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Effect of interleaf sequence on impact damage and residual strength in a graphite/epoxy laminate

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The concept of interleaved composites was introduced by American Cyanamid [1]. In this concept a standard prepreg containing 60 vol% fibers is used as a basis, and a discrete layer of very high toughness, very high shear strain resin is added to it. The interleaved system exhibits superior behavior in compression after impact (CAI) compared to the non-interleaved material using the same basic matrix resin. The major disadvantage of this system is a weight penalty. The tough layers usually have lower stiffness and strength which proportionally reduce the stiffness and strength of the laminate, requiring additional graphite piles to maintain design properties [2].

Inspired by the natural laminates e.g., shells and woods with typically periodic layered structures, we arrived at a concept [3] for graphite/thermosetting laminates by periodically interspersing solid thermoplastic thin films of about a few tens of microns between each graphite ply. An interaction between them occurs at elevated temperatures typically during the curing reaction, resulting in a granular phase-inversion structure. Thus, the procedure can be regarded as *ex-situ* compared to the traditional *in-situ* phase separation. For the 16-ply *ex-situ* graphite/epoxy laminates with 15 tough interleaves [4], the CAI value of 345 MPa was far higher than the 267 MPa of the *in-situ* toughened specimen, though their basic compositions were exactly same. It was also proven that the in-plane mechanical properties of the *ex-situ* laminates were mostly retained and the material exhibited a rather balanced specific profile of strength, stiffness, impact resistance and processing abilities [5]. The *ex-situ* concept would be an all-purpose concept for toughening any laminates regardless of the chemistry of matrix resins.

A comparison between the interleaved system with only one discrete tough, thick layer and the periodically interleaved *ex-situ* system leads to the question of how the interleaf sequence affects the impact damage in graphite/epoxy laminates.

The base matrix composition for this study was a combination of standard diglycidal ether of bisphenol-A epoxy (DGEBA, E-54, Wuxi Resin Factory) and tetraglycidyl methylene dianiline epoxy (TGMDA, AG-80, Shanghai Institute of Synthetic Resins), with DDS as the curing agent. They were mixed at a ratio of E-54:AG-80:DDS = 2:3:2 to make the control specimen. The toughening agent was PEK-C, an amorphous polyether ketone with phenolphthalein group, developed in China (Institute of Applied Chemistry, CAS) [6]. Its properties are similar to those of polyetherether ketone (PEEK) in many respects. PEK-C is dissolvable in tetrahydrofuran (THF), and thus, can be sprayed directly onto the individual prepreg surfaces. The PEK-C coating was almost continuous and its thickness was only a few microns. The prepared plies were finally cured according to the conventional specification and there was about 60 vol% T700SC graphite fiber in the resulting laminates.

The impact damage resistance was evaluated by CAI using (QMW) specimens [7] developed at Queen Mary College, Westfield of London. They were 89 mm × 55 mm quasi-isotropic laminates with plies of $[45/0/-45/90]_{2S}$. The impact energy used was 2 J/mm. Boeing CAI specimens were also used in some cases for comparison. Fig. 1 indicates the individual interleaf sequence and the C-scan results after the impact in comparison. For the interleaf sequence, difference in fiber orientation is shown as being 90 ° ply; 0 ° ply; 45 ° ply; and −45 ° ply, respectively. Light (yellow) background represents the matrix, while the interleaved region is dark-colored (red) as). For all the specimens shown in the figure the upper side was impact-loaded.

Fig. 2 summarizes the compression after impact dependence on the interleaf sequence. It is obvious that the CAI value is proportional to the damaged area as the C-scan results indicated.

In the figures, "A" stands for the single-phase epoxy matrix without any toughening agent as control (AG-80/E-54/DDS). As expected, its CAI-value of 180 MPa is the lowest. "B" stands for the symmetrically interleaved system where the 4 PEK-C coatings were located on both side-surfaces sides. This architecture led to a CAI value of about 211 MPa. By altering the symmetric location of the total 8 interlayers from both side-surfaces to one side of the laminate ("C" and "D") the

