PREPARATION OF SOLUTION IMPREGNATED CONTINUOUS CARBON FIBRE REINFORCED POLY (PHTHALAZINONE ETHER SULFONE KETONE) COMPOSITES

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ABSTRACT

Continuous carbon fibre reinforced Poly (phthalazinone ether sulfone ketone) (PPESK) composites were prepared using a solution impregnation process and hot-press molding method. The effects of polymer solution viscosities on fibre impregnation, fibre volume fraction and thereby on mechanical properties of composites were studied. The results show that the fibre impregnation and fibre volume fraction decreased with increasing polymer solution viscosities, and the mechanical properties of composites mainly depended on the fibre volume content.

Keywords: Composites, Carbon fibre, PPESK, Solution impregnate

1. INTRODUCTION

Continuous carbon fibre reinforced high performance thermoplastic composites have been extensively used in high technological fields owing to their excellent mechanical properties, stiffness-to-weight ratios, toughness, shorter processing time, and repairability. In order to wet together the two components, the thermoplastic matrix and the reinforcement fibres, several combining processes have been developed [1-6]. Among these processes, the hot-melt process has much more extensively been adopted, especially in the case that the matrix is a semi-crystalline high performance thermoplastic like polyetheretherketone (PEEK). Generally, the polymer matrix should go through a melting stage before it physically wets the reinforcement fibres and several alternative approaches have been developed to achieve this goal. However, since polymer melt usually has a very high viscosity, the hot-melt process melt are very difficult to carry out and have to reduce the production rate and provide relatively high processing pressures.

In addition to hot-melt process, composite prepregs can also be made by impregnating carbon fibres with suitable polymer solution. The benefit of this process is derived from the low viscosity of the resulting polymer solution, allowing both unidirectional composite tapes and woven fabrics to be processed. The processing cost may be substantially reduced. Therefore, solution impregnation is the preferred method to prepare high performance composites based on a soluble high performance thermoplastic [7].

Poly (phthalazinone ether sulfone ketone) (PPESK) is an amorphous high performance thermoplastic copolymer, having a special noncoplanar-twisted-aramatic molecular structure as a result of the introduction of phthalazinone moiety. It, therefore, shows many outstanding properties, such as very high glass transition temperatures (T_g), outstanding high temperature stability, excellent mechanical properties, and radiation stability [8]. PPESK with the ketone-to-sulfone ratio of 1 in the backbone has a T_g of 284°C, higher than most of commercial thermoplastics. Moreover, PPESK shows good solubility in some organic solvents, resulting in the possibility to prepare continuous fibre reinforced composites by the solution impregnating method. All of these make

PPESK a very ideal alternative of matrix for high performance composites.

In this study, continuous carbon fibre reinforced PPESK composite prepregs were prepared using a solution impregnation process. Then these prepregs were molded into unidirectional composite panels by hot compression. The effects of polymer solution viscosities on fibre volume content and thereby on mechanical properties of composites were investigated.

2. EXPERIMENTAL

The carbon fibre employed in this study was T700 (12k) produced by TORAY. Fine powders of PPESK with the ketone-to-sulfone ratio of 1 were provided by Dalian Polymer New Material Co. The inherent viscosity of PPESK is 0.55 dL/g (25 °C, in chloroform).

Different concentration solutions of PPESK with a mixture solvent were firstly prepared. The mixture solvent was composed of NMP, DMAc, chlorobenzene with the equal volume composition. Then continuous carbon fibre bundles were impregnated for 25sec in polymer solution, and the individual bundles were wound on to a stainless steel plate. The aligned bundles were welded together owing to the viscosity of polymer solution. Subsequently, the solution-processed prepreg was allowed to dry on the spin drum and then vacuum oven treated at 100 °C to 200 °C for 10h before cutting.
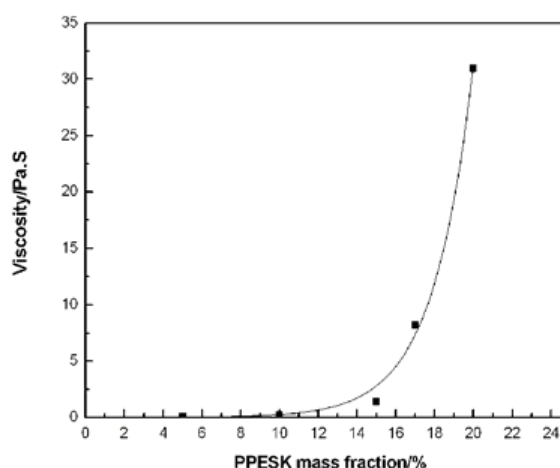
The prepreg was laid up and compressed into unidirectional composite panels using a hot-press molding method. In the first stage of the cycle the press was closed with a steel mould comprising the test laminate stack and the main pressing plates were heated from room temperature to 320 °C at heating rate of 10 K/min. Under this temperature, a pressure of 12 MPa was used to mold the prepreg and kept for 30 min. Thereafter the mould was cooled down to 130 °C at the rate about 5 K/min under the constant pressure until finally the laminate was permitted to take out of the steel mould.

