SELF-HEALING FUNCTIONALITY FOR CFRP

Mr. G.J. Williams, Dr. R.S. Trask and Dr. I.P. Bond

ACCIS - Advanced Composites Centre for Innovation and Science, 
University of Bristol, Department of Aerospace Engineering, Queen’s Building, University Walk, Bristol. BS8 1TR. UK
Tel: +44 (0) 117 331 7499
Fax: +44 (0) 117 927 2771
E-mail: G.J.Williams@bristol.ac.uk
www.aer.bris.ac.uk/research/fibres

Self-healing is receiving an increasing amount of research worldwide as a means of autonomously addressing damage in fibre reinforced polymer composite materials. This paper describes the results of four point bend flexural testing (ASTM-D6272-02 [26]) of T300/914 epoxy Carbon fibre reinforced plastic (CFRP) with embedded resin filled hollow glass fibres (HGF) which provide a self-healing functionality. The study investigated the effect of the embedded HGF on the host CFRP and also the healing potential of the laminates after they were subjected to quasi-static impact damage. Specimens were tested undamaged, damaged and healed using a commercial two-part epoxy healing agent. Microscopic characterisation of the HGF embedment was also used to understand the effect on the host laminate fibre architecture. Double Cantilever Beam testing combined with microscope fractography were also used to investigate the interaction between HGF and a crack front propagating in Mode I.

1 Introduction

The development of high performance fibre reinforced polymer’s (FRP’s) to achieve performance improvements in engineering structures focuses on the exploitation of the excellent specific strength and specific stiffness that they offer. Recently, research has focused on taking advantage of their hierarchical microstructure to facilitate the incorporation of additional functionality giving further benefit to their use over metallic rivals. However, this planar nature of an FRP’s microstructure results in relatively poor performance under impact loading. Furthermore, significant degradation in material performance can be experienced with minimal visual indication of damage being present, a design scenario termed Barely Visible Impact Damage (BVID). Current damage tolerant design philosophies incorporate large margins to account for reduction in structural performance due to impact events, resulting in overweight and inefficient structures. This impact damage sensitivity can be mitigated by imparting an ability for the material to undergo self-healing thereby reducing the importance of detecting and alleviating damage. Self-healing composites would allow lighter, more efficient structures which would have a significant influence in addressing environmental concerns regarding aviation and would also offer a potentially substantive reduction in maintenance and inspection schedules and their associated costs.

Self-healing takes inspiration from biological repair systems, and currently assumes a much simplified approach. To date, applications for different materials have been considered by various authors, e.g. bulk concrete [1-3], bulk polymers [4,5] and polymer composites [6-9].
In particular, self-healing for polymer composites has seen significant developments in recent years [10-21] using the inspiration of biological self-healing applied with broadly traditional engineering approaches. Self-healing in polymer composites can be divided into two main approaches: 1) modification of the matrix material to contain a healing agent or facilitate healing [4,8,10], and 2) inclusion of storage vessels containing a healing agent within the material [11-21]. This paper considers the latter.

HGF [22,23] are used in this study in preference to embedded microcapsules [24,25] because they offer the advantage of being able to store a significant volume of functional agents for self-repair, as well as allowing easy integration with the surrounding reinforcement.

A bespoke HGF making facility [22,23] has been used to produce fibres between 30 and 100 μm diameter with a hollowness of approximately 50%. These are embedded within either glass fibre-reinforced plastic (GFRP) or carbon fibre-reinforced plastic (CFRP) and infused with uncured resin to impart a self-healing functionality to a laminate. During a damage event some of these resin filled fibres fracture, releasing the stored healing agent into a damage site, thus initiating the recovery of properties (healing). This ameliorates critical effects of matrix cracking and delamination and most importantly prevents further damage propagation. The precise details of the healing agent can take several forms whereby a one-part system, a two-part resin and hardener system, or a resin system with a catalyst or hardener could be contained within the HGF [9].

The exact nature of the self-healing method to be deployed depends upon (i) the nature and location of the damage, (ii) the choice of repair resin, and (iii) the influence of the operational environment. The HGFs can be introduced within a laminate as additional plies [17-19] at each interface, at damage critical interfaces or as individual filaments spaced at predetermined distances within each ply [20,21].

This paper considers self-healing CFRP, and demonstrates the strength recovery possible when the HGF system is distributed along interfaces within a laminate, minimizing the reduction in mechanical properties whilst maximizing the efficiency of the healing event. Furthermore, in order to fully understand the mechanism by which the HGF interact with propagating cracks during a damage event, double cantilever beam (DCB) testing was selected to assess the interaction between a propagating crack, the host laminate and the embedded HGF during mode I fracture.

