Hybrid composites based on polyethylene and carbon fibres. Part 3: Impact resistant structural composites through damage management

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Hybrid composites based on polyethylene and carbon fibres  
Part 3: Impact resistant structural composites through damage management  

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Damage tolerance and impact resistance have become key parameters for composite materials in structural applications. In this paper a toughening concept for structural composites based on the hybridization of carbon fibres with high performance polyethylene (HP-PE) fibres is presented. Impact behaviour of hybrid HP-PE/carbon laminates was studied using a falling weight impact test. The effect of the addition of HP-PE fibres as well as the effect of the adhesion level of these fibres on the impact resistance of hybrid HP-PE/carbon structures was investigated. Hybridization results in structural composites exhibiting a significantly better resistance to impact damage than all-carbon laminates due to a change in energy absorption mode. After hybridization more energy is stored in the HP-PE component and consequently less energy is available for damage in the structural carbon component, resulting in a reduction in impact damage and improved post-impact properties.

Key words: composite materials; hybrids; polyethylene fibres; carbon fibres; epoxy resins; impact testing; laminates; residual strength

The use of carbon/epoxy composites as structural materials developed to replace metals has become well established. Composites based on epoxy matrices have several attractive features. These resin systems show a good compatibility with carbon fibres and satisfy matrix-dominated properties such as hot/wet performance. However, the advantages of these materials are significantly reduced because of their susceptibility to damage by low-velocity impacts. To account for impact damage, structures are often 'over-designed' resulting in non-optimum structures in terms of strength/stiffness per unit of weight. The susceptibility of composite materials to foreign object impact damage has been studied rather extensively. Upon impact loadings transverse cracking, delaminations, fibre/matrix debonding and fibre fracture are the potential failure modes in laminated composites. It has been demonstrated that damage due to impact substantially reduces the residual strength after impact of a composite structure, even when damage cannot be visually observed.

The principal mechanism of compressive strength reduction is local buckling of the sub-laminates formed in the delaminated area. In tensile loadings the strength reduction mechanism is dominated by fibre fracture. For these reasons, impact damage is generally recognized as the most severe threat to composite structures.

Several approaches have been taken to improve the penetration resistance and damage tolerance of carbon/epoxy composites. The main developments in toughened composite materials include:

- toughened thermoset matrices—modification is established through the addition of rubber or thermoplastic compounds;
- thermoplastics—an example of a high performance thermoplastic composite system is APC-2 based on a polyether-etherketone (PEEK) matrix reinforced with carbon fibres;
- interleafing—this concept of composite toughening was introduced by American Cyanamid and involves the incorporation of discrete layers of a high shear strain resin at the lamina interfaces thereby giving the composite the ability to undergo higher shear deformations without forming delaminations.

A different toughening approach is to replace brittle
carbon fibres by more ductile fibres such as glass and aramid. A relatively new, tough reinforcing fibre is high performance polyethylene (HP-PE), currently produced based on solution (gel)-spinning of ultra-high molecular weight polyethylene (UHMW-PE), possessing unique mechanical properties in terms of high specific strength and stiffness\(^1\). Moreover these HP-PE fibres possess a high elongation at break leading to high values of work to break compared with other reinforcing fibres such as carbon, aramid and glass. Due to these properties, HP-PE fibres have a high potential for use in composite structures requiring good impact properties\(^1\).\(^2\).\(^3\).

During the mid-1970s various reports concerning impact improvement of carbon-reinforced composites via hybridizing by glass and aramid fibres were presented\(^4\)-\(^6\). Most papers were directed towards enhanced energy absorption capabilities utilizing Charpy-type tests. Although widely used, these tests are not necessarily all suited to an understanding of impact behaviour of composite laminates since the test geometry does not represent the end-use application of the composite structure\(^2\).\(^1\). Instrumented dart or falling weight impact tests on laminated plates have the advantage of a closer approximation to a composite material application.