The viscosities of polymer solutions were tested on a NDJ-4 viscosity meter at 23 °C. The fibre volume content was determined according to GB/T3366-1996. Composite flexural property was carried out according to GB3356-1999 on an AG-2000A testing machine with constant crosshead rate of 5mm/min. Morphologies of composite were characterized using a KYKY2800B scanning electron microscope (SEM).
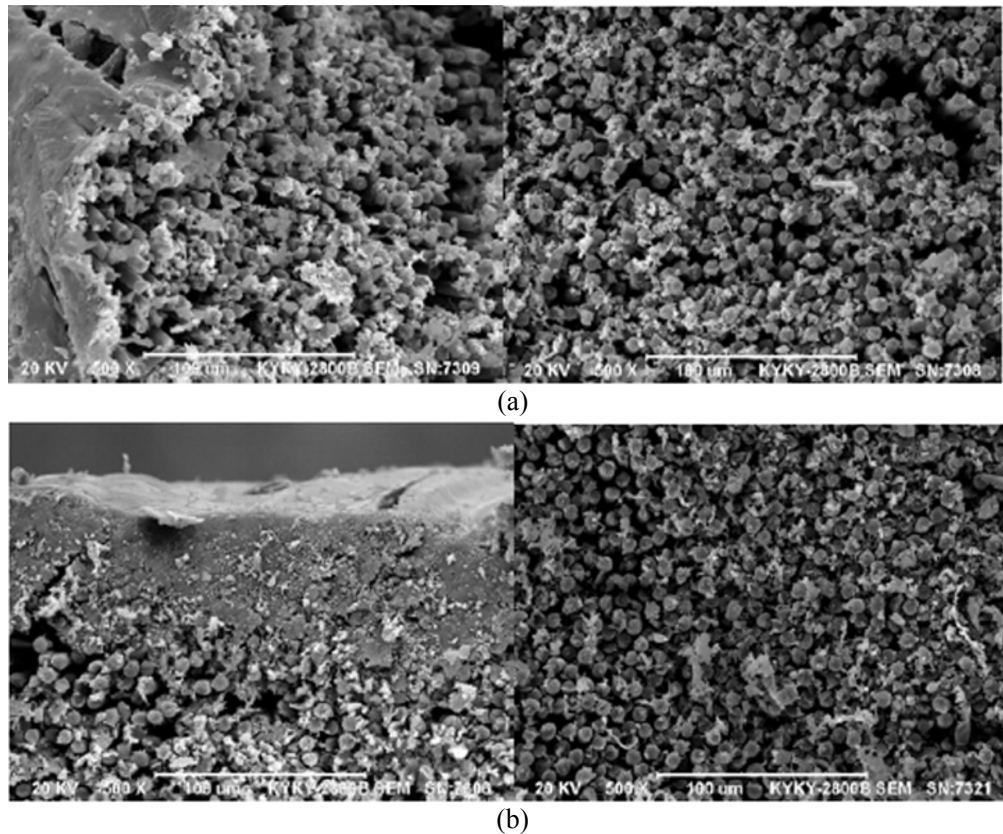
3. RESULTS AND DISCUSSION

Fig.1 shows the viscosity of PPESK solutions as a function of PPESK concentration. The viscosity exhibits an exponential increase with the increase of PPESK content in polymer solution. Compared with the melting impregnation process, the benefit of solution impregnation process is derived from the low viscosity of the resulting polymer solution. The melting viscosities of some high performance thermoplastics are too high for making prepregs with uniform distribution and sufficient impregnation of fibres. Meng et al reported that the melting viscosity was surprisingly about 2×10^6 Pa.s under 350°C and low shear rate, making it very difficult for PPESK to prepare composites by melting impregnation process [9]. Comparatively, the solution viscosity of PPESK is only about 31 Pa.s as PPESK concentration was 20% (Fig.1). Hence, it is preferred to prepare continuous fibre reinforced PPESK composites by solution impregnation process.