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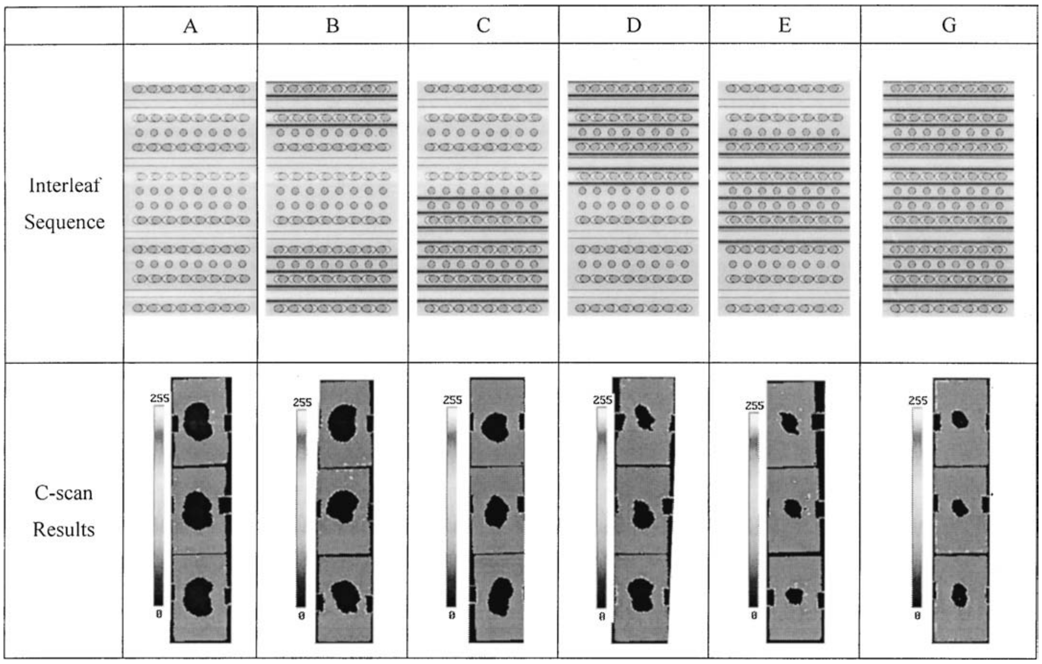


Figure 1 Interleaf sequence of the specimens studied and the C-scan result showing the impact damage.

impact damage changed correspondingly. It was interesting to note that the damage resistance was about 10% higher if the 8 PEK-C interleafs were located on the back side of impact ("D": CAI ≈ 232 MPa, and for "C": CAI ≈ 217 MPa).

Symmetrically interleaving 8 PEK-C layers into the central plies ("E": CAI ≈ 255 MPa) increased further the damage resistance by about 10% compared to the bottom side interleaving ("D"). Increasing the 8 layers to 10 layers ("F": CAI ≈ 271 MPa) made the laminate more resistant against the delamination. Finally, if the laminate was fully interleaved with 16 PEK-C layers ("G"), a higher CAI-value of about 287 MPa was achieved. The highest CAI value of about 345 MPa in this study was achieved for specimen "H", where the PEK-C coating of about a few tens of microns was relatively thicker compared to the "G" specimen. Hence, it is believed that the impact resistance increases proportionally with the volume fraction of toughing agent or the interleaf thickness. If only the interleaf sequence is concerned, the central interleaving (i.e., the case of "E") seems more effective compared to the one-side (cases of "C" and "D"), and its CAI datum is much higher than that of the side-surface sides interleaving (i.e., case of "B").

If we changed the interleaf sequence from strictly per each ply (from "A" to "H" in Figs 1 and 2) to that of every two plies ("1", Fig. 3), the laminate becomes less damage resistant. This behavior may be due to the high volume fraction of graphite fibers and constrained deformation condition in advanced laminates, relatively thin tough interlayers and particularly to the absence of toughening agent within each graphite ply.

