

# BEHAVIORAL INVESTIGATION ON CF/PEEK ROD BY COMPRESSIVE PREPREG TAPE PULTRUSION PROCESS

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### ABSTRACT

This study focuses on the pultrusion process of CF/PEEK rods and their mechanical properties. A laboratory-scale thermoplastic tape pultrusion machine with compressive insert mould was developed to manufacture round rods using unidirectional carbon fibre reinforced poly-ether-ether-ketone (CF/PEEK) prepreg tape. High dense CF/PEEK round rod with no visible voids and fibre bundle layers could be manufactured. Furthermore, in order to investigate the effects of fibre orientations on the mechanical properties of the CF/PEEK rod, the prepreg tape was pultruded in a twisted state. A threepoint bending test and a lateral compression test were performed on the round rod, and the damage behavior was investigated by cross-sectional observation after the test. In addition, the strain distribution of the round rod during a lateral compression test was obtained with a non-contact strain distribution measurement based on digital image correlation (DIC) method. It was found that the maximum load in the three-point bending test and lateral compression test varied depending on the position in the compressive mould along the pultrusion direction, and stabilized and reached a peak when cooled below the glass transition temperature of PEEK polymer. The maximum lateral compression load of the CF/PEEK rods manufactured using twisted prepreg tape increased with increasing the twist amount. This is because the orientation of the fibres was disturbed by twisting, and the transverse cracks of the CF/PEEK rod did not propagate straight in the longitudinal direction of the rod. This fibre orientation contributes to improve the resistance to the transverse compression loading and the stress redistribution. However, the flexural modulus and maximum bending load of the CF/PEEK rod manufactured using the twisted prepreg tape were lowered because the longitudinal fibres were oriented obliquely.

# **1 INTRODUCTION**

The greatest advantage of advanced thermoplastic composites is that their main actions are melting and solidifying by heating and cooling, therefore, they are highly productive and reusable, and secondary processing such as metallic plastic working and welding is possible. As one of the effective applications, thermoplastic composite rivet fastening [1,2] that can be joined thermosetting composite frames and thermoplastic composite beam stringers for reinforcing outer skin panels in addition to assembling an EV battery boxes and lattice structures and so on. While the manufacturing costs and environmental payloads of forming and joining traditional composite structures are still relatively high, thus the various mobilities and infrastructures of the future require more efficient structural design and low-cost manufacturing process. Thermoplastic CFRP pultrusion process has the potential to produce long-span reinforcements at low cost and with high efficiency [3,4]. In addition, the lattice structure can be assembled using thermoplastic CFRP pultruded rods, which is very effective for lightweight and tough structure such as cylinders and domes. It was not easy to continuously produce high-density molded parts by completely impregnating carbon fiber bundles with the high-melt-viscosity super engineering plastics. In recent years, these members can be manufactured by thermoplastic pultrusion process and continuous compression moulding (CCM) using consolidated prepreg sheets, but the details of the manufacturing mechanism have not been elucidated [5, 6].

In this study, CF/PEEK prepreg sheets were used to manufacture round rods using a laboratory-scale compact machine developed originally. The behavior of this process was investigated by measuring the internal temperature of the rod and observing cross-section of the rod. Also, the mechanical properties of the rod were evaluated by a lateral compression test and a three-point bending test. Furthermore,

pultruded rods manufactured from twisted prepreg tapes to improve mechanical properties. The fibres are helically twisted at an angle to the axis, providing not only unidirectional strength, but also strength against lateral forces such as lateral compression and shear.

### 2 EXPERIMENTAL METHODS

#### 2.1 Compressive prepreg tape pultrusion process

A laboratory-scale thermoplastic prepreg tape pultrusion machine developed originally is shown in Figure 1. In this drawing, processing proceeds from left to right. The machine consisted of slitter, heating die, press die, holding die, cooling die, and drawing device. Compact and high-quality manufacturing is achieved through two major features: 1) in-situ slitting to reduce the size of equipment by a unique tape feeding system, and 2) high consolidation of pultruded products through using compressive mould insert. Since the prepreg sheet is already fully consolidated, there is no need to struggle to impregnate the fibre bundles with the high melt viscosity polymer. In the preforming zone, the prepreg sheet emerges from rolls, is continuously slit to the desired tape width, and is fed directly into the square holes of the heating die. In the moulding zone, these tapes pass through upper and lower die cavities heated by cartridge heaters. During the time, these tapes are continuously pressed by lowering the mould insert from a square-shaped cavity to a round cavity. The rod is then cooled to the glass transition temperature of PEEK polymer under compressive loading in a straight cavity. Finally, in the drawing zone, a double belt drawing machine is used to continuously force the rod out of the die at a constant speed.



Figure 1: Manufacturing of CFRTP round rod by laboratory-scale prepreg tape pultrusion machine.