Recently some studies on hybrid composites were directed to the system carbon/HP-PE\(^2\).\(^2\)-\(^2\).\(^5\). It was shown that impact performance and ductility of carbon/epoxy composites can be increased by hybridizing with HP-PE fibres. Previous studies demonstrated that the amount of improvement was strongly dependent on the adhesion level of the HP-PE fibres\(^2\).\(^4\). \(^2\).\(^5\). Experiments using instrumented dart impact tests on laminated plates also indicated that, apart from the adhesion level, the position of the HP-PE plies in a layered carbon/HP-PE hybrid structure is an important parameter for impact resistance\(^2\).\(^4\). The highest impact energy values for penetration were obtained by positioning the HP-PE plies at the opposite side to the impacted surface in the tensile zone and at a low level of adhesion.

The objective of the present study was to investigate the effect of hybridization with HP-PE on impact damage in the structural carbon component of the hybrid under low-velocity non-penetrating impact conditions including the effect of surface treatment of the HP-PE fibres on the impact toughness of the hybrids.

**EXPERIMENTAL DETAILS**

**Materials**

Hybrid laminates were moulded from woven fabrics of high strength carbon fibre (Grafil XA-S/3K) and HP-PE fibre (Spectra 1000, 650 denier) in an epoxy system of Ciba-Geigy (Araldite LY 556/HY 917/DY 070) based on bisphenol-A with an anhydride curing agent. Carbon and HP-PE fabrics, both of 200 g m\(^{-2}\), were plain weave fabrics with 50% of fibre in the warp and 50% in the weft direction. One ply of HP-PE was approximately twice as thick (0.43 mm) as one ply of carbon fabric (0.20 mm).

Laminated structures were prepared by stacking pre-impregnated plies of carbon and HP-PE fibre woven fabric together in a (0,90) lay-up in a mould to ensure even compaction and curing for 4 h under combined vacuum and pressure conditions in a hot press at 80°C. After curing, samples were post-cured for 12 h at 110°C.

The reference material was a six-ply carbon/epoxy laminate \([C_6]\). A variety of hybrid configurations was prepared based on this basic six-ply carbon laminate to which, on the bottom side, one, two or three plies of HP-PE fabrics were added. These laminates are designated as \([C_6/PE_1]\), \([C_6/PE_2]\) and \([C_6/PE_3]\) respectively. HP-PE fabrics were stacked at the non-impacted surface in the tension zone to obtain an optimum effect in impact improvement. To investigate the effect of improved adhesion of the HP-PE fibres on the impact behaviour of such hybrids, composites incorporating untreated and treated polyethylene fibres were manufactured. Improved adhesion was obtained by immersing HP-PE fibres in chromic acid for 15 min\(^2\).\(^6\)-\(^7\); this proved to be an easy and effective way to increase the interfacial bond strength with epoxy matrices without reducing significantly the tensile strength of the fibre. The total volume fraction of fibre in all laminates was approximately 60%. Plate thicknesses of the various laminates are presented in Table 1. Plane samples of dimensions 100 × 100 mm were cut from these laminates using a diamond cutting wheel.

**Impact testing**

Impact tests were conducted with an instrumented falling weight impact tester. Impact energies were generated by dropping a hemispherical impactor (diameter of 12 mm), loaded with different weights at different heights. Typical incident energies were 2.5, 5, 10 and 15 J. The laminates were clamped between two plates with a square opening of 40 mm. Both force vs. time and impact velocity just before impact were recorded and stored in a digital storage oscilloscope. Postprocessing of data results in the complete energy history during impact and the energy absorbed by the composite. At least four specimens of each laminate configuration were impacted at a fixed energy level.

