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Feasibility analysis of inter-laminar toughening for improving delamination resistance

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Abstract

This letter discusses the effect of bonding energy on the delamination resistance of laminate composites using a ductile interleaf. The mechanism and methods for interleaf toughening are reviewed and the improvement in delamination resistance discussed. The bonding energy between the ductile interleaf polymer and the fibers as well as between the ductile interleaf polymer and the brittle matrix polymer is shown to be a limiting factor in the interleaf toughening of epoxy based laminates. A dual bonding approach, that is, melt bonding and diffusion bonding, is evaluated as a means to increase the bonding energy between the interleaf and laminate components.

Keywords: Composites; Delamination resistance; Inter-laminar toughening

Polymer matrix composites (PMCs) are the most widely used composite material in industrial applications due to their high strength-to-weight ratio and lowest manufacturing and material costs of any matrix. Laminate PMC fabrication involves the molding of fiber fabrics, either in the form of polymer matrix impregnated tapes (prepreg) or un-impregnated woven fabrics (preform), in successive layers called laminas or plies to form a solid structure. Development of vacuum assisted resin transfer molding (VARTM) of low viscosity thermoset (TS) epoxy matrix materials has led to the application of pre-form fabrics in large composite structures [1].

An important application of glass fiber reinforced (GFR) PMCs is wind turbine blades: over 180 feet in length fabricated from stitched fiber pre-forms and VARTM. Blade max tip deflection, buckling resistance and weight considerations dictate the need for significant tapering of the blade and main spar from hundreds of mm at the root to only a few mm at the tip [2]. Fabrication of tapered wind turbine blades incorporates repeated ply terminations (called ply drop-offs shown in Figure 1a) between the root and tip sections. Ply drop-offs induce shear lag and out-of-plane stress concentrations at the drop-off when the composite is loaded in-plane [3]. In the effort to reduce fabrication costs, ply thickness is scaled up with blade length, leading to thicker drop-offs. Stress concentrations are magnified by the thickness of the dropped ply, the number of plies dropped, and the proximity of ply drops [4]. Delamination initiation at ply drop-offs under critical static and fatigue loading are an ever greater concern in wind turbine blades as the length and thickness of these laminate structures increase [5]. Once initiated, cracks propagate through the brittle matrix material (epoxy) between two plies, resulting in a characteristically brittle delamination fracture surface (Figure 1). Inter-laminar toughening is of critical importance in composite applications with demanding geometric requirements such as tapers, free edges, and holes.

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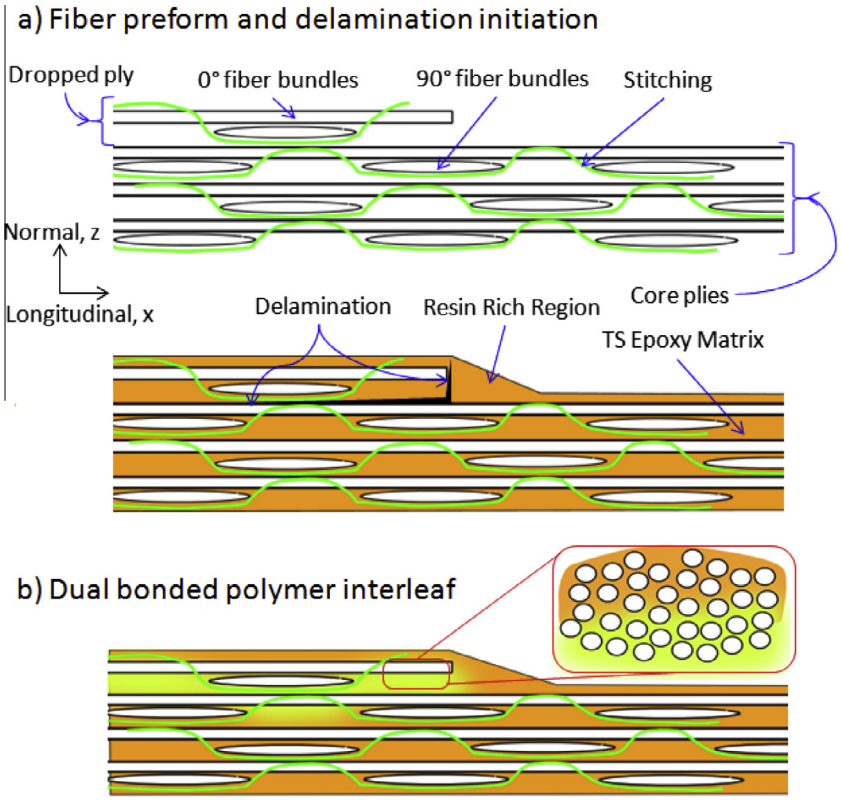

Figure 1. (a) Schematic of the bonded interleaving process: an amorphous TP interleaf is inserted and infiltrated into the fiber bundles through a hot melt bonding process. (b) During elevated curing, TP–TS interdiffusion occurs at the interleaf/resin interface forming a molecular bond between the epoxy and interlayer materials.

