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Optimisation of interfacial properties for tensile strength by plasma polymerisation

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Abstract

Plasma polymerisation was used to coat E glass fibres with an acrylic acid/1,7-octadiene copolymer at varying concentrations of acrylic acid. These fibres were then used to produce unidirectional composite panels. By so doing it was possible to produce composites with varying degrees of adhesion between fibre and matrix. The panels were then prepared into test pieces and tested in tension.

It was found that stress, strain and energy absorbed to failure were higher in specimens produced from plasma polymer coated fibres than from uncoated controls. Tensile strength and energy absorbed in failure were dependent on the concentration of acrylic acid in the monomer flow used to coat the fibres but strain to failure and modulus were not. The presence of a plasma polymer coating increased the longitudinal tensile strength by 20%. By tailoring the composition of the coating to create the optimal degree of fibre–matrix bonding the tensile strength was increased by a further 30%. This optimal degree of bonding was obtained at an intermediate level of interfacial adhesion which was less than the maximum achievable. It was also observed that the plasma polymer coating can change the failure mechanisms and fracture behaviour of the composite.

Keywords: A. Polymer–matrix composites (PMCs); A. Glass fibres; B. Fibre–matrix bond; C. Stress transfer; E. Plasma deposition

1. Introduction

The performance of most polymer composites has long been recognised to depend on a good interface allowing optimal stress transfer between fibre and matrix [1–3]. To achieve this in the case of glass fibres water based sizes containing silanes [4] are common, whereas in the case of carbon fibres, oxidative treatments followed by a polymeric size [5] are used. More recently plasma polymerisation has been proposed as a method of coating fibres to promote the formation of a good interphase between fibre and matrix [6–8].

Plasma polymerisation is a process which allows nanodimensional, conformal and pinhole free polymer coatings to be applied to a substrate of choice [9]. For this work a plasma polymer (PP) produced from co-polymerising acrylic acid and 1,7-octadiene was applied to the surface of glass fibres prior to their incorporation in a high volume fraction (V_f) composite in order to control the degree of adhesion between the fibre and the matrix. It has been shown by Marks [8,10] using the technique of X-ray photoelectron spectroscopy (XPS) with trifluoroethanol derivatisation (TFE) that by varying the ratios of the above two monomers the concentration of carboxylic acid groups on the surface of the polymer coating can be varied. It has been shown by Marks [8] and by Kettle [6,8] using the fragmentation test that the strength of the interfacial bond between a plasma polymer coated glass or carbon fibre and an epoxy resin can be controlled by the concentration of surface carboxylic acid groups.

Longitudinal tensile strength of a unidirectional glass fibre composite was chosen as the parameter to be

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measured against the strength of the interface. Longitudinal tensile strength is often considered to be less dependent on the strength of the fibre–matrix interface than on factors such as the strength and volume fraction of the fibres [1]. However it is also well known that a bundle of fibres wetted with resin in the form of a composite will support substantially more load than a similar bundle of dry fibres. The increase is not due to the load carrying capability of the resin itself, which is relatively weak, but due to its ability to redistribute load from broken to unbroken fibres [11]. The matrix can only redistribute the load if the interface (or the interphase region) is strong enough to transfer the stress. It is also known that in the case of a strong fibre–matrix bond a failure of the fibre can lead to micro cracking of the matrix at the fibre-break [12,13]. A crack in the matrix will lead to a higher stress concentration in the matrix than a debond [12], and it is thought that stress concentrations at fibre-breaks have an effect on the tensile failure of a composite [14]. A mechanism whereby an excessively strong fibre–matrix bond could reduce tensile strength therefore exists, making it likely that an optimal value of interfacial strength for maximum tensile strength will exist. Furthermore a ductile interphase can also redistribute the stored energy in a fibre which is released on fracture rather than debonding or fracturing the matrix.

2. Experimental

2.1. Materials

E-glass fibres were supplied by Owens Corning Ltd. of diameter 14.32 ± 1.47 μm, density 2.60 g/cm^3, and tensile modulus 76 GPa. The fibres were supplied as water sized; that is no silane or other coupling agents or lubricants had been applied but the fibres were sprayed with water during the manufacturing process.

The monomers, acrylic acid and 1,7-octadiene utilised for plasma deposition were obtained from Aldrich Chemical Company, UK with a purity of 99% or more. Acrylic acid was used to introduce carboxylic acid functional groups into the plasma polymer and 1,7-octadiene was used as a diluent.

The matrix resin used for the experiment was a mixture of 100 parts (by mass) Epikote 828 which is a diglycidyl ether of bisphenol-A (Robnor Resins, UK), 90 parts 1-methyl-5-norbornene-2,3-dicarboxylic anhydride (NMA), a curing agent (Robnor Resins, UK), and 1 part DY062, Benzyldimethylamine (BDMA), used as the accelerator (Huntsman Advanced Materials Ltd., UK). The resin was cured at 80 °C for 2 h, 120 °C for 3 h, 150 °C for 4 h, with ramp rates of 2 °C/min.

2.2. Plasma polymer coating

The fibre bundle was separated by a jet of blown air (to minimise shadowing effects), rolled onto spools and loaded into the plasma reactor. The fibres were then coated by a continuous process by being wound from spool to spool through the plasma chamber. The speed of the receiving spool was adjusted to maintain a constant deposition time of 10 min on the fibre bundle. The baseline pressure within the reactor without monomer flow was less than 3×10^{-3} mbar. The total flow rate of monomers was set to 2 sccm using needle valves. The ratio of acrylic acid: octadiene was varied from 100%:0% to 0%:100% in 20% increments. A low power of 1 W was used to maintain polymer functionality. Fibres used to create control specimens were produced in the same way, but without monomer flow or power.

2.3. Specimen preparation

The coated fibres were hand wound onto frames measuring 20 × 95 × 1 mm. 3.5 m of fibre bundle was used per frame. These were placed in a resin bath and left in a vacuum oven at 80 °C for 1.5 h to degas and to impregnate the fibres. The impregnated fibres remained on the frames and were cured in a vacuum bag within an autoclave at a pressure of 2 bar. The frames were then cut away to leave the composite panels, end tabs were adhered with PTFE inserts to reduce the stress concentration at the end tab and the panel was cut into tensile test specimens with the geometry shown in Fig. 1.

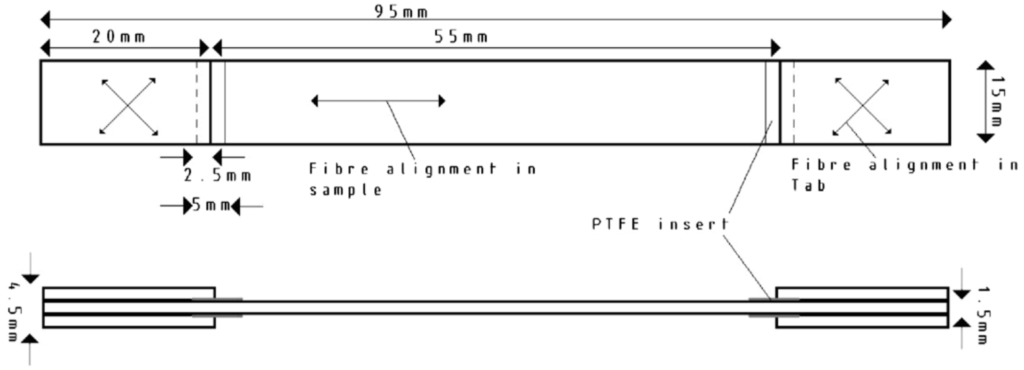

Fig. 1. Tensile test piece geometry.