Preparation and Properties of Continuous Carbon Fiber Reinforced Polyether ether ketone Prepreg Tapes

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Abstract. The continuous carbon fiber reinforced polyether-ether-ketone (CCF/PEEK) prepreg tapes were prepared by a wet powder impregnation process. The effect of processing parameters on fiber content, tensile strength and fracture morphology of the prepreg tapes were investigated. The results show, that the fiber content increased with increasing pulling speed and reducing concentration of PEEK suspension. The results of tensile property show remarkable enhancements in tensile strength of prepreg tapes with increased processing temperature and fiber content. Scanning electron microscope observed the plasticizing of PEEK matrix and interfacial adhesion between fibers and PEEK matrix were improved with increasing processing temperature.

Introduction

In recent decades, improvements in the properties and manufacturing of fiber reinforced polymer composites led to increase in application of these composites in aerospace, automotive [1], etc. The bulk of such structures used thermostet composites despite the excellent mechanical and thermal properties exhibited by composites based on high-performance engineering thermoplastic polymers. Presently, it has attracted great interest in fiber reinforced thermoplastic composites based on polyaryletherketones (PAEKs) with excellent mechanical properties, high temperature resistance, corrosion resistance and so on, which applied in automotive, aerospace motor casings, heating-removal fans and alternative energy [2,3]. Polyether ether ketone (PEEK) was the most widely applied in high pressure, high temperature in the PAEK family. PEEK can withstand higher service temperatures due to their higher glass transition temperatures and higher melt temperature.

A great amount of effort has been devoted to manufacturing continuous fiber reinforced thermoplastic composites (CFRTPCs). A key element was achieving full impregnation into
reinforcing fibers during manufacturing CFRTPCs. PEEK matrix had very high viscosity, which added difficulties to manufacturing composite. To solve this problem, it developed new technology for the fabrication of CFRTPCs, for example, micro-braiding fabrication [4], the wrapped yarn [5], solution impregnation [6] and powder impregnating method [7] and so on. The first three methods introduced above which were complex and had high production cost, which limited their application in industrial production. However, a wet powder impregnation process had advantages of low concentration of surfactant additive, few health hazard and easy handling of additives, simple processing method and low cost. In recent decades, researchers prepared natural and glass fiber reinforced polypropylene composites [8,9], continuous carbon fiber reinforced polyphenylenesulfide composites [10,11] by the powder impregnation process. However, there were few studies on the preparation of continuous carbon fiber reinforced polyether-ether-ketone (CCF/PEEK) composites by the wet powder impregnation process, especially the CCF/PEEK prepreg tapes.

In order to further improve the preparation efficiency and reduce the cost of CCF/PEEK composites, in the present work, CCF/PEEK prepreg tapes were prepared by the wet powder impregnation process. Meanwhile, the effect of processing parameters on fiber content and tensile strength of prepreg tapes were investigated. The fracture morphology of prepreg tapes with different processing temperature were examined by SEM.

Experimental

Materials. The thermoplastic matrix used in this work was PEEK powders, purchased from Jilin Joinature Polymer Co., Ltd., China. Continuous carbon fibers (CCF: T700SC-12K) were purchased from Japan Toray with tensile strength of 4900 MPa and modulus of 230 GPa. Deionized water was homemade in the laboratory. Solvents and other additives were domestic industrial products.

Preparation of Prepreg Tapes. The preparation of CCF/PEEK prepreg tapes included the following steps. Firstly, weighing a certain amount of PEEK powders, solvent and surface active agent were slowly added into deionized water by magnetic shirring for 4h. The PEEK suspension were well maintained. Secondly, the PEEK suspension put in the sizing tank. And then, the CCF were spread evenly in the sizing tank by adjusting the tension, which made the PEEK attach to the surface of the CCF. The fiber bundles were pre-prepared in the self-designed preheated melting mould, and the CCF/PEEK prepreg tapes were prepared by extrusion and finally coiled. Schematic diagram for process of CCF/PEEK prepreg tapes is shown in Fig. 1.

![Schematic diagram for process of CCF/PEEK prepreg tapes](image-url)
Measurements

**Tensile Tests.** Tensile properties were measured using a Model 5969 universal material testing machine manufactured by INSTRON, Inc. Tensile specimen of the prepreg tapes were prepared according to ASTM D3039/D3039M-14. The specimens dimensions were 250 mm×15 mm×0.1 mm and the stretching speed was 10 mm/min.

**Microscopy Analysis.** Fracture surfaces of tensile specimens for the prepreg tapes were sprayed with gold, and were observed by a Quanta 250 scanning electron microscope manufactured by FEI Inc. of the United States.