2 Specimen manufacture

2.1 Four point bend flexure

The HGF self-healing studies at Bristol to date [17-21] have incorporated HGF within GFRP laminates as discrete plies. It was decided that this approach would not be suitable for CFRP laminates as it would effectively produce a hybrid glass-carbon laminate and result in a significant reduction in mechanical properties. A less detrimental method is to directly embed a small number of discrete HGF’s within a CFRP ply where they act solely as distributed storage vessels for the healing agent. This method requires optimisation of the HGF distribution in order to limit the detrimental affects on the mechanical performance of the laminate and to maximize the volume of healing agent available to address any damage.
The optimisation was assessed with the use of four point bend flexural testing (according to ASTM-D6272-02 [29]) as it had already been used successfully to demonstrate self-healing in GFRP.

Carbon fibre/epoxy (T300/914 Hexcel Composites) pre-impregnated tape was selected as the host laminate as it is typically used for aerospace applications. Quasi-isotropic (QI) plates were manufactured [16 ply (-45°/90°/45°/0°)\textsubscript{2s}] using a hand lay-up technique (230mm x 160mm x 2.5mm). Cure was undertaken according to manufacturer’s recommendations. HGF (60\(\mu\)m external diameter, 42\(\mu\)m internal diameter, 50% hollowness fraction) with a pitch spacing of 70\(\mu\)m were wound directly onto CFRP plies prior to lamination, located at two 0°-45° interfaces within the lay-up as follows:

\((-45°/90°/45°/0°/\text{HGF}/-45°/90°/45°/0°/0°/45°/90°/-45°/\text{HGF}/0°/45°/90°/-45°)\)

### 2.2 Double cantilever beam

Two configurations of CFRP laminate were assessed. Both were fabricated from 32ply IM7/8552 pre-impregnated CFRP tape (Hexcel Composites) in which HGF spaced at 70\(\mu\)m were aligned in the 0° fibre direction on the mid-plane. The first considered only unidirectional reinforcement with the fibre direction parallel to the direction of crack propagation.

The second configuration consisted of the following lay-up [(0\(_2\),90\(_2\))\textsubscript{S},(0\(_2\),90\(_2\))\textsubscript{S} HGF (-45\(_2\),45\(_2\))\textsubscript{S} (45\(_2\),-45\(_2\))\textsubscript{S}]. The purpose of this latter configuration was to investigate local fibre orientation on crack propagation, and in particular the 0°/45° interface that could delaminate within a QI laminate as considered in the flexural testing work detailed above. The 0° direction for this case was perpendicular to the direction of crack propagation.

### 3 Mechanical testing

#### 3.1 Four point bend flexure

A support span to depth ratio of 32:1 and a support to load span ratio of 3:1 were selected in accordance with ASTM D6272-02 [29]. This resulted in specimen dimensions of 100mm x 20mm x 2.5mm. Ten samples were cut from a plate with the use of a water-cooled diamond grit saw. The sample edges were smoothed (SiC P2500) to avoid any unwanted edge effects. Samples were then dried, sealed in sample bags and stored in a temperature and humidity controlled environment prior to testing.

Immediately prior to testing, the HGF within each specimen were infiltrated with pre-mixed two-part epoxy resin (Cycom 823 Cytec) using a vacuum assist technique. Quasi-static impact damage was imparted to each specimen using a 5mm spherical tup mounted on a Hounsfield H20K-W electromechanical test machine running in load control with the sample supported by a steel ring of 27mm outer diameter and 14mm inner diameter. The indentations were stopped at a peak load of 2000N. Up to this point the damage is contained within the laminate and can be likened to BVID, as the impact surface suffers a minimal indent and the back face experiences minimal distortion due to back face delamination.
After indentation, the specimens were left at 70°C for 45 mins to facilitate resin infiltration at minimum viscosity (25cps) followed by 125°C for 75 mins which initiates cure. The use of a pre-mixed healing resin and elevated temperature after the damage event was an attempt to achieve the maximum healing effect (and thus a baseline of maximum performance) from the Cycom 823 resin.

This resin system is not designed for use as a healing agent, in fact, no resin systems currently exist which are specifically formulated for such an application. Some desirable characteristics for an ‘ideal’ healing resin have been identified;

- Low viscosity to aid flow into damage site
- Insensitivity to precise stoichiometric mix ratio
- Ambient temperature cure
- Longevity in uncured state.

However, the advantage of using Cycom 823 at this time derive from the fact that temperature activation provides excellent control of the cure initiation, eliminating time constraints on the testing/manufacturing process.

After healing, test specimens were mounted on a Roell Amsler HCT25 electromechanical test machine with roller spacing determined by the specimen dimensions (ASTM D6272-02 [29]). An Instron 8800 controller/data-logger was used to control the test machine and record data. Specimens were loaded to catastrophic failure at which point the cross-head displacement was stopped and the load removed. Specimens were monitored to ensure a consistent failure mode and optical microscopy used to record detailed observations. Results were obtained from 10 undamaged, 5 damaged and 5 healed specimens.

