultrusion molding. The braided yarn technique manufactured preform which had CF diagonally oriented at certain angles with the CF inserted into the braiding yarns along the longitudinal direction as shown in Figure 3 by using 16 braiding yarns with 50° of braiding angle and 8 middle end yarns on mandrel diameter 15mm for 3 layers with ideal volume fraction 59wt%.

Pultrusion system is one of continuous molding method. The CF braided fabric was inserted into the pultrusion machine. The molding die and each designation of the heating device are shown in Figure 4. Molding die had a tapered part at the entrance side of die. Diameter at entrance side was 20mm and diameter at exit side was 18mm. The total length of the molding die was 270mm. Molding temperature could be controlled by heating device at the eight zones. In this study, heating temperature at zone number eight (exit die) was 220°C, 200 °C and 160°C, respectively. When CF braided yarn was passed through the resin bath and heater at zone number two, CF braided yarn was subjected to heat and polymerization stating. The temperature of resin bath and entrance die (heater zone number one) was set at the room temperature because during the pultrusion process, the resin temperature in the resin bath should be between 70-80°C. Otherwise, it will polymerization and change to solid phase. The pulling speed was 16 Hz (60mm/min). With this molding die, the tubular composite with diameter of 18mm was molded. The impregnation quality was evaluated by using an optical microscope observation of the CF/Thermoplastic composites cross-sections. The processing ability was evaluated by visual check.

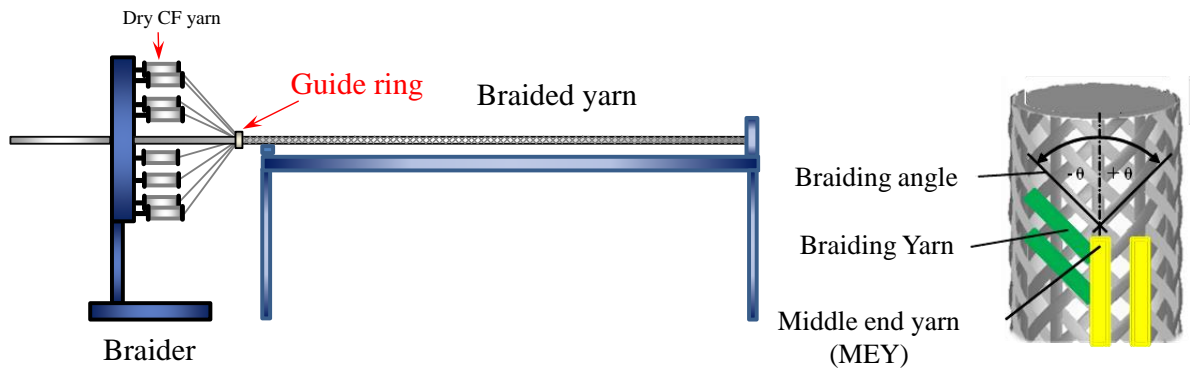
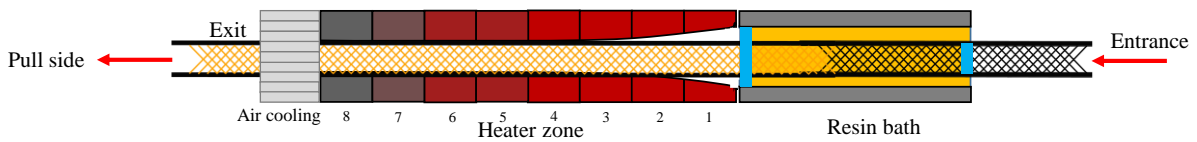


Figure 3 Schematic diagram braiding process and braided fabric



	Zone 8	Zone 7	Zone 6	Zone 5	Zone 4	Zone 3	Zone 2	Zone 1	Resin bath	Mandrel
1	220°C	170°C	150°C	130°C	110°C	90°C	70°C	RT	RT	RT
2	200°C	170°C	150°C	130°C	110°C	90°C	70°C	RT	RT	RT
3	160°C	150°C	130°C	110°C	90°C	80°C	70°C	RT	RT	RT

Figure 4 Schematic diagram Pultrusion process

3 TESTING METHOD

To observe the impregnation state of the composite, the cross-section of composites was embedded in epoxy resin and then polished in order to observe the fiber bundle impregnation by inverted microscope Olympus-GX41F.