After impact the specimens were examined for visible damage and indentation depth. In order to determine the impact damage of the carbon component in the hybrid, the outer layers of HP-PE were removed from the bottom surface of the laminate. Ultrasonic C-scan was used to determine the extent of impact-induced delaminations in this carbon component. Optical microscopy on polished longitudinal cross-sections of

<table>
<thead>
<tr>
<th>Lay-up</th>
<th>Average thickness (mm)</th>
<th>Average density (g cm(^{-3}))</th>
</tr>
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<tbody>
<tr>
<td>([C_6])</td>
<td>1.19</td>
<td>1.57</td>
</tr>
<tr>
<td>([C_6/PE_1])</td>
<td>1.57</td>
<td>1.45</td>
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<tr>
<td>([C_6/PE_2])</td>
<td>1.92</td>
<td>1.37</td>
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<tr>
<td>([C_6/PE_3])</td>
<td>2.25</td>
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tested samples was used to reveal the type of impact damage in the carbon laminates and to discriminate between matrix cracking, fibre breakage and delaminations.

Residual flexural strength of the structural carbon part of the hybrid was measured by loading statically to failure on a Zwick testing machine using a three-point bending fixture \((L/d = 40)\) at a rate of 6 mm min\(^{-1}\). Specimens with a width of 35 mm (slightly larger than the width of the largest delamination) and a length of 100 mm were cut from the impacted laminates.

RESULTS AND DISCUSSION

Laminates with untreated HP-PE fibres

1. Impact energies

Postprocessing of the measured impact parameters—force, time and impact velocity—results in the complete energy history and the energy absorbed during impact by the composite laminate. Upon impact the total impact energy can be divided into two parts:

1) the elastically stored energy in the composite plate which is released after maximum deflection by rebouncing of the laminate. This rebouncing energy is successively transferred back to the impactor; and
2) the energy absorbed by the composite laminate which is available for damage and consequently controls the extent of damage and residual strength after impact.

In Fig. 1 the elastic energy \((= \text{impact energy minus absorbed energy})\) is plotted against the impact energy for the all-carbon laminate and three hybrids with, respectively, one, two and three plies of HP-PE fabric added to the non-impacted surface of this carbon laminate. For the \([C_6]\) laminate an apparent maximum in elastic energy storage appears at an impact level of approximately 4 J. At higher impact energies elastic energy storage decreases. Consequently more energy is absorbed in the laminate and is available for damage. The addition of HP-PE results in a change of the energy absorption mode. After hybridization, more energy is stored elastically in the HP-PE fibres and consequently there is an increase in elastic energy transferred back to the impactor. Within the covered impact energy region all hybrids showed a linear increase in elastic energy with increasing impact energy values.

2. Damage characterization

Following the impact tests the laminates were examined for the amount and extent of damage. It can be expected that higher levels of absorbed energy results in more damage.

Fig. 2 shows the visual observations made on the tension side of the carbon laminate after removal of the HP-PE plies. For the basic \([C_6]\) laminate a strong increase is seen in visual tensile face damage with increasing impact energies, resulting in full penetration of the laminate at an impact energy of 15 J. After hybridization a striking reduction in visual damage is observed in the laminate, mainly as a result of the change in energy absorption mode.

The extent of internal damage in the carbon component resulting from the impacts was examined using ultrasonic C-scan imaging. The internal delaminated areas for both the non-hybridized and hybridized carbon laminates are compared in Fig. 3. The overall level of damage in all the carbon laminates increases proportionally with increasing impact energy. However, hybridized laminates showed a smaller level of impact damage at a given incident energy and less sensitivity to higher impact energies as reflected by the decrease in slope of the straight lines with increasing number of HP-PE plies.

In Fig. 1 it was shown that one of the principal toughening mechanisms for HP-PE/carbon hybrid laminates is the improved ability of elastic energy storage. Consequently, less energy is absorbed in the hybrid laminate to be converted into damage. An additional effect which results in a reduction in energy available for damage in the structural carbon component is energy absorption by damage in the HP-PE laminate and at the carbon/HP-PE interface.

