Development of multi-functional, high performance, pseudo-ductile hybrid composites

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1. Introduction

The failure of conventional structural materials (steel and other metals, alloys) has a favourable ductile character. High performance fibre reinforced polymer matrix composites (e.g. carbon fibre-reinforced epoxy) have been in the spotlight for the last few decades because of their outstanding specific strength (strength/density ratio), but their fundamental limitation is sudden brittle failure, which renders them unfavourable for a range of high volume, safety-critical applications where the loads are not entirely predictable and abrupt failure is not tolerated. Composites only withstand a limited strain before the usually catastrophic failure, and they do not provide a detectable sign before the final rupture. Ductile or pseudo-ductile behaviour and overload indication functionality could enable safer operation and the penetration of composite components into high-volume applications (e.g. transportation and construction industries). Damage and impact resistance can help exploit the full potential of this unique material family. A further drawback of thermoset matrix composites against metals is that there is no available simple repair technology like welding of metals, which is capable of extending damaged components' lifetime. Significant cost and environmental footprint could be saved if minor defects of valuable composite components could be repaired effectively; therefore, repairability is considered as a valuable added function.

The overall aim of the research programme was to develop pseudo-ductile composites further and add valuable functions like reparability and damage indication features to them.

More specific key challenges of the research programme are summarised below:

- Proving our strain overload indicating concept on the component level and making it an additional function to pseudo ductile hybrid composites
- Development of pseudo-ductile hybrid composites based on standard thickness pre-impregnated fibre (prepreg) plies to improve performance and reduce cost
- Development of repairable pseudo-ductile composites
- Out-of-plane damage tolerance characterisation of composites containing interface modification

2. Results

2.1. Development of self-indicating pseudo-ductile composite component

Design, manufacturing and structural testing of a pseudo-ductile composite sandwich panel with an additional visual overload indicating feature were completed. A 300x550x10 mm size panel with carbon fibre/epoxy composite skins and foam core was fabricated with a full layer of glass/epoxy-carbon/epoxy hybrid composite visual overload sensor integrated into the bottom skin. This rather large size was selected to demonstrate that the patented overload indication technology is suitable for high performance, industrial-scale structural components. The inspiration came from aircraft floor panels, which must be light, strong and durable. Falling objects may overload the floor panel, so the indication of damage accumulation observable from the underside is a valuable added function. During the experiments, the sensing layer provided clearly visible signs of overload in the form of light stripes when the high-modulus carbon fibre reinforced sensing layer started to fragment and delaminated locally around the fractures at a pre-defined strain (see Fig. 1.).

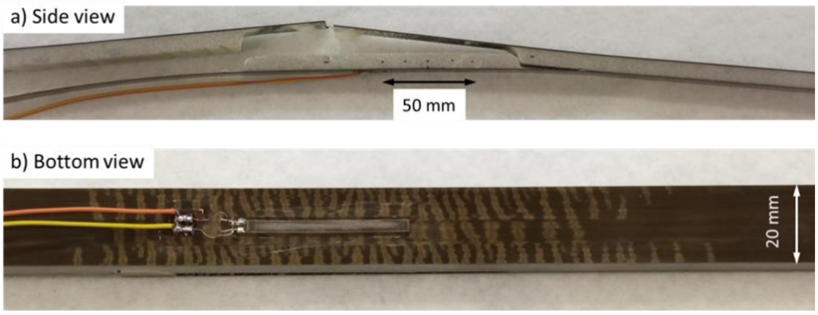

Figure 1. Failure mode and overload indication pattern of a 550x20x10 mm sandwich beam specimen

40 mm long electronic strain gauges were applied successfully and provided accurate strain readings over the highly variable strain field after the triggering of the overload sensing layer. The trigger strain of the visual overload sensor was in line with the expected failure strain of the sensing layer. Acoustic emission damage monitoring was able to detect the triggering of the visual overload sensor, and the obtained data correlated well with the knee point after the initial linear part of the load-displacement diagrams and the small drops in the load signal from the test machine, indicating the fragmentation of the sensing layer. We plan to publish the results in an international conference paper.