### 3.2 Double cantilever beam

Damage initiation and propagation caused by impact events can be assumed to result from aspects of Mode I, Mode II and Mode III loading of varying contributions depending on the location of the damage within the laminate, loading conditions, support conditions material properties [26-28]. Double cantilever beam (DCB) testing is thus appropriate for and can be used to investigate the interaction between mode I crack propagation and HGF located along a ply interface, in accordance with ASTM D 5528 -94a [30]. This testing, whilst not fully representative of what takes place in a real impact event, provides a simple and effective way to recreate one of the crack propagating mechanisms involved. Further work is underway to determine the effects of mode II and mixed mode loading on crack interaction with embedded HGF.

A pre-crack was created within the specimens by use of PTFE inserts (15μm). Piano hinges were bonded to the load ends with the use of Araldite 2010 after lightly abrading the surface with SiC P120 abrasive paper.

A non-standard crosshead displacement of 50mm/min was selected for testing in an attempt to recreate loading rates more akin to impact damage. A more comprehensive range of loading rates will be assessed in further work.
4  Results

4.1  Four point bend flexure

Analysis of the flexural test data in Table 1 shows that fibres spaced at 70μm caused a significant reduction in the host laminate’s undamaged strength (8%). This can be attributed to the disruption in fibre architecture observed in Figure 1. However, after a quasi-static impact event of peak load 2000N, this configuration also exhibited a large amount of damage tolerance when compared to the plain host laminate. This can be attributed to energy absorbed by the crushing of HGF. It appears that the initial reduction in strength due to the presence of a significant volume of HGF is offset by the increased damage tolerance during an impact event. The relatively large fibre volume fraction of HGF (~3%) in this configuration also provides a significant reservoir of healing agent. This is confirmed by a 97% recovery in undamaged strength (equivalent to 89% of the host laminate undamaged strength).

Table 1: Assessment of Flexural Strength for Undamaged, Damaged and Healed Laminates with HGF spaced at 70μm

![Table 1](image1)

Figure 1: Optical micrograph images of disruption to host laminate architecture due to embedded HGF spaced at 70μm
5 Fractography

5.1 Four point bend flexure

It is known that the strain to failure ($\varepsilon_f$) of commercial glass reinforcing fibre is higher than carbon which suggests that fracture of the carbon should precede that of HGF fracture. However, the $\varepsilon_f$ of HGF is likely to be significantly lower than commercial glass fibre due to a number of factors:

- Larger volume/unit length (i.e. 60$\mu$m vs. ~10$\mu$m),
- Significantly greater surface area (inner plus outer)/unit length (1000%) giving rise to increased surface defects/weaknesses,
- Thermal residual stresses created during the manufacturing process.

Furthermore, there may be more effects contributing to HGF fracture during an impact event such as propagation of stress waves and interaction with propagating crack fronts during shear cracks and delaminations.

SEM micrography has been used to identify HGF fracture after a drop weight impact event. Figure 2 provides examples of a propagating crack fracturing around a fibre/matrix interface (Figure 2a) and directly through a single HGF (Figure 2b). It also shows a crack diverging into multiple cracks as it intercepts a cluster of HGF (Figure 2c) whereby one fibre is fractured through its centre whilst another exhibits interfacial failure at the fibre edge. These images highlight the complexity of the crack paths and give clear examples of HGF failure as damage propagates through a laminate due to low velocity impact.

![Figure 2: SEM micrographs of impact damage in a 16ply Q1 CFRP. Propagating cracks are observed a) edge b) centre and c) within a cluster of HGF](image)

5.2 Double cantilever beam

It can be seen from Figure 3 that mode I crack propagation can initiate HGF fracture at the 0°/45° interface. Although the crack path was seen to deviate from the mid-plane, HGF rupture was evident before deviation occurred. However, was not the case for the unidirectional configuration (Figure 4) which provided little evidence of HGF damage. This suggests that the ability for self-healing to occur (via HGF fracture and the release of healing resin) will be fibre orientation dependent.
When designing a self-healing laminate approach, care must be taken in locating HGF at the interfaces most likely to be susceptible to damage [19] and at the interfaces where crack propagation is likely to rupture the fibres.

Figure 3: SEM micrographs of fractured HGF in DCB sample at 0°/45° ply interface

Figure 4: SEM micrographs of undamaged HGF in DCB sample at 0°/0° ply interface

6 Conclusions

Self-healing of a quasi-isotropic 16 ply T300/914 CFRP laminate has been achieved with the use of resin filled 60μm HGF with 70mm spacing, embedded along two 0°/45° interfaces. The use of a temperature activated pre-mixed resin system (Cycom 823) to provide fully optimised healing has demonstrated a 97% recovery in flexural strength.

Fracture of HGF has been observed to precede failure of the surrounding reinforcing carbon fibre under quasi-static impact events. DCB testing was used to further investigate this phenomenon, with visual evidence acquired, showing HGF fracture on a 0°/45° interface under mode I crack propagation.

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