Fig.1: Viscosities of polymer solutions as a function of PPESK mass fraction.
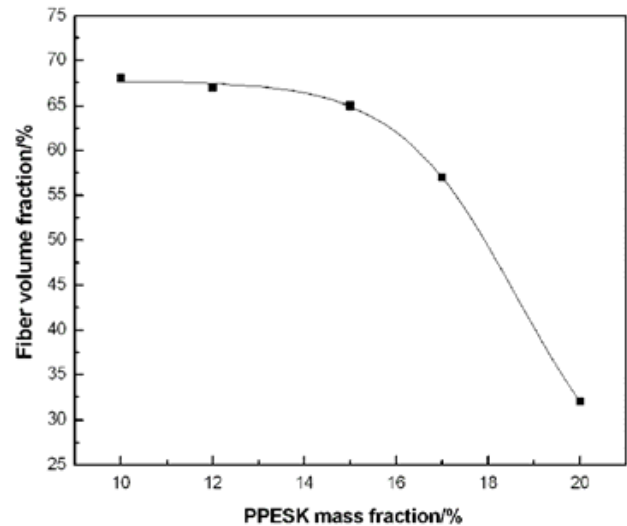
SEM micrographs of transects of the single carbon fibre bundle prepregs impregnated in two different

(a)

(b)

Fig.2: SEM micrographs of transects of the single carbon fibre bundle prepregs impregnated in different solutions (a) PPESK=10 wt% and (b) PPESK=20 wt%

concentration solutions were shown in Fig.2. Because of the same parameters in the solution impregnation process, the viscosity of polymer solution is critical to the impregnation qualities of the single carbon filaments, especially the filaments inside the bundles, and fibre volume fraction. Fig.2a shows that the polymer layer on the bundles' surface is thin, uniform polymer distribution and good fibre wetting inside of the fibre bundles has been achieved when the viscosity is low, indicating that the polymer solution with low viscosity has good permeability and high impregnation velocity. It could be attributed to that easily flowing of the solution with low polymer concentration. In addition, the low viscosity results in the high fibre volume fraction and low polymer content. Contrarily, the polymer solution with high viscosity has relative poor permeability and low impregnation velocity, resulting thick polymer layer on the surface of fibre bundles, scarcity of polymer inside of the fibre bundles, low fibre volume fraction and high polymer content (Fig.2b).

Since most underlying structure and performance

Fig.3: Fibre volume fraction of composites as a function of mass fraction of PPESK solutions

are related to volume fraction, analysis of composite properties is normally carried out considering fibre volume fraction [10, 11]. Fig.3 shows that the fibre volume content of CF/PPESK composites decrease with increasing PPESK content in polymer solution. The main reason was the increasing of the viscos-
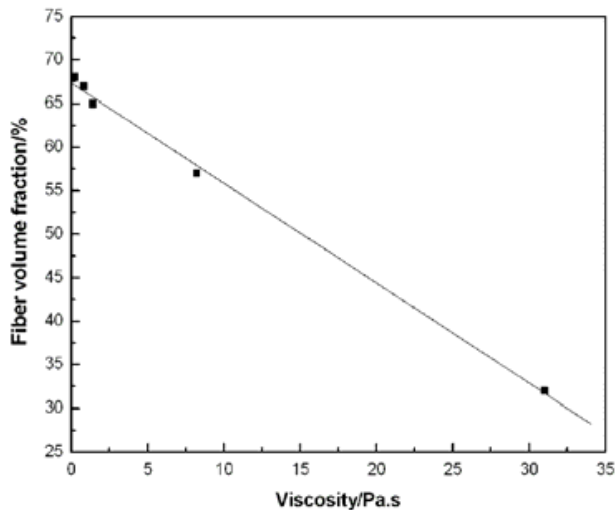
Fig.4: Fibre volume content of composites as a function of viscosities of PPESK solutions

ity of polymer solutions. Fig.4 shows that the fibre volume content of CF/PPESK composites decrease linearly with increasing the viscosity of polymer solutions which increase exponentially with increasing PPESK content in polymer solution (Fig.1).