Results and Discussion

**Composites Preparation.** The effect of concentration of PEEK suspension on carbon fiber content of the prepreg tapes is presented in Fig. 2. With the relative concentration of PEEK suspension increased from 6.5 wt% to 8 wt%, the fiber contents decreased from 70 wt% to 40 wt%, respectively. These phenomenon indicated that the more PEEK particles attached to the surface of the CCF when the high concentration of PEEK suspension was in the sizing tank. Fig. 3 shows the influence of pulling speed on carbon fiber content of the prepreg tapes. It can be seen that pulling speed had a significant effect on carbon fiber content of the prepreg tapes. The carbon fiber content of the prepreg tapes increased from 60 wt% to 82 wt% with the pulling speed increased from 4 r/min to 14 r/min at the concentration of PEEK suspension of 7 wt%. Therefore, it can be concluded that increasing pulling speed resulting in decreasing the capacity of PEEK particles attaching to the surface of the CCF.

Fig. 2. Effect of PEEK powder concentration of suspension on carbon fiber content of prepreg tapes (pulling speed was 4 r/min)
Fig. 3. Influence of pulling speed on carbon fiber content of prepreg tapes (concentration of PEEK suspension was 7 wt%) 

Tensile Strength 

**Processing Temperature.** The tensile strength of CCF/PEEK prepreg tapes with different processing temperature is presented in Fig. 4. The tensile strength of CCF/PEEK prepreg tapes was 1466 MPa when the processing temperature was 320 °C. It improved gradually with increasing the processing temperature. When the processing temperature was 350 °C, the tensile strength of CCF/PEEK prepreg tapes increased by 18.15% from 1466 MPa to 1732 MPa. Therefore, the results demonstrated that increasing processing temperature was an effective way to improved tensile strength of the prepreg tapes.

Fig. 5 shows the fracture morphology of CCF/PEEK prepreg tapes with different processing temperature. As depicted in Fig. 5(a), the plasticizing of PEEK matrix and interfacial bonding between CCF and PEEK matrix were poor. PEEK matrix was in the “high elastic state" while the processing temperature was 320 °C, which influenced the melt flow behavior during impregnation. As can be seen from Fig. 5(b) and Fig. 5(c), when the processing temperature increased from 320 °C to 340 °C, the impregnation was remarkably improved as compared with Fig. 5(a), but some gaps were observed between CCF and PEEK matrix. As shown in Fig. 5(d), fibers uniformly dispersed and wrapped with substantial PEEK matrix with less voids, which illustrated the better interfacial adhesion between fibers and PEEK matrix. It can be concluded that the impregnation of CCF/PEEK prepreg tapes was improved with increasing processing temperature, which was an important reason for the tensile strength of CCF/PEEK prepreg tapes improved gradually in Fig. 4.

Fig. 4. Tensile strength of CCF/PEEK prepreg tapes with different processing temperature
Fiber content. The mechanical properties of fiber reinforced composites were mainly determined by fiber. The composite was subjected to an external load, which could be effectively transmitted to the fiber through the resin matrix, thus carbon fiber played a critical enhancement role in the composite. Fig. 6 shows the tensile strength of CCF/PEEK prepreg tapes with different carbon fiber content. It can be seen that, the tensile strength of CCF/PEEK prepreg tapes increased with increasing the carbon fiber content. The tensile strength of pure PEEK was only 103 MPa, and the tensile strength of CCF/PEEK prepreg tape with CF content 70 wt% was up to 1732 MPa, and increased by 16.8 times comparing with pure PEEK, which was improved by 61.87% as compared to that of CCF/PEEK prepreg tapes with the 50 wt%. It can be attributed to the carrying capacity of fiber was higher than the matrix, resulting in tensile strength of the prepreg tapes improving with adding the carbon fiber.
Conclusions

In this work, CCF/PEEK prepreg tapes prepared by a wet powder impregnation process. The effect of processing parameters on fiber content, tensile strength and fracture morphology of the prepreg tapes were investigated. The results show, that the carbon fiber content increased with increasing pulling speed and reducing concentration of PEEK suspension. Mechanical tests show remarkable enhancements in tensile strength of prepreg tapes with increasing processing temperature and fiber content. The tensile strength of the prepreg tape was 1732 MPa, after increasing fiber content and processing temperature to 70 wt% and 350 °C, respectively. It can be attributed to the loading external force of fiber content increasing with adding carbon fiber, when the CCF/PEEK prepreg tapes was subjected to an external load. Morphological observations revealed that interfacial adhesion between fibers and PEEK matrix were improved as increasing processing temperature, which was an important reason for the tensile strength of the prepreg tapes.

References