Fig. 4 shows representatively the interlaminar morphology of the interleaved system studied. It is, at first glance, nothing new except for the phase-inversed granular structure usually observed for the reaction-induced phase decomposition reaction. However, the granular structure did not penetrate into the graphite plies; thus,

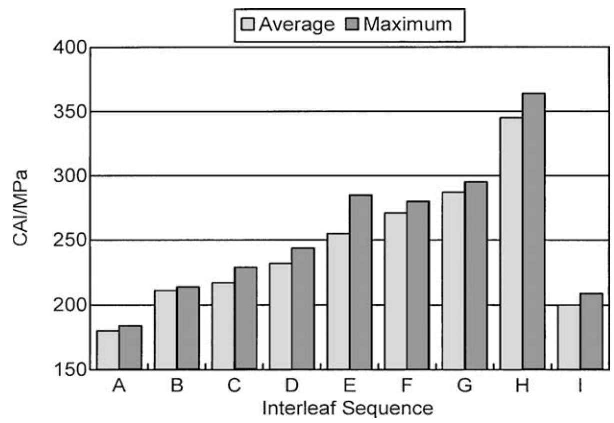


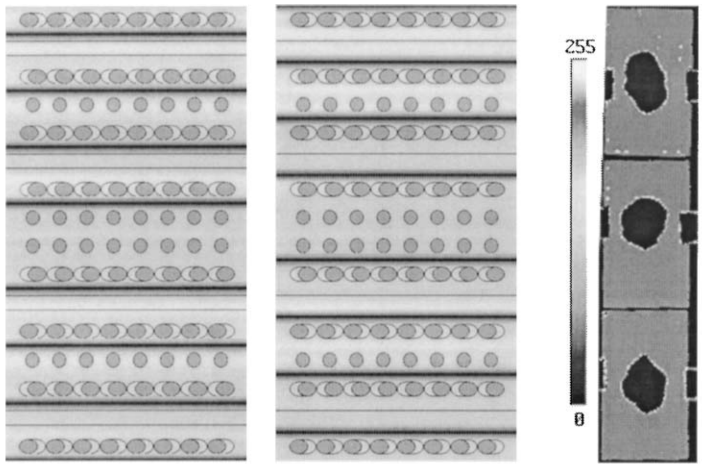
Figure 2 Compression after impact (CAI) of the specimens in dependence on the interleaf sequence.

PEK-C retained high concentration only within each interply layer. It is conceivable that the matrix crack is initiated in the non-toughened surface plies by transverse impact, leading usually to delamination [8]. On the other hand, the thin *ex-situ* interlayers cannot provide sufficient resistance against the delamination, unless there are several strictly alternatively interleaved tough interlayers like the interleaf sequences in cases of "A" to "H" in Figs 1 and 2.

From this experimental investigation, the following conclusions may be drawn:

1. The interleaf sequence affects considerably the impact damage and post-impact compression strength. For this reason, an interleaved system can be designed for desired CAI properties using sufficient toughening agents.

2. The *ex-situ* concept creates a unique laminated structure for advanced composites: graphite/non-toughened epoxy plies and PEK-C modified phase-inversion interply layers. Once the matrix crack is



"I"

Figure 3 Interleaf sequence per every two plies and the corresponding C-scan results.

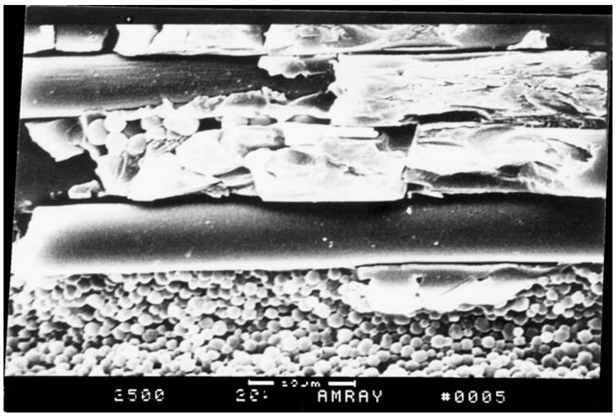


Figure 4 Granular phase-inversion structure of PEK-C modified epoxy in the interply layer and the single-phase epoxy structure within an epoxy/graphite ply.

initiated by transverse impact within the graphite ply, each single toughened thin interlayer cannot provide sufficient resistance against the delamination. For this reason, the system should be designed to be alternatively interleaved throughout several layers, as in this study with 8 layers.

3. It can be deduced that the *ex-situ* concept attributes the in-plane mechanical properties mostly to the graphite/non-toughened epoxy plies and the damage resistance properties mostly to the thermoplastic-rich interlayer. The interaction between them determines the post-impact behavior in the laminates.

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