In this report, the effects of fibre volume contents on the flexural strength and modulus were also investigated (Fig.5). Generally, the flexural strength firstly shows an increased tendency to the peak value of 1092.4 MPa with the increase of fibre volume content, and sharply decreases with the further increase of fibre volume content. However, the flexural modulus keeps increasing with the increase of fibre volume fraction.

Using a calculated regression, R^2 for fibre volume

fraction equals to 0.5568 that can only explain 55% of the variation in flexural strength around the regression line (Fig 5a). It indicates that not all flexural strength experimental date accord with the rule of mixture theoretical date. However, dividing all the experimental date to two separate segments, bellowing 57 vol. %, the flexural strength increased linearly with increasing the fibre volume fraction (R^2=0.9878). It indicates that the CF/PPESK composite samples were composed of sufficiently wetting fibres and load can efficiently be transferred between fibres and matrix. In the other segment, above 57 vol. %, there appears to be distinct deviation from linearity and the decreased tendency. It perhaps is because the polymer matrix fraction is too low to transfer the load between fibres and matrix efficiently. Different from the trend of the flexural strength with fibre volume content, the flexural modulus of CF/PPESK composites increase linearly with increasing the fibre volume content (Fig. 5b). It indicates that the flexural modulus of this class composite has good correlation with the fibre volume percent.

4. CONCLUSIONS

Poly (phthalazinone ether sulfone ketone) (PPESK) is a novel high performance thermoplastic with outstanding high temperature resistance, excellent mechanical properties and special solubility. Meanwhile, PPESK possess very high melting viscosity, but its solutions can offer very low viscocities. Hence it is preferred to prepare continuous carbon
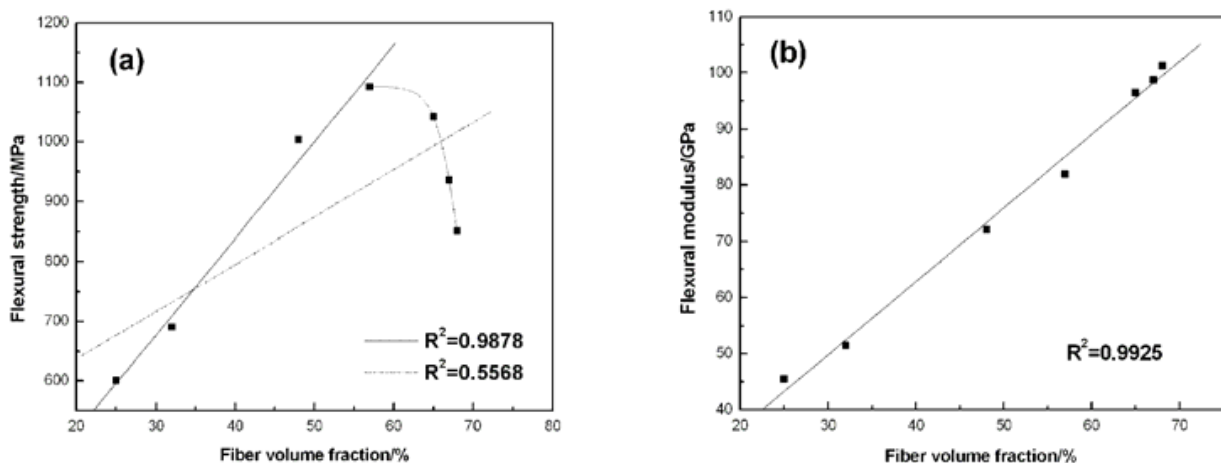

Fig.5: (a) flexural strength and (b) flexural modulus of composites as a function of the fibre volume fraction

fibre reinforced PPESK composite using a solution impregnation process. The results indicated that viscosity of PPESK solutions has critical effects on the impregnation and fibre volume fraction of composites. In the all compositions, the flexural modulus showed a good agreement with rule-of-mixture equation of composites. For the composites with lower fibre content, the flexural strength could be also modeled using the rule-of-mixtures equation. However, as fibre volume content was increased up to above 57%, the flexural strength of the composites sharply decreased with the further increase of the fibre content.

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