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MICROCRACKING MECHANISMS AND INTERFACE TOUGHENING OF SEMI-IPN POLYIMIDE MATRIX COMPOSITES

by

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One of the most critical issues for structural durability of fiber-reinforced composites with normally brittle *thermoset resin* matrix has been the occurrence of *localized micro-scale damage* (often called *crazing* or *microcracking*) in the form of either *resin fracture* near the fiber-matrix interface or *fiber-matrix debonding*. Localized micro-damage occurs at a relatively low level of applied strain (less than 0.3 to 0.5% in static tension) under both *mechanical* and *thermal* loading. For the composites with high degree of differential shrinkage of matrix and fibers in cooling, the micro-damage can be induced by *residual stresses* even in the absence of external loading. Once initiated, microscale damage of matrix/interface develops into *matrix-dependent macroscopic failure* modes such as interfiber splitting, constrained ply cracking (first ply failure) and delamination.

As a means of increasing the resistance of fiber composites against *matrix damage accumulation*, particularly *delamination growth*, past empirical efforts of toughening brittle thermoset resins with the inclusion of *elastomer particles* or *ductile thermoplastic domain* were successful to various extents. In fact, the use of elastomer-toughened epoxy resin matrix became an almost standard practice of composite prepreg industry. In the case of brittle thermoset polyimide matrix composites, one promising approach of increasing matrix toughness with minimal change of modulus and temperature resistance has been the addition of secondary domains of ductile thermoplastic polyimide to the base resins, which results in *semi-interpenetrating network* (semi-IPN) type polymer alloys .

In contrast to the success in reducing damage accumulation of composites, toughening of resin matrix was <u>not</u> successful in raising the resistance of composites against *damage initiation* under *cyclic loading*. Fiber-reinforced resin composites in general do not exhibit clear-cut *fatigue endurance limit* (threshold cyclic stress for infinite fatigue life) which indicates the *onset of critical micro-damage or cracks* in other structural materials. However, as Owen pointed out in his earlier work, all S-N curves of composite systems with thermoset resin matrices of varied toughness or flexibility tend to <u>converge</u> at low levels of stress amplitude such as *10^7 cycle fatigue endurance strength*. In other words, the effects of matrix toughening on fatigie lifetime of composites become less and less noticeable in longer term loading. This fact suggests that the *fatigue damage intiation resistance* of composites is not governed by fracture toughness of resin matrix and instead may be more dependent on the *resistance to interfacial debonding*.

In controlling the *resistance to interfacial debonding* of fiber-resin composites, as Plueddemann pointed out, we have to consider the following two requirements of contradictory nature. Firstly, optimum stress transfer between a high modulus fiber reinforcement and a lower modulus resin matrix requires an *interphase region of intermediate modulus* (which leads to so-called *restrained layer theory* in surface science). Secondly, the toughness of composites and the ability of interface region to withstand differential shrinkage between matrix and reinforcement (i.e. residual stresses) require a *flexible interphase region* to relieve local stresses (*deformable layer theory*). The latter requirement was examined in epoxy resin matrix composites by adding flexible or ductile interlayer between the fiber and matrix. Although the approach was shown to increase impact fracture energy of composites, enough data were not available on its effects on fatigue damage resistance of composites

In view of the facts discussed so far, a new research program has been initiated with the 1990 Summer Faculty Research Project (SFRP) as a preliminary phase. The following three objectives are being pursued for the overall program: (a) to elucidate the mechanisms of microcracking for graphite fiber-reinforced semi-IPN polyimide matrix composites under mechanical and thermal cyclic loading, (b) to devise material engineering solutions for possible *improvement of fatigue damage resistance* (or the increase of fatigue endurance strength) of semi-IPN matrix composites by *tailoring of modulus and toughness of fiber-resin interface region*, (c) to assess processing characteristics of the composites and their roles in controlling the resistance of composites to microcracking and the effectiveness of interface toughening.

In the 1990 SFRP, main emphasis was placed upon the initial screening of material systems and optimization of processing conditions for semi-IPN matrix composites with *tailored interface*. As a first set of control material systems to study, the composites were prepared with unsized Celion 6000 graphite fiber reinforcement and the following resin matrices of varied fracture toughness: (a) PMR-15 thermoset polyimide, (b) semi-IPN of PMR-15 thermoset polymide and NR150B2 thermoplastic polyimide in 75/25 ratio, (c) semi-IPN of PMR-15 and NR150B2 in 50/50 ratio. The measurement of rheological behavior of composite prepreg in squeeze flow condition indicated progressive lowering of resin flowability with increasing content of NR150B2 thermoplastic polyimide in semi-IPN. Confirming the rheological property measurement, curing of semi-IPN matrix composites required higher molding pressure to obtain adequate flow.

For the composites with the resin matrix of semi-IPN in 75/25 ratio, interface tailoring was attempted by using graphite fibers coated with the resins of systematically varied *fracture toughness*. Our initial trial was limited to sizing of graphite fibers with a dilute solution of either brittle PMR15 or ductile NR150B2 resin. In both cases, relatively uniform coating of fibers was obtained. The PMR15-sized fibers were B-staged prior to their use in prepreg. In our continuing work, a broad range of interlayer toughness will be achieved by coating the fibers with the reactants of semi-IPN having lower or higher content of thermoplastic constituent in comparison with the composition of surrounding resin matrix.

In pursuing the objectives of overall research program, we intend to define the respective roles and interaction of critical parameters such as *modulus transition* at the fiber-resin interface, *residual stresses* due to cooling, fracture *toughness* of interlayer, *flow* characteristics of resin matrix, and fiber *wetting* behavior. The assessment of damage initiation and accumulation of composites under mechanical cyclic loading will be based on the S-N (strss amplitude vs fatigue lifetime) data, the measurement of dynamic creep rate, and microscopic examination of fracture modes. In thermal cyclic loading, similar approaches will be taken to assess the occurrence and extent of local damage with the data of temperature range vs strength/stiffness retention, thermal strain for full strength retention and thermal strain for full stiffness retention replacing S-N curve, fatigue endurance strength and dynamic creep rate respectively.