1. Materials and methods

Alumino silicate E-glass fibers (10 μm in diameter) are used in this study. E-glass is the most widely used grade of glass fiber reinforcement in PMC applications for its high strength-to-weight ratio, low cost, and high glass transition temperature ($Tg = 846°C$, $\eta = 10^9$ Pa s). Sample specimens are composed of unidirectional stitched fabrics with an areal density of 980–1800 g/m^2. Thermoplastic (TP) ($Tg = 150–250$ °C) thin films (50–500 μm thickness) are used as interleaves to be melt-bonded to E-glass fibers below their softening temperature. Of the available TP types, highly ductile, amorphous, low transition temperature grades containing bisphenol A is desired. TP interleaves will be placed in between preform plies at the desired locations and hot pressed between 200–320 °C and 0.5–4 MPa into the fiber preform architecture prior to VATRM matrix infusion and curing.

The chemical compatibility of the TP interleaf to the TS matrix controls the inter-diffusion depth between the two polymers during curing at elevated temperatures. TP's formed through the polymerization of bisphenol A, such as polysulfone (PSU), contain monomer groups that are soluble in amine, thereby allowing inter-diffusion of the epoxy resin into the interlayer. The final diffusion depth of the TS–TP interface is controlled by the temperature during the curing process and the maximum allowable curing temperature of the resin. Three amorphous TP interleaves will be evaluated in this study. Polycarbonate (PC) has very high plasticity, low glass transition temperature and poor chemical affinity to epoxy. Polysulfone (PSU) has similar plasticity to PC with higher glass transition temperature and good chemical affinity to epoxy. Polyetherimide (PEI) has higher strength than PC or PSU, lower plasticity, higher glass transition temperature and some chemical affinity to epoxy.

2. Theory

Matrix fracture toughness (J_{IC}, J_{IIC}) at the location of stress concentrations determine the delamination initiation behavior and resistance (G_{IC}, G_{IIC}) of the laminate. Once initiated, inter-laminar cracks are detrimental to the life of the composite structure, resulting in bulk delamination and buckling. In addition to ply drop-offs, stress concentrations and delamination initiation sites also include free edges, holes and joints. The creation of tough selective interleaves in these highly stressed regions will significantly improve composite delamination resistance.

The proposed process has its roots in thermoset (TS) matrix modifications and TS interleaving applications first investigated by Aksoy et al. [6]. Adhesive inter-layering and interleaving methods have shown significant improvements in delamination resistance of thermo-set matrix composites

but are limited in toughness by the base thermo-set resin. The delamination resistance limit of TS-adhesive interlayering is characterized by the maximum size of the plasticity zone ahead of a crack tip, depicted in Figure 2. The proposed interleaving process enables the direct replacement of the brittle matrix with a highly ductile thermoplastic polymer, increasing the size of the plasticity zone, and doing so within the widely adopted VARTM framework.

The benefits of selecting a thermoplastic (TP) interlayer are depicted in Figure 2. The theoretical delamination resistance linearly increases with the inter-leaf thickness along the slope given by the total internal energy of the interleaf material: $G_{IC} = t\int_0^\infty \sigma d\varepsilon, G_{IIC} = t\int_0^\infty \tau d\gamma$ [7], where t is the interlayer thickness, and the integrals are the total internal energy of the interlayer. By picking a highly ductile TP interleaf material with a greater total internal energy, a greater delamination resistance per unit interlayer thickness is achievable. Thinner interleaves minimize the reduction in fiber volume fraction and in plane stiffness. By maximizing the bonding energy between the interleaf and the fiber architecture, we expect to exceed the limit of TS adhesive plastic zone size, approaching the plastic zone limit of the base TP. TP interleaving was first investigated by Aksoy et al. [6] but has not been widely adopted due to the limitations in TP-fiber bonding energies. More recent investigations into improving interleaf bonding energies by Hojo et al. [8] has demonstrated the potential in delamination resistance improvements using TP interleaves. Hojo et al. [8] highlighted that the significant improvement in delamination resistance was achieved through a bonding depth of only two fiber diameters, as is limited by the ionomer interleaving process. Through hot melt infiltration and polymer inter-diffusion methods, we expect to improve the bonding energy between the TP and fiber architecture significantly, thereby exceeding the maximum delamination resistance previously achievable through adhesive interlayering methods. The authors have also investigated an inter-laminar reinforcement approach based on direct glass joining [12,13].