#### 2.2 Material and CF/PEEK rod

A unidirectional carbon fiber reinforced poly-ether-ether-ketone (CF/PEEK) prepreg sheet with fiber volume fraction of  $V_f$ =58% (Toray Advanced Composites, Cetex<sup>®</sup>, TC1200) was used, and the round rod with outer diameter of  $\phi$ 6.5mm was manufactured under the pultrusion conditions of temperature and pressure as shown in Figure.2. Preset temp,  $T_p$  is the temperature set in the heating controller and Pressure, P is the assumed pressure rate in the die. The materials are preheated reaches to the melting temperature in heating die (z=150-330mm) and pressed gradually by tapered insert die in pressing die (z=150-330mm) at constant temperature. After that, the moulding pressure is kept, and the temperature decreases up to the glass transition temperature in holding die (z=330-420mm). The moulding pressure is held, and the temperature decreases below the glass transition temperature in cooling die (z=420-600mm). Temperature, T shown in Figure.2 was measured during pultrusion process using a thermocouple set at z=300mm from the end of the prepreg sheet which was insulated with

polyimide tape. The temperature was set at 450°C for the pultrusion in which the temperature was measured. The temperature inside the rod rises slightly later than the temperature set for the mold. The rise from set temperature observed due to friction during pultrusion.



Figure 2: Pultrusion conditions for CF/PEEK rod.

# 2.3 Evaluation of CF/PEEK rods by mechanical test

The mechanical properties of CF/PEEK round rod were evaluated by (a) three-point bending test and (b) lateral compression test, as shown in Figure 3. (b) In lateral compression test, a rod placed on a flat table is compressed by an upper flat table. Rods manufactured by heating and pressurizing laminated tapes may have continuous large voids between layers if they are formed under insufficient heating and pressurization. Therefore, it is necessary to check the damage behaviour due to the pressure applied between the upper and lower contacts of the rod. Testing was performed at room temperature.



Figure 3: Testing methods for evaluation of CF/PEEK round rod.

#### **3 RESULTS AND DISCUSSIONS**

#### 3.1 Cross-sectional images of pultruded rod

Figure 4 shows the cross-sectional images of pultrusion of a CF/PEEK rod taken out from above after opening the die. In the heating die and former part of pressing die (z=0-200mm), the multilayered prepreg tape can be seen separately in the height direction. There are no visible voids in the prepreg sheet because the compaction has been completed. However, after these slitted tapes are inserted into heating die, the tapes were melted and softened at suitable location by heating. There are many air gaps between the prepreg tapes, the tapes are extended in the thickness direction, and some voids can be seen in these thin layers, as shown in Figure 4(i) z=75 mm. In pressing die, as shown in Figure 4(iii) z=300mm, the shape is not circular but elongated elliptical, leaving linear voids between the tapes. Then, during pultrusion, a compressive load was gradually applied to increase the density. In holding and cooling die, the shape became circular and small burrs occurred on both sides of the matched dies of the mating mold parting line, as shown in Figure 4(iv) = 500 mm. After exiting the die, the shape is circular without no visible voids as shown in Figure 4(v) z=700 mm. Further, from the axial cross-sectional image of the rod shown in Figure 4(vi), it can be seen that striate grooves are formed on the rod surface along the longitudinal direction of the carbon fibers. Also, from the cross-sectional images of the rod shown in Figure 4(vii), it was confirmed that uniform and highly dispersed carbon fibers and polymers were observed inside the rod.



Figure 4: Cross-sectional images in prepreg tape pultrusion processing of CF/PEEK rod.

#### 3.2 Mechanical properties of pultruded rod

Figure 5 plots the maximum loads for lateral compression test and three-point bending test towards position from the die gate of CF/PEEK rods. The maximum lateral compression load increases up to the position of z=350mm and remains constant along the pultrusion direction after this position. Also, maximum three-point bending load increases linearly up to the position of z=300mm and remains constant over this position. After 300mm, it is cooled down to below the glass transition temperature about 420mm, and the strength stabilizes when compression and cooling are completed.



Figure 5: Max lateral compressive load and max Three point-bending load in Position from die gate.

Figure 6(a) shows the three-point bending test results of pultruded CF/PEEK rods with various mold set temperature of 400°C, 450°C, and 500°C, respectively. Figure 6(b) is an image of a longitudinal section of the sample after testing. It can be seen that compression buckling occurs on the compression side at the highest point of 450°C, indicating sufficient strength. However, no significant compressive buckling was observed at 400°C and 500°C. However, rods manufactured at 400°C and 500°C show no obvious buckling, so the damage behavior needs further confirmation.



(a) Three-point bending test results



(b) Images of the longitudinal section

Figure 6: Three point-bending test of CF/PEEK rod with various mould set temperatures.