By plotting the delaminated area in the carbon component vs. the energy absorbed by the hybrid laminate and assuming that there is a direct correlation between this delaminated area and the absorbed energy in the basic \([C_6]\) laminate*, it is possible to discriminate between energy absorption in the carbon and HP-PE components (Fig. 4). Addition of HP-PE plies causes a shift of the straight lines which suggests that, as expected, energy is also absorbed in the HP-PE component. At a given delaminated area in the carbon component an increase in absorbed energy after hybridization is observed. This increase in absorbed energy can be related to the energy absorbed by damage in the HP-PE component and at the interface between the carbon and HP-PE plies. Since the latter is independent of the number HP-PE plies (there is only one delamination possible between carbon and HP-PE components), additional effects in absorbed energy for

* The validity of this assumption will be discussed below in the section concerning all-carbon laminates.
Fig. 2  Effect of hybridization on visual damage showing carbon tensile face damage after removal of HP-PE plies
Fig. 3 Delaminated area of carbon component vs. impact energy: +, [C₆]; ○, [C₆/PE₁]; △, [C₆/PE₂]; and ◇, [C₆/PE₃]

Impact energy (J)

Elastic energy storage (J)

Fig. 4 Delaminated area of carbon component vs. absorbed energy: +, [C₆]; ○, [C₆/PE₁]; △, [C₆/PE₂]; and ◇, [C₆/PE₃]

Absorbed energy (J)

To illustrate this we will discuss the energy absorption for [C₆] and [C₆/PE₁] laminates at an impact energy of approximately 10 J. From Figs 1 and 3 it can be shown that in the case of the [C₆] laminate this yields a value of 0.6 J for elastic energy storage and 8.8 J for energy absorbed in the laminate, resulting in a carbon damage area of 4.5 cm². With the addition of one ply of HP-PE fabric elastic energy storage increases up to 2.9 J, leaving 6.6 J for energy absorption in the hybrid laminate and causing a delaminated area in the carbon component of 2.8 cm². From Fig. 4 it can be shown that the amount of absorbed energy necessary to create this damage, assuming a direct correlation between delaminated area and absorbed energy, is 4.3 J in a [C₆] laminate. Consequently 2.3 J (= 6.6 - 4.3 J) is absorbed in the HP-PE component and at the carbon/HP-PE interface. In conclusion the total effect of hybridization on the energy absorption mode in a [C₆/PE₁] laminate at an impact energy of approximately 10 J is 2.3 J (= 2.9 - 0.6 J) of additional energy storage and 2.3 J absorbed energy by damage in the HP-PE ply and at the carbon/HP-PE interface. This leaves 4.9 J absorbed by the carbon laminate and available for damage in the structural part of the hybrid.

Micrographs of cross-sections of hybridized carbon laminates subjected to an impact energy of 10 J are shown in Fig. 5(a)-(d). For comparison a non-hybridized [C₆] laminate subjected to an impact of 7.5 J is shown because damage in the 10 J impacted specimens was too severe, preventing a clear overall picture of the damage. Although the [C₆] laminate shown was impacted at a lower energy level than the hybridized laminates, the strong reduction in extent of damage is clearly visualized. At an impact of 7.5 J there is growth of large fibre cracks under the point of impact as a result of the flexural response of the laminate. Failure is initiated at the bottom side of the laminate by tensile stresses exceeding the failure stress of the composite and resulting in the ejection of a shear plug. After hybridization the amount of delamination, fibre and matrix cracking is greatly reduced so that, for the carbon component of the [C₆/PE₁] laminate, damage is more or less the combination of one strongly reduced delamination and one single transverse crack.

Impact damage significantly reduced the residual flexural strength of both non-hybridized and hybridized carbon laminates (Fig. 6). In all cases the curves exhibit basically the same shape. However, the reduction in strength is more pronounced in the non-hybridized case. The flexural strength of the [C₆] laminates after, for example, an impact of 10 J was reduced to 30% of the undamaged value compared with approximately 40%, 50% and 60% of the undamaged values for hybrids with one, two and three plies of HP-PE respectively. The reason for this improved residual strength of the carbon component, by as much as 100% for the [C₆/PE₁] laminate, is undoubtedly the reduction in delamination and fibre fracture. Particularly the latter will have a strong effect on residual strength as tested here in a three-point bending mode.