2.2. Mode II interfacial fracture toughness improvement by interleaving

High initial elastic modulus can be achieved in sandwich hybrid composites if the ratio of the low elongation (carbon fibre/epoxy) and high elongation (glass fibre/epoxy) layers is increased. However, a thick carbon/epoxy ply releases too much energy upon its first fracture to keep the failure process stable and catastrophic delamination of the layers takes place. In order to ensure stable, pseudo-ductile failure (i.e. fragmentation of the carbon/epoxy layer), the layer interfaces have to be modified to have high enough mode II interlaminar fracture toughness. Thicker carbon/epoxy plies are also cheaper than the unique ultra-thin versions, which have been used to demonstrate pseudo-ductility so far. A successful experimental programme was designed and executed using 30 and 60 μm thick thermoset resin films (the same epoxy as the matrix of the composite plies) and thermoplastic acrylonitrile butadiene styrene (ABS) films at the layer interfaces to increase the mode II interlaminar fracture toughness and stabilise the delamination in the hybrid laminates made with standard thickness carbon/epoxy plies sandwiched between glass/epoxy layers. The key toughening mechanism in the case of the more successful epoxy film interleaving was the local decrease of the fibre volume fraction at the modified interfaces of the unidirectional hybrid composite, which increased the shear compliance and damage tolerance. The results were published in a journal paper [1].

The effect of oxygen plasma treatment on the bonding between the ABS films and the composite layers was also analysed in detail. We concluded that the plasma treatment promoted a better adhesion, but no breakthrough was achieved with styrene-based interleaf films. The results of this study were also published in a journal paper [2].

Excellent results were achieved with another thermoplastic film: polyamide 12 (PA 12), which has not been applied in film format before as an interleaf layer for toughening laminated composites. The hybrid laminate specimens, which failed with unstable delamination without the PA 12 film interleaves, demonstrated favourable pseudo-ductile behaviour with 20 μm PA 12 films at the glass/epoxy-carbon/epoxy layer interfaces. The mode II interlaminar fracture toughness of the hybrid laminates was increased by two-fold (from 2.0 kJ/m^2 to 4.0 kJ/m^2) [3]. Good potential was identified in this new type of interleaf film, and we decided to use PA 12 for the reparability study.

One of the co-investigators of the programme was working actively to develop new electrospinning techniques to enable high throughput production of wide but uniform nanofibrous mats of low diameter fibres, which can cover larger areas and, therefore, suitable for interleaving composites. Two new methods (i.e. the shear-aided needleless and self-feeding electrospinning) were proposed, and the results were published in journal papers [4]-[6]. Our electrospinning group also developed special nanofibrous mats for mask filters and respiratory monitoring purposes. These new applications arose in response to the pandemic. The results were published in journal papers [7],[8]. Furthermore, the developed poly-lactic acid (PLA) nanofibrous layers were also applied as interleaves in the same 3D-printed thermoplastic material. The new concept yielded materials that can be considered unique self-reinforced nanocomposites, which showed superior crystallinity and mechanical properties and offered excellent scope for complete biodegradability. The results were published in a journal paper [9].

Inspired by the success of our electrospinning technique developments, we decided to use nanofibrous mats as an additional modification approach to the layer interfaces of our hybrid laminates made with standard thickness carbon/epoxy prepreg plies to make their failure pseudo-ductile. Electrospun nanofibrous layers made of polyamide 6 (PA 6) were used as interleaf layers in different areal densities ranging from 2 to 20 g/m^2 between the carbon/epoxy and the glass/epoxy composite layers of our hybrid laminates. The interleaved nanofibrous layers helped to control the thickness of the well-defined interlayers between the composite blocks with thicknesses in correlation with the areal densities of the applied interleaves. Thicker interlayers led to lower interfacial shear strength values determined based on the fracture spacing of the fragmented specimens obtained visually due to the translucency of the outer glass/epoxy layers of the hybrid laminates. On the other hand, the mode II fracture toughness increased with interlayer thickness, which is identified as an interesting trade-off in the interfacial properties of our pseudo-ductile hybrid composites. The results were published in a journal paper [10].