Figure 7 shows the lateral compressive test results of CF/PEEK rods with various mould set temperatures as well as the three-point bending test. Figure 7(b) shows the damage state of cross-sectional images of the sample after the lateral compression test. Lateral compression test results showed the highest strength at 450°C as well as three-point bending. Cross-sectional observations show that longitudinal cracks predominated in fracture in rods pultruded at 400°C, but radial cracks are more frequent than vertical cracks. At 500°C, longitudinal cracks are dominant, but delamination is observed in part of the cross-section, and the effect of temperature must be confirmed.







Figure 7: Lateral compression test of CF/PEEK rod with various mould set temperatures.

#### 3.2 Mechanical properties of twisted pultruded rod

Next, the evaluation results when inserting the twisted material into the mold and pultrusion forming the CF/PEEK rod are shown. The number of twists was 30T/m and 66T/m. First, Figure 8 shows a cross-sectional observation result of the CF/PEEK rod by X-ray CT. Figure 8(a) shows a cross-sectional image of an untwisted CF/PEEK rod, and Figure 8(b) shows a cross-sectional image of a pultruded CF/PEEK rod with a twist number of 66T/m. Cross-sections of untwisted straight rods show no discernible pattern of fiber orientation. However, the cross-section of a rod with a twist of 66T/m exhibits a concentric circular pattern due to the applied twist.



Figure 8: Cross-sectional image of straight and twisted CF/PEEK rod by X-ray CT.

Figure 9 shows (a) three-point bending test results and (b) longitudinal section images of the straight rod, 30T/m and 66T/m twisted rod. It was found that an untwisted rod simply with prepreg tape overlaid achieved the highest the three-point bending maximum load. However, rods with twist numbers of 30T/m and 66T/m had lower elastic moduli and significantly reduced maximum three-point bending load. It has been suggested that the flexural modulus is significantly reduced because the longitudinal fibres are angled respect to the axial direction. Fracture behaviour of the twisted rod did not show compressive buckling, and significant fracture occurred on the tensile side, and the crack propagated diagonally from the longitudinal cross-sectional images. Obliquely oriented fibres were observed in the image of the rod with a twist number of 66T/m, and it was observed that the cracks also propagated along the fibre orientation.



(a) Three point-bending test results



(b) Images of longitudinal section

Figure 9: Three point-bending test of CF/PEEK rod changed number of twists.

Figure 10 shows the maximum load and the displacement at the maximum load in three-point bending test when the numbers of twists is changed. The results show the maximum load in three-point bending decreases at an almost constant rate with increasing number of twists. Also, the displacement at maximum load tends to be large.



Figure 10: Maximum load and displacement at maximum load in three-point bending test versus numbers of twists.

Figure 11 shows the results of the lateral compression test of CF/PEEK rod changed number of twists. For untwisted rods, the maximum load is reached at lower load levels and decreases rapidly. However, for rods with twist numbers of 30T/m and 60T/m, a linear increase in load was observed, followed by stress redistribution after primary failure (S-(ii)) and load increase again. From the results of strain distribution measurements, it can be observed that untwisted rod mainly develops longitudinal cracks, becomes embrittled and fractures. It was the main factor in the load reduction. On the other hand, cracks were observed in various directions in the rod with a twist number of 66T/m. Then, when load and displacement are applied, the cross-sectional shape changes from circular to elliptical. Fracture behavior is not brittle and cracks propagate in multiple directions. Cracks propagating for multiple directions result from fiber orientation. This fiber orientation contributes to the resistance to compression and stress redistribution after primary failure (T-(ii)).



Figure 11: Lateral compression test of CF/PEEK rod changed number of twists.



Figure 12: Strain distribution and cross-section images in lateral compression test of CF/PEEK rod changed number of twists.

# **4** CONCLUSIONS

The maximum load in lateral compression and three-point bending tests of untwisted CF/PEEK rods depends on the position in compressive die along the pultrusion direction, reaching a maximum upon cooling below the glass transition temperature of PEEK polymer and then stable. CF/PEEK rods pultruded at different mold set temperatures were evaluated through three-point bending and lateral compression tests. In the three-point bending test, the rod pultruded at 450°C showed the highest strength, and there was a difference in the presence or absence of compression buckling. No compressive buckling was observed at 400°C and 500°C. Damage behavior needs to be further confirmed for rods manufactured at these temperatures. A significant temperature dependence was observed in the lateral compression test. The rods exhibited variability in vertical cracking and delamination. However, further investigation is needed to confirm these findings. Pultruded CF/PEEK rods with twist numbers of 30T/m and 66T/m exhibit lower elastic moduli in three-point bending due to the angular orientation of the machine direction longitudinal fibers and lower flexural modulus, maximum load is reduced. However, this fiber orientation contributes to the resistance to compression and stress redistribution.

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