INTRODUCTION

Fiber-reinforced organic composites are increasingly being considered for structural applications in aerospace and advanced marine systems. This emphasis is basically due to the very high specific modulus and tensile strength the composite materials have to offer. The thrust behind this trend is the potential for replacing as much metallic components as possible with composite parts such that weight reduction may be achieved resulting in energy saving. In current organic composite systems, thermosetting resins such as epoxy and polyimide are widely used as matrix materials. These resins offer the combined properties of high modulus and creep resistance because of their highly cross-linked structures. However, high cross-linking generally leads to brittle materials that are subject to failure by the growth of internal flaws and progressive crack propagation. The flaws that may be anticipated include internal cracks, surface cuts and microvoids inherently present as a result of the current processing methods employed in fabricating composite parts. The growth of these flaws and the subsequent crack propagation usually cause catastrophic breakdown in structures. The importance of the fracture properties of matrix resins in the performance of composite materials has therefore been realized and emphasized in recent years\textsuperscript{1-4}. In the future, both next generation commercial and military aircraft will demand high strain-to-failure in composites for increasing structural applications such that the projected weight reduction and fuel efficiency may be met. For a reliable design of structures against impact damage and fatigue cracking, a minimum of 2\% strain in the composites will be required to increase the current design allowables of 0.003 inch/inch. Such a requirement
therefore demands improved fracture toughness in matrix resins in addition to high modulus and low creep.

On a scale of increasing fracture toughness, Fig. 1 shows that thermostetting resins are only slightly tougher than inorganic glass, having fracture energies ranging from 80 to 200 J/m². On the other hand, thermoplastic resins such as polymethyl methacrylate are tougher than the thermosets, having fracture energies of the order of 1 kJ/m². New high performance sulfone polymers exhibit even higher toughness with fracture energies of several kilo-Joules per square meter, which represent values not far from that of, say, the 7075-T6 aluminum. The high fracture toughness of the thermoplastics is not unexpected because of the large free volume available in these amorphous materials to absorb the energies associated with crack growth and propagation. However, it is surprising to note that there is a unique class of thermostetting materials having fracture energies matching that of thermoplastic polysulfones, namely the elastomer-modified epoxy resins.

In order to mitigate the brittleness of epoxy materials, many industrial formulators use elastomeric additives in their epoxy formulations in an attempt to create a multi-phase system. This results in a soft rubbery inclusion dispersed in the brittle epoxy matrix, which is widely known by now to enhance the toughness of epoxies. The effect of the particulate elastomeric modifiers on the fracture behavior of epoxy polymers was studied by Sulton and McGarry, Riew et al., and Bascom and Cottington, among others. These investigators have shown that, when an elastomer was copolymerized with the epoxy resin prior to gelation, a uniformly dispersed phase of small rubber particles was formed in situ. These particles have diameters of a few microns or less, and they enhance the toughness of the unmodified epoxy by several orders of magnitude. The toughening mechanisms were only recently understood to involve triaxial dilatation of the rubber particles at the crack tip, particle elongation and plastic flow of the epoxy matrix. However, when such a rubber-modified resin system is used in fiber-reinforced composites, the effects of the added elastomer particles on the mechanical properties of the composite are not clear. In this paper, the balance between matrix fracture behavior and composite mechanical property will be discussed based on the result of an extensive experimental study. Epoxy polymers modified by using carboxy-terminated butadiene acrylonitriles (CTBN) from the Hexcel Corporation (Dublin, CA) were used. These epoxy formulations utilize both a liquid CTBN and a solid rubber in order to achieve a bimodal distribution of particle sizes, which was shown to give even greater fracture toughness in neat resin form than liquid CTBN additives alone. This enhanced resin toughness has also been shown to be transferrable to increase the interlaminar fracture energy of composite laminates. The results that will be reported here include those from shear, tensile, flexural and fatigue tests.