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Experimental investigation of carbon fiber reinforced poly(phenylene sulfide) composites prepared using a double-belt press

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ABSTRACT

The high-performance carbon fiber reinforced poly(phenylene sulfide) composites were continuously fabricated using thermoplastic prepregs in a double-belt press. The effects of process velocity on the composite consolidation quality and mechanical properties were investigated. It is found that the tensile and interlaminar shear properties of composites prepared using the double-belt press are comparable to that of compression-molded composites when the process velocity is no more than 0.20 $m \cdot min^{-1}$. The composite fracture morphologies also show different failure mechanisms between different samples and indicate that the interfacial adhesion strength may play a vital role in the mechanical properties of CF/PPS composites. Furthermore, experimental results show that the heating time above 330 °C should be over 440 s and the void content should be lower than 2.38% in order to obtain high performance CF/PPS composites.

1. Introduction

Advanced polymer composites (APCs) have obtained significant applications as structural materials in aerospace and civil engineering industries due to their outstanding mechanical properties, low weight and elevated temperature resistance [1–3]. High-performance thermoplastic composites are playing an increasingly important role in APCs because thermoplastic composites have many advantages over thermosetting composites, such as high impact tolerance and toughness, improved chemical and corrosion resistance, short processing cycle, unlimited shelf life of prepregs and recyclability [4–6]. Poly(phenylene sulfide) (PPS) is a widely-used semicrystalline polymer and possesses superior mechanical and thermal properties, good chemical and aging resistance, inherent flame retardancy, low water absorption and excellent friction properties [7,8]. Its mechanical properties can be further enhanced by reinforcing carbon fibers and CF/PPS composites have promising applications in aeronautical and other industrial fields as excellent engineering materials [9–12]. In recent years, the booming global automobile industries bring great opportunities and markets for thermoplastic composites. However, the mass production of thermoplastic composites has been a great challenge for their wide applications. Intermittent production techniques, e.g. compression molding press and autoclave, often prepare composites with extremely low flaws and excellent mechanical properties. These methods are acceptable for aircraft manufacturing which particularly emphasizes quality and safety, but they are not suitable for civil industries due to high production costs. A rapid and low-cost manufacturing method is of great interest to the industries [13]. Double-belt press is a high-efficiency machine to produce fiber reinforced thermoplastic composites [14], in which raw materials are successively and automatically conveyed through heating and cooling zones under a certain pressure and meanwhile resin matrix is melted and impregnates reinforcing fibers, followed by consolidation and solidification [15,16]. Thus, the production efficiency of the double-belt press is considerably higher than that of intermittent methods. Many literatures [4,5,7,8,17–21] have reported the influence of the filler contents, processing parameters, thermal treatment and other factors on consolidation, mechanical and tribological properties of the CF/PPS composites. Some researchers [15,16,22] also have studied the impregnation and mechanical properties of PA and PP matrix composites fabricated using the double-belt press technology. However, little research has been conducted on CF/PPS composites prepared using the double-belt press method.

In this paper, CF/PPS composites were prepared using thermoplastic prepregs in a double-belt press. The effects of process

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velocity on void contents and distribution, mechanical properties and morphologies of CF/PPS composites were investigated. The crystalline properties were examined by differential scanning calorimetry (DSC) and thermo gravimetric analyzer (TGA). The composite fracture morphologies were detected using scanning electron microscopy (SEM).

2. Materials and methods

2.1. Materials and composites preparation

Two types of single-sheet thermoplastic CF/PPS prepregs were used to prepare composites and their basic properties are listed in Table 1. Unidirectional (U) and twill (T) fabric prepregs were hot-melt impregnated using PPS films mixed with carbon fiber yarns and fabrics on a self-developed prepreg machine in our laboratory, respectively. The prepregs were cut into 45 cm × 25 cm, with the length direction parallel to the fiber yarns of the unidirectional prepregs. Five fabric layers and eight unidirectional layers were stacked as the following sequence: [TUUTUUTUUTUUT]. The forming temperature was set at 340 °C and the double-belt velocities were 0.15, 0.20, 0.25 and 0.35 m·min^{-1}, respectively. The gap between the upper and the lower steel belts was fixed at 2.5 mm. When the temperature was stable in the range of 340 ± 5 °C, the stacked prepregs inserted between two polyimide films were put into the double-belt press. Depending on the velocity, raw materials were retained in the heating zone of the double-belt press for 6 ~ 14 min. The polyimide films were 15 μm in thickness and were used to prevent the melt resin from adhering to the steel belts. They were easily removed after the composites were cooled down. Another CF/PPS composite laminate was also prepared by a compression molding press, with a temperature of 340 °C, a pressure of 4 MPa and a clamp time of 10 min.