Laminates with treated HP-PE fibres

Fig. 7 shows the elastic energy vs. impact energy for the hybrids incorporating chromic-acid treated HP-PE fibres with an improved level of adhesion. In accordance with the data shown for untreated HP-PE there is also an increase in elastic energy storage with the addition of treated HP-PE plies. However, in contrast to untreated HP-PE, a maximum in elastic energy storage ability occurs within the impact energy range tested, revealing the embrittlement of HP-PE composites with increasing levels of adhesion. This more brittle failure behaviour results in the fracture of HP-PE fibres at higher impact energies and consequently a loss of impact protection by these fibres (Fig. 8). This implies that above the
Fig. 5 Micrographs of cross-sections of impacted laminates showing damage in the carbon component: (a) [C₆], 7.5 J; (b) [C₆/PE₁], 10 J; (c) [C₆/PE₂], 10 J; and (d) [C₆/PE₃], 10 J.

impact energies corresponding to these maxima, more energy is absorbed within the carbon laminates.

From the C-scan data it could not be concluded that there was a significant effect of fibre treatment on the delaminated area in the carbon component as measured after removal of the treated HP-PE plies. Within experimental scatter all hybrids incorporating treated HP-PE showed similar trends to those shown in Fig. 3.

The effect of surface treatment of HP-PE fibres on the residual flexural strength of the carbon component is shown in Fig. 9. In this typical case the effect is shown for the [C₆/PE₁] and [C₆/PE₃] laminates. As expected, residual strength decreases with increasing impact energy. However, it is demonstrated that the hybridized carbon laminates incorporating treated HP-PE have a substantial higher reduction in strength at higher impact levels than the laminates with untreated HP-PE fibres. As the origin of this more pronounced reduction in residual flexural strength cannot be explained by the
Fig. 8 Tensile face damage in a $[C_s/PE_t]$ laminate incorporating treated HP-PE showing fibre fracture in the HP-PE ply after an impact of 15 J.

Fig. 9 Residual flexural strength of carbon component vs. impact energy—effect of HP-PE fibre surface treatment: +, $[C_s]$; $[C_s/PE_t]$; $[C_s/PE_t]$ treated; $[C_s/PE_t]$ treated; $[C_s/PE_t]$ treated.

Effect of fibre treatment on the delaminated area in the carbon component, other strength reducing factors must be the origin of these additional reductions in strength.

By plotting the residual strength of all the impacted carbon laminates, hybridized as well as non-hybridized, against delaminated area it can be demonstrated that, for a given delaminated area, the residual strength of the carbon laminate hybridized with untreated HP-PE fibres is superior to that of laminates incorporating treated fibres (Fig. 10). This indicates that in the case of treated fibres the damage in the carbon component is more localized around the impact centre and more harmful to the flexural load-carrying capacity of the composite than in the case of laminates incorporating untreated HP-PE fibres. This more intensified damage in the carbon component can be the result of more delamination, debonding, matrix cracking and fibre fracture of which, as already stated, fibre fracture will have a predominant role in flexural strength reduction.

This intensified localized damage cannot be revealed using C-scan because this technique is only capable of giving an integrated picture of the overall damage. Fig. 11 compares the indentation depth of the carbon component of hybrid laminates incorporating treated and untreated HP-PE fibres with increasing impact energies. It can be observed that at higher impact energies the indentation depth is much greater for hybrid laminates reinforced with treated HP-PE fibres than for the hybrids with untreated fibres as a result of fibre fracture in the HP-PE plies. As indentation in the case of a brittle material such as carbon/epoxy always involves fibre fracture, it supports the earlier assumption that hybrids with treated HP-PE fibres have a more intensified localized damage area in the carbon laminate with more severe fibre fracture around the impact centre.

All-carbon laminates

Besides a change in energy absorption mode there is an additional effect of increased laminate stiffness on the
The extent of damage in the composites with the addition of HP-PE plies. After hybridization the increase in stiffness of the laminate results in smaller deflections and consequently in lower flexural stresses. These stresses cause fibre and matrix fracture in the lower outer ply. This crack deflects when encountering the next ply, where it forms a plane of delamination. Cantwell and Morton showed that laminate stiffness is a dominant parameter in impact response, thereby controlling the mode of fracture. Impact damage in thin flexible laminates initiates due to flexural action of the laminate, whereas in thicker stiffer laminates damage is initiated by contact stresses concentrated around the impact centre.

