

# Overview

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In May, 1983, NASA-Langley Research Center held a Workshop on Tough Composite Materials which resulted in NASA conference publication CP-2334. The conference reviewed most of the NASA-sponsored research on this subject, holding workshop sessions in three areas: fracture toughness/impact, constituent property-composite property relationships, and matrix synthesis and characterization. The workshop was of great benefit to NASA and its industrial and university peers. It was hoped a second meeting on this topic could be held again in two years.

The ideal forum for such an encore was an ASTM national meeting. Under the perceptive leadership of Chairman W. W. Stinchcomb, ASTM Committee D-30 on High Modulus Fibers and Their Composites invited NASA to cooperate in sponsoring a Symposium on Toughened Composites. It was held in Houston, Texas, 13–15 March 1985.

The purpose of the symposium was to provide a state-of-the-art perspective of the on-going research to develop tougher high performance continuous graphite fiber reinforced composite materials. A second objective was to make the symposium as multidisciplinary as possible from a materials standpoint. To do this, papers were invited and classified in five categories:

- toughened composites: Prospectives From Industry,
- micromechanics,
- interlaminar fracture,
- thermoplastics, and
- thermosets.

Twenty-six papers presented at the symposium are included in this STP which brings together a wide range of disciplines currently involved in the research and development of tough high performance composites.

Research that is focused on some ultimate end use is usually multidisciplinary in nature, for example, the development of composite materials for aerospace applications. What is often missing in the process is the interdisciplinary activity. Researchers from different disciplines need to make a conscious effort to communicate and exchange ideas with each other. Such intercourse is indispensable to the ultimate success of the activity. It also helps one shed the myopic view so easily gained and retained in the practice of highly technical skills. The papers in this volume will challenge the reader to make a conscious effort at this much needed interdisciplinary communication.

The papers in this volume should also provide the reader with some answers, or at least suggest approaches, to the following questions of general import to the development of toughened composite materials.

- What is toughness and how is it measured?
- How much toughness is needed for a particular application?
- What methods, structural, mechanical, and chemical, can be used to optimize toughness and damage tolerance with acceptable property trade-offs?
- What generic research should be pursued to understand the behavior of tough composites, for example, viscoelastic effects?
- What new concepts need to be developed and pursued?
- Do tougher composites possess long-term durability under a real-world environment?
- What correlations are needed between resin properties and composite properties to help guide synthetic efforts on new matrices?
- What part does fiber-resin interfacial adhesion play in controlling composite properties?

Often, an interdisciplinary approach is required to develop solutions to the challenges raised by these questions. For example, toughness is approached by the chemist at the molecular level, by the fracture mechanician at the ply and sublaminate level, and by the structural engineer at the subcomponent and component level. Ultimately, contributions from all three disciplines must be combined to bring about the successful development of a tough, damage tolerant composite structure.

The reader probably will not be completely satisfied with the answers provided herein. Nor should he be! A paper may fall short of his expectations or may not focus on certain aspects of the problem he perceives as important. Hopefully, the results (or lack of them) published in this STP will stimulate the search for more and better solutions and generate the desire and need for improved research cooperation across disciplines. The fruitage from such activities may well require a third multidisciplinary meeting on toughened composites within the next several years.

A summary of the contents of this volume follows.

### **Toughened Composites: Prospectives from Industry**

*Kam and Walker* presented basic guidelines for the selection of a composite material system for application to aircraft structures. Two major selection criteria, structural characteristics and process characteristics, were discussed. *Griffin* concluded, on the basis of a variety of mechanical tests on advanced toughened composites, that increased values of design allowable tensile strength can be easily obtained. However, similar increases in compressive strength allowables cannot be achieved without additional improvements in matrix properties.

## Micromechanics

*Sohi, Hahn, and Williams* investigated the effect of resin modulus and toughness on compressive behavior of unidirectional and quasi-isotropic graphite/epoxy composites. Mechanisms of failure and failure propagation were also studied using undamaged, impact damaged, and open hole specimens. *Hirschbuehler* attempted to correlate mechanical properties of neat resins and their respective composites in order to develop predictive relationships. Resin flexural modulus, resin strain-work-to-failure, as well as composite compressive strength after impact, interlaminar fracture toughness ( $G_{Ic}$ ), short beam shear strength after impact, and open hole compressive strength were studied. *Hunston, Moulton, Johnston, and Bascom* established a relationship between values of neat resin  $G_{Ic}$  and composite interlaminar  $G_{Ic}$  as measured by the double cantilever beam (DCB) specimen. They found that for a variety of reasons tougher resins translate less than 50% of their toughness to the composite, and that many thermoplastic composites exhibit poor interfacial bonding and lower than expected  $G_{Ic}$  values.

*Jordan and Bradley* conducted a detailed in situ scanning electron micrograph (SEM) study of the delamination failure of brittle and rubber toughened epoxy laminates tested under a variety of Mode I/Mode II ratios. The nature and extent of the crack tip deformation/damage zone, and the development of microcrack zones ahead of the crack tip were observed. *Hibbs, Tse, and Bradley* determined the Mode I, Mode II, and mixed mode delamination fracture toughness and controlling micromechanisms of fracture for five graphite-epoxy composites containing a systematic variation in toughness and interfacial bonding. Toughness was highly dependent on interfacial adhesion and mode of loading. In the brittle systems, an increase in the percentage of Mode II loading led to a dramatic increase in delamination toughness through the development of hackles. *Bascom, Boll, Hunston, Fuller, and Phillips* also presented SEM fractography of delaminated surfaces observing features such as fiber pullout, hackle markings, resin fracture, and resin shear yielding. The effects of changing fiber mechanical properties and matrix resin fracture energy on the fractography of delamination were described.