For the mechanical property, 3-point bending test of composites was performed by using Tensilon RTC-1350A 5kN. The specimen size was 100mm in length, 15mm in width and 1.8~2.0 mm in thickness. The span length was 80 mm and the test speed was 5mm/min.

4 RESULTS AND DISCUSSION

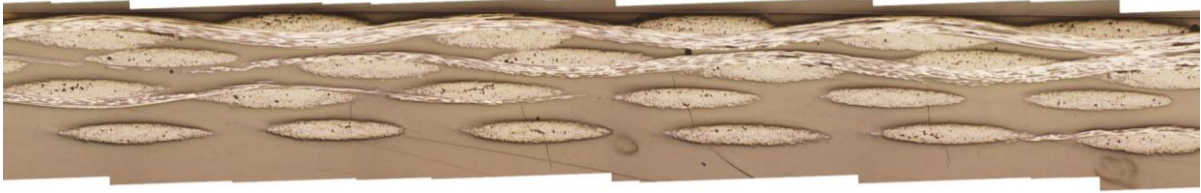
Temperature reaches of resin before mixing (°C)	Gelation time (min)
105	~1
105»70	~20
70	~25

Table 3 Gelation time of thermoplastic epoxy resin

In Table 3 shows the gelation time of thermoplastic epoxy resin after added the accelerator (XNH6850B). The temperature reaches of thermoplastic epoxy resin for pultrusion process, it was found that the temperature reaches 105»70°C and 70°C showed better gelation time than the temperature reaches 105°C. The resin temperature reaches 105°C, the gelation time is around 1 min

because the particles of accelerator (XNH6850B) are dissolved completely at resin temperature 105°C so the polymerization is occur very fast.

Temperature reaches: 105°C, Un-impregnated area: 1.71%



Temperature reaches: 105»70°C, Un-impregnated area: 0.95%



Temperature reaches: 70°C, Un-impregnated area: 0.33%

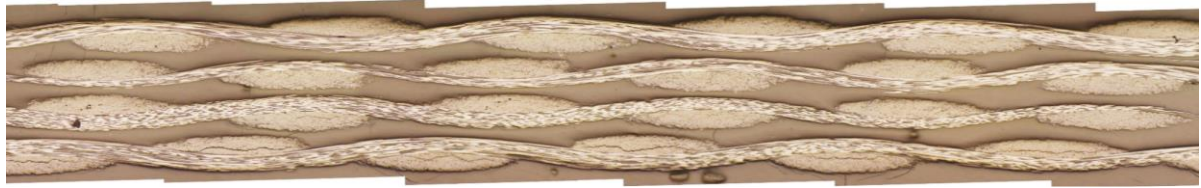


Figure 5 Cross section observation of CF/ Thermoplastic epoxy resin composites

In Figure 5 shows the cross section observation of CF/ Thermoplastic epoxy resin composites with various the temperature reaches of resin. The un-impregnation area of the resin temperature reaches 105»70°C and 70°C was lower than 105°C because the gelation time of the resin temperature reaches 105»70°C and 70°C longer and lower viscosity than resin temperature reaches 70°C.

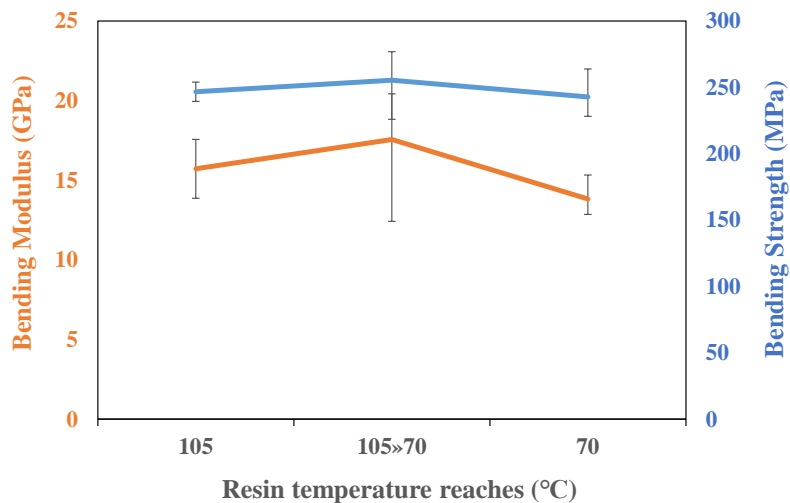


Figure 6 The bending properties of composites

In Figure 6 shows the bending properties of CF/ Thermoplastic epoxy resin composites. At the resin temperature reaches 105»70°C could produce composites with better mechanical properties by increased the bending modulus and strength of composites. In the case of specimen 70°C, the mechanical properties was lower than 105°C and 105»70°C because the specimen 70°C was brittle

than specimen 105°C and 105»70°C.