3. Results

TP interleaving has been carried out using an amorphous TP and stitched glass fiber preform. Interleaf melt bonded has been carried out on two plies of unidirectional stitched E-glass fabrics (with a glass transition temperature (Tg) of 812 °C) and polycarbonate (Tg = 155 °C), a highly ductile amorphous TP. Cross section imaging of these samples exhibit the desired viscous flow and fiber encapsulation behavior. The hot melt bonding process was carried out on a bench top furnace and press fixture in the range between 220 and 275 °C. The significant difference in softening temperatures between the fiber and interleaf materials allows for a wide processing window for the melt bonding process. As seen from the cross section in Figure 3, the TP interleaf is able to flow into and encapsulated the fibers up to a depth of five fiber diameters. The interleaf is shown to wet and bond to the fibers and the process largely preserved the integrity of volume fraction of the fibers. Using a polymer film with a uniform film thickness, the thickness of the interleaf is well controlled by the temperature and pressure of the bonding process. The two ply pre-form configuration evaluated thus far needs to be extended to multiple plies and ply drop off configurations for delamination resistance and onset testing. Viscous flow of softened TP into and encapsulation of glass fibers need to be further investigated.

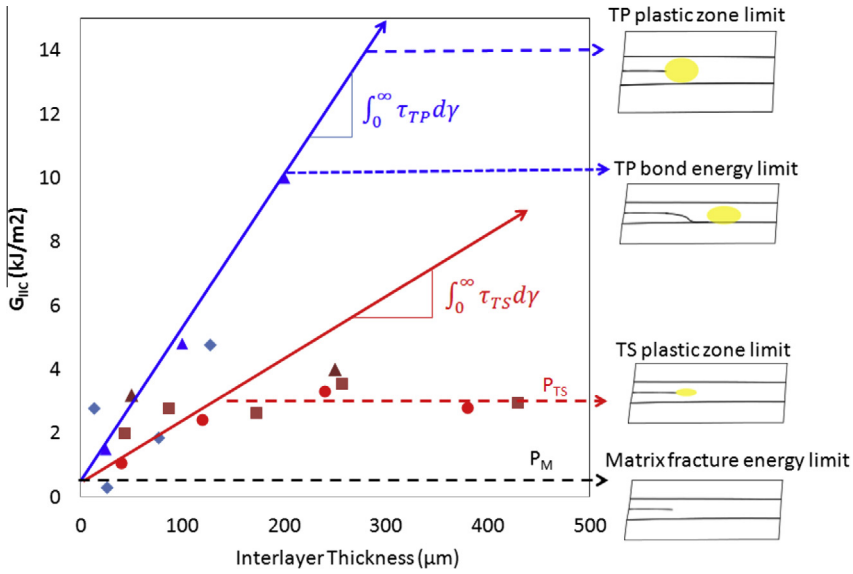

Figure 2. Theoretical and experimental mode II delamination resistance of TS (red) and TP (blue) inter-layered composites. TS inter-layer delamination resistance is limited by the maximum plastic zone ahead of the crack tip while TP inter-layers are limited by TP-fiber bonding. TS data (red) by [6,9,10], TP data (blue) by [6,11]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

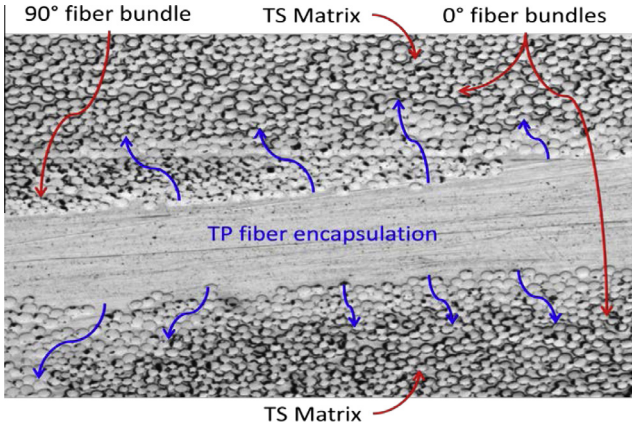

Figure 3. Polycarbonate (PC) thermoplastic infusion and encapsulation of glass fibers prior to VARTM resin infusion. TP infiltration of over five fiber diameters is observed in some places. No inter-diffusion zone is observed for PC-epoxy material pairs.