In order to ensure that the observed improved toughness was not simply due to the effect of increased thickness of the hybrid laminates, all-carbon laminates were fabricated containing 8, 10 and 12 plies of carbon fabric, corresponding to the thickness of the hybrids with one, two and three plies, respectively, of HP-PE fabric added. The effect of the addition of carbon plies to the basic $[C_6]$ laminate on the delaminated area in these composite laminates is given in Fig. 12. The slope of these straight lines can be considered as a standard for impact toughness. Consequently it may be concluded that the susceptibility to impact damage of carbon/epoxy composites is not altered with increasing thickness. This in contrast to the hybrid laminates which showed a decreasing slope with increasing amount of HP-PE, resulting in a far more effective reduction in impact susceptibility. This superiority becomes even more pronounced if the weight reduction is taken into account. The hybrid $[C_6/PE_a]$, for example, is more than 20% lighter than the $[C_{12}]$ laminate at approximately the same thickness.

By plotting the delaminated area of the all-carbon laminates against the absorbed energy it can be shown that all the data fall on the same straight line (Fig. 13). This supports our earlier assumption, made for discriminating between energy absorption in carbon and HP-PE components, that the delaminated area in the carbon part of the hybrid can be directly correlated to the absorbed energy in this component.

**CONCLUSIONS**

It has been shown that the addition of HP-PE plies to the non-impact side of a carbon laminate results in structural hybrid composites exhibiting a significantly better resistance to impact damage by altering the energy absorption mode via hybridization. With the addition of HP-PE, more energy is stored elastically in the HP-PE fibres and consequently less energy is available for damage in the structural carbon component of the hybrid. An additional effect which leads to a reduction in energy absorbed by the carbon component is energy absorption by damage in the HP-PE laminate and by delamination at the carbon/HP-PE interface. The increase in damage tolerance is reflected by a significant reduction in delaminated area of the carbon laminate after hybridization as indicated by the C-scan data. Microscopic investigations of sectioned specimens also indicated that the number of delaminations and extent of fibre/matrix fracture was also reduced in the hybridized carbon laminates. As a result of this an improvement in residual flexural strength after impact is obtained.

Hybrids incorporating surface-treated HP-PE fibres also showed improved damage tolerance. However, at high impact levels fibre fracture occurs in the HP-PE plies due to a more brittle failure behaviour of HP-PE composite with increasing levels of adhesion. After fracture of the HP-PE fibres the elastic energy storage capability in this component diminishes, resulting in more extensive localized carbon damage in the vicinity of the impact centre. Consequently the residual strength of these hybridized carbon laminates impacted at higher energy levels is lower than that of hybridized carbon laminates incorporating untreated fibres, which did not show any fibre fracture in the HP-PE plies within the tested impact energy range.

It was also found that hybridization with HP-PE fibres resulted in composites which were far more superior in
impact resistance than thicker and heavier laminates constructed entirely of carbon/epoxy.

Finally, it can be concluded that the proposed hybrid toughening concept results in composite laminates with no penalty from the standpoint of structural applicability and with superior damage tolerance and penetration resistance compared with that of non-hybridized carbon/epoxy laminates.

Additional benefits of this toughening concept are the possibilities of selective toughening, i.e., application of HP-PE plies at critical locations in a composite structure with respect to impact damage, and impact modification of existing composite structures. At the same time it does not exclude the possibility of composite toughening, such as toughened matrices and interleafing which should result in composites with more superior impact resistance than those presented in this paper. Additional benefits of this toughening concept are the possibilities of selective toughening, i.e., application of HP-PE plies at critical locations in a composite structure with respect to impact damage, and impact modification of existing composite structures. At the same time it does not exclude the possibility of composite toughening such as toughened matrices and interleafing which should result in composites with even more superior impact resistance than those presented in this paper.

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