Based on the cross section observation and bending properties results of CF/ Thermoplastic epoxy resin composites, we decided to use the resin temperature reaches 105»70°C for the pultrusion process.

① Exit die temp (Z8): **220°C**



② Exit die temp (Z8): **200°C**



③ Exit die temp (Z8): **160°C**



Figure 7 Visual check surface of composites

In Figure 7 shows the visual check surface of CF/ Thermoplastic epoxy resin composites with various the exit die temperature by using pultrusion technique. The CF fabrics can be extruded from mandrel and mold. The surface of specimen exit die temperature 220°C and 200°C could be observed the smooth and good surface than exit die temperature 160°C. The surface of specimen exit die temperature 160°C could be observed rich resins.

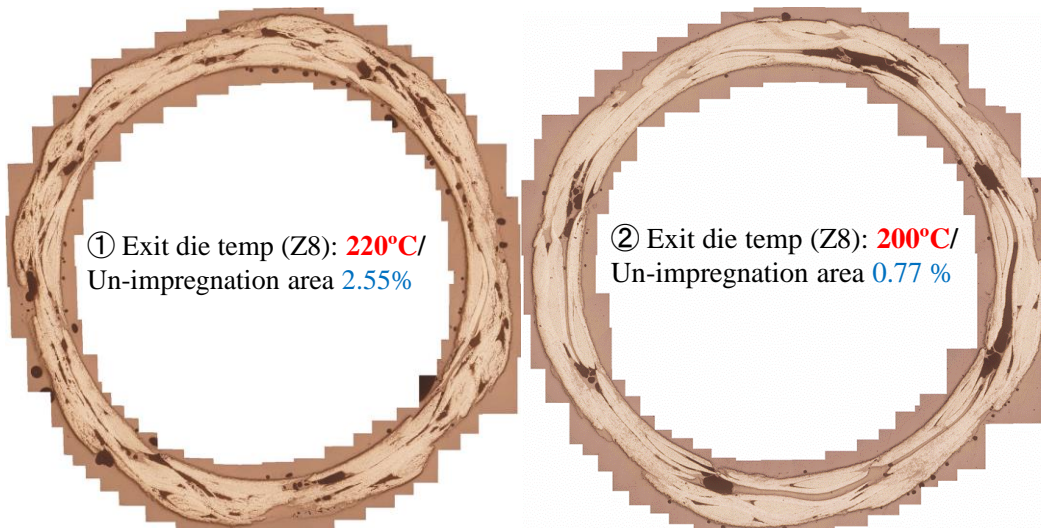




Figure 8 Cross section observation of CF/Thermoplastic epoxy resin composites

In Figure 8 shows the cross section observation of CF/Thermoplastic epoxy resin composites with various the exit die temperature by using pultrusion technique. The un-impregnation area of exit die temperature 200°C was lower than of exit die temperature 220°C and 160°C. It indicated that the molding temperature at the exit die 200°C could be improve the impregnation of resin into CF bundle better than the molding temperature at the exit die 220°C and 160°C.

5 CONCLUSIONS

Based on the above results,

The optimum condition of the resin temperature reaches for pultrusion process which good gelation time and mechanical properties is 105 °C and decreased to 70°C. While, the optimum molding condition for the good impregnation of thermoplastic epoxy resin into carbon fiber bundle is condition No.2 (Z8: 200°C, Z7: 170°C, Z6: 150°C, Z5: 130°C, Z4: 110°C, Z3: 90°C, Z2: 70°C and Z1: 0°C). During the pultrusion process, the best resin temperature in the resin bath should be between 70-80°C. Otherwise, it will polymerization or change to solid phase.

ACKNOWLEDGEMENTS

This work is a part of the Center of Innovation Science and Technology based Radical Innovation and Entrepreneurship Program (COI STREAM) supported by Regional Innovation Strategy Support Program (The Ministry of Education, Culture, Sports, Science and Technology (MEXT)). The authors would like to acknowledge the support of the Thermoplastic epoxy resin and knowledge by Nagase ChemteX Corporation in Japan. The authors would like to thank Dr.Hirofumi Nishida from Innovative Composite materials research & development Center (ICC), Kanazawa Institute of Technology for his wisdom and advice.

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