After hot melt bonding, an epoxy resin is infused into the TP bonded fiber preform and cured using conventional VARTM methods. Preliminary high temperature cure tests using polysulfone (PSU) has yielded observable diffusion depths within the range of curing temperatures used during VARTM curing (Figure 4). A linear trend between curing temperature and diffusion depth is observed using the low viscosity epoxy typically used in liquid infusion processes. Further VARTM curing trials are needed to investigate the effects of fiber confinement along the matrix-interleaf boundary on the diffusion depth and morphology within a melt bonded structure.

4. Discussion

The chemical compatibility of the TP interleaf to the TS matrix controls the inter-diffusion depth between the two polymers during curing at elevated temperatures. TP's formed through the polymerization of bisphenol A, such as polysulfone (PSU), contain monomer groups that are soluble in amine, thereby allowing inter-diffusion of the epoxy resin into the interlayer. The final diffusion depth of the TS-TP interface is controlled by the temperature during the curing process and the maximum allowable curing temperature of the resin. Three amorphous TP interleaves will be evaluated in this study. Polycarbonate (PC) has very high plasticity, low glass transition temperature and poor chemical affinity to epoxy. Polysulfone (PSU) has similar plasticity to PC with higher glass transition temperature and good chemical affinity to epoxy. Polyetherimide (PEI) has higher strength than PC or PSU, lower plasticity, higher glass transition temperature and some chemical affinity to epoxy.

5. Conclusions

A dual bonding approach is evaluated as a means to increase the bonding energy between the interleaf and laminate components to significantly improve delamination resistance. The mechanism and methods for interleaf toughening are reviewed and the improvement in delamination resistance has been discussed. The bonding energy between dissimilar polymer pairs is shown to be a limiting factor in the interleaf toughening of epoxy based laminates

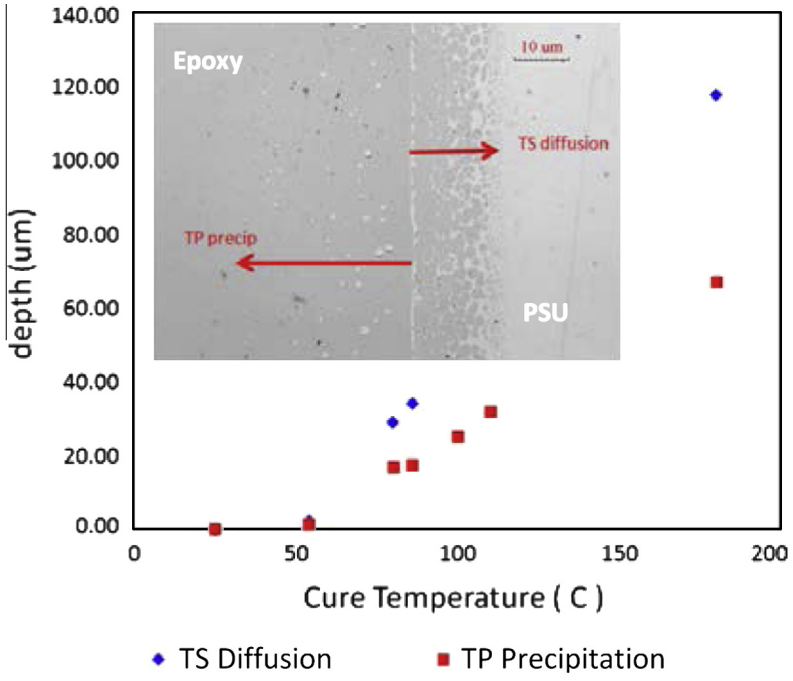

Figure 4. Polysulfone (PSU) thermoplastic diffusion depth into epoxy during elevated curing is measured from optical microscopy imaging of the TP–TS interface. Thermo-set diffusion into the thermoplastic is characterized by a web-like PSU structure. The displaced PSU is precipitated in the thermo-set after curing.

and experimental results in two interlayer bonding methods have been presented. Further work is needed to confirm the effect of interlayer bonding energy on delamination resistance. Rigorous mechanical testing of interleaf toughened laminates is ongoing after VARTM processing to determine the effects of selective toughening on delamination initiation, resistance, and strength. Mode I, mode II and mixed mode double cantilever beam testing are under way to measure the fracture energy of interleaved composites.

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