2.2. Characterization

The experimental density (ρ_e) of CF/PPS composites was measured using a densimeter (AND EK-300iD) by measuring the mass of specimens in the air and in distilled water. The theoretical density (ρ_t) of fully consolidated composites was calculated by Eq. (1):

$$\rho_t = \frac{\rho_f \cdot \rho_m}{w_f \cdot \rho_m + w_m \cdot \rho_f} \tag{1}$$

where ρ_f is the density of carbon fibers, 1.8 g·cm^{-3}; ρ_m is the density of PPS, 1.35 g·cm^{-3}; w_f and w_m are the weight fraction of the fiber and matrix, respectively. The matrix weight fraction of composites was tested using the digestion method in concentrated nitric acid at 100 °C for 2 h according to ASTM D3171. The apparent void contents (X_v) of composites were calculated using Eq. (2):

$$X_v = \frac{\rho_t - \rho_e}{\rho_t} \tag{2}$$

Table 1
Properties of the unidirectional and twill fabric CF/PPS prepregs.

	Unidirectional	Fabric
Raw fibers	T700, 12 K, Toray	T300, 3 K, 2/2 twill, Mitsubishi
Areal weight/g·m^{-2}	203 ± 7.6	404 ± 7.9
Thickness/mm	0.16	0.31
Resin content/wt.%	51.3 ± 0.8	50.6 ± 0.2
Fiber weight/g·m^{-2}	96 ± 3.7	194 ± 7.9
Volatile content/wt.%	<2	<2
Degree of consolidation	Partially	Partially

The crystal property of the matrix PPS was examined by DSC (Perkin–Elmer Pyris Diamond). Fragments around 7 mg were carefully cut from the pre-dried specimens and scanned from 30 °C to 350 °C at a heating rate of 10 °C·min^{-1} under a N_2 atmosphere. Then the samples were pyrolyzed in TGA (Perkin–Elmer Pyris Diamond). The temperature range and the heating rate were 60 ~ 700 °C and 20 °C·min^{-1}, respectively. N_2 atmosphere (200 mL·min^{-1}) was applied to prevent carbon fibers from weight loss. The degree of crystallinity, X_c, was calculated according to the following equations:

$$w_m = \frac{1 - R_s}{1 - R_{pps}} \tag{3}$$

$$X_c = \frac{\Delta H_c}{w_m \times \Delta H_m^0} \tag{4}$$

where w_m is the PPS weight fraction of the sample; R_s and R_{pps} are the residue rate of the sample and neat PPS after pyrolysis in TGA, respectively; ΔH_c is the melting enthalpy of the sample; ΔH_m^0 is the melting enthalpy of fully crystalline PPS, 112 J/g [21].

The mechanical properties of CF/PPS composite laminates were tested on an Instron 5985 universal testing machine. The length direction of specimens was parallel to the unidirectional carbon fibers in the composite laminates. The tensile strength, modulus and breaking elongation were measured at a speed of 2 mm·min^{-1}, according to ASTM D3039. The tensile fracture toughness was evaluated from the area under the tensile stress–strain curve. The flexural properties were determined using the three-point bending method conforming to ASTM D7264, the span-to-thickness ratio was 32 and the crosshead speed was 1 mm·min^{-1}. The interlaminar shear strength (ILSS) was measured according to ASTM D2344, the span-to-thickness ratio was 4 and the crosshead speed was 1 mm·min^{-1}. The final results and standard deviation for each group of samples were derived by averaging 6 specimens.

The distribution of voids and carbon fibers in the composites were detected using SEM (TM1000 Hitachi). The cross-sections of samples were polished and then cleaned in an ultrasonic bath. Morphological observation was carried out using tensile and interlaminar shear fractography by SEM. All surfaces were coated with gold using an ion sputtering machine.

3. Results and discussion

3.1. Consolidation properties of CF/PPS composites

After immersed in the hot concentrated nitric acid for 2 h, the matrix PPS was completely digested. The matrix weight fraction of the prepared CF/PPS composites was 48.0% ± 0.16%. This value is slightly less than that of the unidirectional and fabric prepregs, indicating that a very small amount of resin was squeezed out during the thermoforming processes. According to Eq. (1), the ρ_t of fully consolidated CF/PPS composites was 1.552 g·cm^{-3}, and the void contents and fiber volume fraction (V_f) of composites were calculated and summarized in Table 2. The composites prepared using the compression molding press and the double-belt press are noted by “CFPPS-M” and “CFPPS”, respectively. The process velocity is noted after the sample ID. For example, CFPPS-0.15 means a velocity of 0.15 m·min^{-1} in Sample CFPPS.

As shown in Table 2, the velocity has little effect on the V_f of CF/PPS composites. It seems that the sample of CFPPS-M has the largest value of ρ_e (1.549 g·cm^{-3}) while the lowest void content value of 0.19%, which means that CF/PPS composites are fully consolidated using the compression molding press method. The