*Schwartz and Hartness* attempted in a rather Edisonian manner to improve interlaminar fracture toughness, 90° tensile strength, and 0° compressive strength by applying model tough polymeric thin coatings on the fiber surface. *Weinberg* used liquid wetting studies on graphite and glass filaments to calculate solid surface energies which, when combined with thermoplastic polymer surface tensions, were used to predict resin wetting of the fiber in terms of work of adhesion. Notch tensile strength of carbon fiber-thermoplastic composites correlated with the predicted work of adhesion. The results appear to confirm the importance of employing wetting studies in interfacial investigations. *Whitney and Drzal* developed a model for predicting the axisymmetric stress distribution around an isolated fiber fragment. This analytical model should be an improvement over existing "shear lag" models for stress analysis of the single-fiber interfacial shear strength test specimen so commonly used today.

### Interlaminar Fracture

*O'Brien, Johnston, Raju, Morris, and Simonds* further refined the edge delamination test for measuring interlaminar fracture toughness of composites by studying Mode I and mixed Mode I/II versions, varying coupon size and matrix resins, and determining the contribution of residual thermal and moisture stresses on strain energy release rates. *Poursartip* addressed the problem of edge delamination growth under fatigue loading for a brittle and a toughened epoxy graphite composite by developing power law correlations for the two materials. His study showed that energy release must be compared to the increasing resistance to further growth caused not only by new delamination but also by associated off-axis matrix cracking. *Adams, Zimmerman, and Odom* also fatigue tested edge delamination specimens for two toughened epoxies, one an interesting interleaf composition, in two different layup orientations at two test frequencies and two load ratios. Importantly, all laminates exhibited a significant decrease in strain energy release rate with increasing cycles to failure.

*Daniel, Shareef, and Aliyu* determined the effects of loading rates on Mode I DCB interlaminar fracture toughness of a graphite elastomer-modified epoxy composite. Crack extension rates up to 21 mm/s (49.6 in./min) were used; over three decades of crack velocity, a 20% decrease in  $G_c$  was observed. Using the end-notched flexure specimen, *Russell and Street* investigated the static and fatigue behavior of delaminations subjected to pure Mode II shear loading. Their results indicated that increasing matrix toughness improves Mode II shear fracture energy less than Mode I tensile fracture energy, and further, tougher systems are more sensitive to Mode II fatigue crack growth than are the brittle systems. *Johnson and Mangalgiri* presented Mode I, Mode II, and mixed mode interlaminar fracture toughness data for seven composites made with brittle and toughened matrix materials. The study showed that brittle fracture is controlled by the  $G_I$  component, while tough resin fracture is controlled by total strain-energy release rate.

### Thermoplastics

*Beever, O'Connor, Ryan, and Lou* discussed the characterization of semicrystalline polyphenylene sulfide (Ryton®-PPS) neat resin moldings and composites. Molding conditions, especially cool down rates and annealing conditions, influence percent crystallinity and crystalline size which directly influence mechanical properties. *Nairn and Zoller* experimentally determined the residual thermal stresses in crystalline and amorphous thermoplastic composites and also predicted their magnitudes from the properties of the matrix. This work emphasizes the need to determine the effects of residual thermal stresses on composite properties. The next two papers dealt with polyetheretherketone (PEEK). *Cebe, Hong, Chung, and Gupta* conducted isothermal and rate-dependent crystallization of PEEK neat resin to achieve films with varying degrees of crystallinity and crystalline morphology. The room temperature mechanical proper-

ties of films varied with crystal size and size distribution but not significantly with degree of crystallinity. Using semicrystalline PEEK APC-2® composites, *Leach, Curtis, and Tamblin* investigated delamination behavior, damage as a result of low energy impact, and post-impact compressive strength.

### Thermosets

*Yee* critically reviewed the reasons for lower-than-expected toughness in composites with toughened matrices. He discussed various mechanisms for toughening thermoset matrices, including use of second phases, strain softening combined with a high degree of orientation hardening, and low temperature relaxation processes. *Garcia, Evans, and Palmer* fabricated a hybrid composite from a 350°F cure epoxy by adding 3 to 15 parts of silicon carbide whiskers. The 90° tensile strengths and strains and tensile edge delamination strain levels were significantly increased while in-plane fiber dominated properties were all significantly reduced as a result of fiber damage incurred during fabrication. This approach appears promising if the fiber damage can be minimized. *Evans and Masters* disclosed the properties of new one-phase 350°F (176.6°C) cure epoxies with vastly improved mechanical, toughness, and damage tolerance properties over standard brittle systems. Novel improved interleaving materials were also described whose properties exceed the ultimate design strain target for post-impact compression. The interleaf concept exemplifies the ability to “engineer” solutions to critical problems if the mechanics of failure are understood. *Boschan, Tajima, Forsberg, Hull, and Harper-Tervet* describe the application of their recently developed computer assisted process models to facilitate fabrication of high quality graphite-epoxy composites by the standard autoclave/vacuum bag technique with minimum rejection rate. The thermal analyzer and chemiviscosity models were used to fabricate successfully 96- and 384-ply brittle 350°F (176.6°C) cure graphite-epoxy composites, respectively. The models have been extended to toughened matrix materials displaying non-Newtonian melt-flow behavior. *Hartness* explored the use of a novel semi-interpenetrating polymer network as a composite matrix by combining a dicyanate thermoset with a copolyester-carbonate thermoplastic. Composites exhibited substantially high interlaminar fracture toughness than brittle epoxies and excellent flexure properties.

*Norman J. Johnston*

NASA-Langley Research Center, Hampton, VA;  
symposium chairman and editor