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Toughening of carbon fibre/polybenzoxazine composites by incorporating polyethersulfone into the interlaminar region

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ABSTRACT

Two grades of hydroxyl-terminated polyethersulfone (PES), one low and one high molecular weight, were incorporated into carbon fibre/polybenzoxazine (CF/PolyBZ) composites as interlayer toughening agents. A preliminary study on the PES–benzoxazine (BZ) resin mixtures was carried out via hot stage microscopy to observe the dissolution behaviour of the PES in the BZ during the curing cycle. CF/PolyBZ composites with PES in the interlaminar regions were produced by a vacuum resin infusion process and characterised via double cantilever beam (DCB) tests to determine the mode-I type interlaminar fracture toughness (G_{IC}). The DCB fracture surfaces were examined under an optical microscope and scanning electron microscope (SEM). The thermomechanical properties of the composites were also studied via dynamic mechanical analysis (DMA). The incorporation of both grades of PES resulted in a significantly improved G_{IC} and marginally increased glass transition temperature of the composite.

1. Introduction

Benzoxazine (BZ) resins are a relatively new class of thermoset resin which have been developed for high performance composite applications in addition to the traditional resins such as epoxies, phenolics, bismaleimides, polyimides and cyanate esters due to their advantageous features [1]. BZ resins exhibit near-zero shrinkage, very high char yield, fast development of mechanical properties as a function of conversion and high glass transition temperatures [1–3]. Their processing viscosities are suitable for resin transfer moulding (RTM) and vacuum resin infusion, making them excellent candidates for composite applications. However, BZ resins are thermosetting and thus, their high crosslink density does not allow the cured resin to absorb energy under stress and renders them brittle in nature [4,5].

Toughness of thermosetting resins and composites can be enhanced through several techniques such as physical blending with liquid and/or solid rubbers, core-shell particles, thermoplastics, nanotubes and nanofibres and interlaminar toughening via the insertion of thermoplastic films, veils, particles, etc. between layers of prepreg or fibre reinforcements [6,7]. Over the last number decades some of these techniques have been used to toughen mainly epoxy resins for use as composite matrices. A rubber or thermoplastic toughening agent may remain dissolved in the epoxy matrix resin or may undergo reaction-induced phase separation resulting in a morphology and phase size, which are dependent on numerous interacting parameters, giving increased fracture toughness of the composite [8,9,10]. Other toughening agents, such as core-shell particles, remain largely in the form in which they are added. The most widely studied thermoplastics as toughening agents for epoxies have been polysulfones, polyether sulfones, phenoxy resin, polyetherimide, polyphenylene ether, polyethylene terephthalate and polycarbonate [4,8–13].

There are some reports on the toughening of benzoxazine resins. Rimdusit et al. [14] reported that the toughness of polybenzoxazine (BA) can effectively be improved by alloying with isophorone diisocyanate (IPDI)-based urethane prepolymers (PU) or with flexible epoxy (EPO732). The experimental results indicated that the BA/PU alloys rendered superior properties to the BA/EPO732 systems with potential use as high performance polymers or composite matrices. Hamerton et al. [15] synthesised polyarylsulfone (PSU) and polyarylethersulfone (PES) thermoplastics differing in molecular weights and terminal functionalities as toughening agents for benzoxazine resins. These thermoplastics were blended with the benzoxazine resins to enhance the toughness. They observed that PES formed tougher blends with benzoxazine than the equivalent PSU, which might be as a result of the increased phase separation and crack pinning in the PES toughened systems [16]. Although there was a degree of scatter, the trend displayed that increasing molecular weight of the thermoplastic increased the toughness of the blend within each set.

There are very few reports on toughened CF/PolyBZ systems. For the toughening of carbon fibre/polybenzoxazine (CF/PolyBZ)

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Table 1
Loctite® BZ 9130 AERO (Henkel) resin properties cured at 185 °C for 2 h and post-cured at 232 °C for 1 h [19].

Property	Value
Density (g/cm^3)	1.22
Glass transition temperature Onset: dry (°C)	255
Glass transition temperature Onset: wet[a](°C)	196
Flexural strength (MPa)	163
Flexural modulus (GPa)	3.72
Strain energy release rate (J/m^2)	219

[a] 72 h water boil.

Table 2
Properties of the PES particles employed as toughening agents [20,21].

