

Interfacial Debonding in Glass Fiber Reinforced
Polymer Composites

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The durability of polymer composite materials is affected by environmental factors such as temperature, moisture and the state of stress. The interfacial properties are of paramount importance since damage most often initiates and propagates from changes at the fiber-matrix interfaces. The chemical and physical properties of the “interphasial” region between the reinforcing constituent and the polymer matrix can be manipulated to control its chemical stability and mechanical response. Moisture is particularly injurious to the long-term stability of the interfaces in glass fiber reinforced materials. Silane coupling agents are used to chemically bond the reinforcing fibers to the polymeric matrix and to minimize the initiation of fracture at the interface boundary. A direct covalent bonding of a hydrophobic boundary layer to the fiber surface has been shown to maximize the durability of the interface.

In order to produce water impermeable interfaces between glass fibers and Bisphenol-A based matrices, oligomers were grafted at available silanol sites on the glass fiber surfaces by way of a SiCl_4 intermediary [1-3]. The general concept is illustrated in Figure 1.

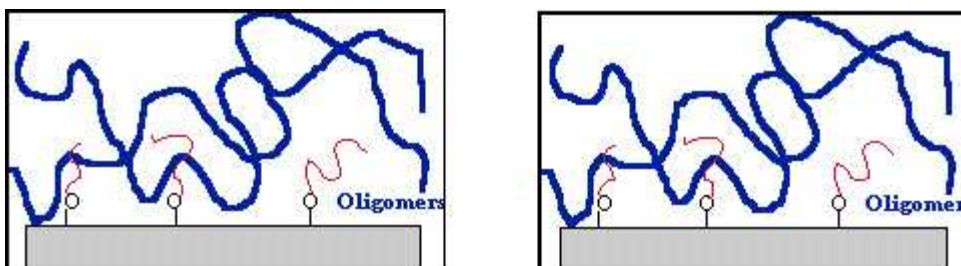


Figure 1

Concept: Create a hydrolytically stable interface with good stress transmission properties by chemical

grafting of molecules on surface.

In polycarbonate composites, the SiCl_4 and oligomeric polycarbonate reacted with hydrogen bonded water and hydroxyl groups at the S-glass interface, and then, during fabrication, the interphase layer transesterified with the polycarbonate matrix. Diffuse reflectance FTIR, GC/MS and TGA measurements were used to show that a tenacious monomolecular layer of grafted polycarbonate (or Bisphenol-A) remained on the fiber surfaces, even after repeated Soxhlet extractions with methylene chloride. Chemical shifts in the high resolution solid state ^{13}C CP/MAS NMR carbon spectra indicate covalent bonding between the grafted monolayers. Measurements of the ^1H spin-lattice relaxation times, $T_{1\rho\text{H}}$, show that the average relaxation times for the grafted monolayers are higher than those for the neat oligomer, and the thermal response shows that the α -transition apparent in the neat oligomer disappears when it is tethered. Examination of all NMR data for the Bisphenol A and oligomer-grafted systems, combined with the evidence of hydrolytic stability, lead us to speculate that the monomolecular layers are covalently tethered to the S-glass surfaces and form a loosely-packed, brush-like layer that shields the S-2 glass surface from diffusing water. Scanning electron photomicrographs of the fracture surfaces of the composites containing surface-grafted fibers indicate strong bonding to the polymer matrix, as illustrated by the presence of strongly adhering polycarbonate on the pulled-out fiber filaments, even after exposure to boiling water for 24 hours. Under the same conditions, debonding of the polymer matrix was evident when commercial sizings were utilized.

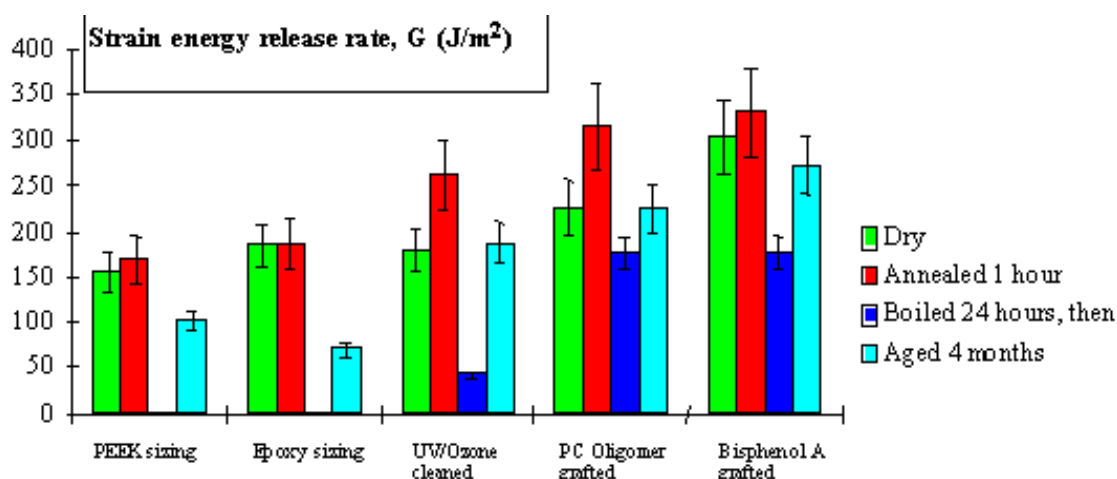


Figure 2.

Fracture Toughness of Fiber Surface Treatments

Micromechanical tests of interfacial strength and toughness were used to evaluate the stability of the interfaces when exposed to boiling water for 24 to 72 hours and the durability of the interfaces when exposed to 37°C water for 4 months. Finite element analysis was used to simulate interfacial debonding and to calculate the strain energy release rate accompanying debonding at a broken filament end [3]. Single fiber fragmentation tests were used to assess the hydrolytic stability and durability [3, 4]. Results were compared to those obtained with composites prepared using fibers treated with commercial sizings

as illustrated in Figure 2. Under all conditions, the debonding energies of the interfaces with monomolecular layers of grafted polycarbonate oligomer or Bisphenol A were higher than those using commercial PEEK or Epoxy sizings. The effects of changing the mechanical properties of the matrix, fiber and fiber sizing on the fracture toughness will also be presented.

References

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