	PES grade Virantage® VW-10700 RFP	PES grade Virantage® VW-10200 RFP
Average molecular weight (g/mol)	21,000	46,500
Solution viscosity (mPas)	560	800
−OH end groups (meq/g)	210	75
Glass transition temperature (°C)	216	220
Particle size (μm)	75	63

composites two types of rubbers have been reported thus far, these being amine-terminated butadiene-acrylonitrile rubber (ATBN) and carboxyl-terminated butadiene acrylonitrile rubber (CTBN). Ishida and Chaisuwan [17] dip-coated de-sized CF in ATBN rubber and investigated the interlaminar toughness of CF/PolyBZ composites. The adhesion between the PolyBZ resin and CF was improved by the ATBN, increasing the delamination toughness. Jang and Yang [18] improved the delamination toughness of CF/PolyBZ composites by using ATBN and CTBN as the toughening agents. ATBN was found to be more effective than CTBN for improvement of mode-II delamination toughness.

No research concerning thermoplastic toughening of polyBZ resin composites has, as yet, been reported. So the overarching aim of the present work was to incorporate PES as an interlayer toughening agent in the CF/PolyBZ composites to improve the Mode-I interlaminar fracture toughness. Two grades of PES of different molecular weight were employed. A morphological study of the BZ-PES mixture was first carried out via hot stage microscopy to understand the behaviour of both PES grades in the BZ. CF/PolyBZ composites with PES particles in the interlaminar regions were produced by a vacuum resin infusion process and characterised via double cantilever beam (DCB) tests to determine the mode-I type interlaminar fracture toughness. The DCB fracture surfaces were examined under optical microscopy and scanning electron microscopy (SEM). In addition, the effect of the PES particles on thermomechanical properties of the composites was studied via dynamic mechanical analysis (DMA).

2. Experimental

2.1. Materials

Benzoxazine resin Loctite® BZ 9130 AERO (Henkel) was employed to manufacture the composites [19] (Table 1). This resin is intended for use in liquid resin infusion processes. Polyethersulfone (PES) particles employed as toughening agents in this study were hydroxyl-terminated polyethersulfone Virantage® VW-10700 RFP and Virantage® VW-10200 RFP, supplied by Solvay Specialty Polymers [20,21] (Table 2). Unidirectional carbon fibre fabrics Tenax IMS60 E13 (Saertex) with an areal weight of 302 g/m^2 were employed for the manufacture of the CF/PolyBZ laminates.

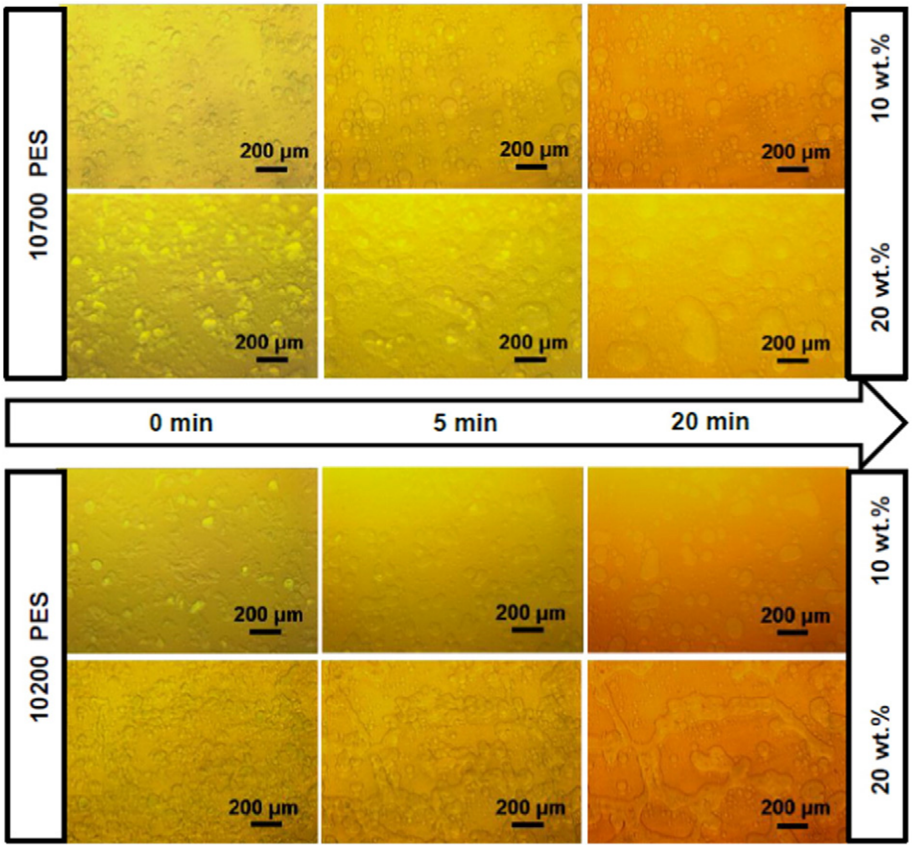

Fig. 1. Images of the PES particles in benzoxazine resin captured in-situ via hot stage microscopy during the resin curing process at 185 °C.