2.3. Development of pseudo-ductile and reparable composite material

We initially proposed two different approaches to repair delamination between the layers of pseudo-ductile hybrid composites i) the creation of an internal vascular network in the material to deliver the liquid repairing agent to the delaminated areas and ii) adding thermoplastic films to the layer interfaces, which can be melted at elevated temperature to re-bond the delaminated areas. We found some interesting studies where the authors managed to make an internal vascular network by including steel wires in the composite laminate, which were extracted (pulled out) after manufacturing to leave small diameter (below 1 mm) channels in the material. However, this approach did not seem feasible at an industrial scale as the wires would break and could not be extracted if they were too long or not perfectly straight. Therefore, we decided to focus on the more promising approach with PA 12 films, where we gained significant experience and promising results in the earlier phase of the research programme.

Three-layer unidirectional hybrid composites were made from prepregs in an autoclave with glass fibre/epoxy and carbon fibre/epoxy layers, including 20 μm thick PA 12 films at the layer interfaces. The carbon-epoxy layer was cut into 50 mm long strips before laying up the hybrid laminates. This way, the fracture of the carbon fibre epoxy layer was avoided, and only delamination took place upon tensile loading of the specimens. The material demonstrated a desirable non-linear pseudo-ductile stress-strain response with smooth transitions between the initial linear, the slightly rising plateau and the second linear phases. The careful control of the damage mode of the material (i.e. no fibre breakage) allowed us to restore the initial stiffness of the specimens, after 30-40% loss due to delamination, by repairing the specimens under heat and pressure. The damaged specimens were placed in our autoclave at a temperature just above the melt temperature of the PA 12 films inside and 7 bar pressure for only 10 minutes. As a result, the delamination visible from the outside of the specimens due to the translucent nature of the outer glass fibre/epoxy layers disappeared. At the same time, the stiffness of the specimen was fully restored to that of the pristine samples (see fig. 2.) right until the strain and stress corresponding to the limit of linear stress-strain response (i.e. knee point).

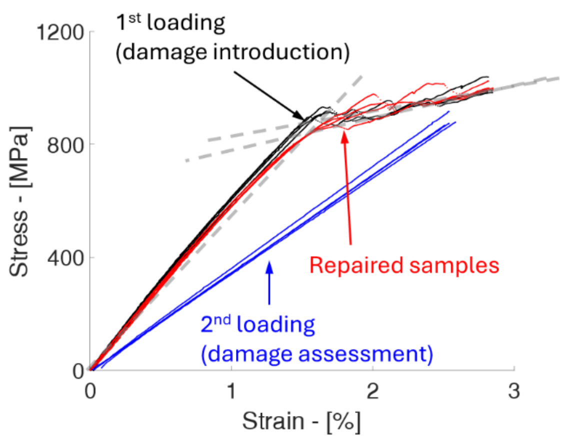

Figure 2. Stress-strain response of our reparable hybrid composite material

We summarised our results in a journal paper [3]. This was the first time to demonstrate reparability and pseudo-ductility at the same time in the field of high performance fibre reinforced polymer composites. Therefore, we believe that our concept and results can open a new field for demanding applications.

2.4. Out-of-plane damage characterisation, damage control by interleaving

After an extensive literature survey in the field, we realised that impact loading to a supported panel would induce damage in a dynamic manner, which is hard to control. Therefore, we decided to start the experimental campaign by quasi-static indentation loading, which is analogous to impact in direction and potentially in the total energy exposure, but it is much easier to control by stopping the displacement of the indenter at any point. We carefully selected the support for the indentation tests to be the same as the standard support frame for compression after impact testing. We designed 4 mm thick quasi-isotropic laminates with and without interfacial modification at the mid-plane of the laminates, where the highest shear stress is expected during out-of-plane indentation. The scope of the study was to analyse the extent of the damage introduced in the 100x150 mm test panels by the penetration of a 16 mm diameter hemispherical indenter until a given displacement. Since these techniques have not been applied at our lab before, we had to pioneer the area by the first attempts to set up the test environment, fit the support frame to our test machine, design and manufacture the indenter tool using an existing falling-weight dart tip and execute the first preliminary tests. After the first series of indentation tests on carbon/epoxy panels that did not show a clear effect of interfacial modification at the midplane, we decided to switch to glass fibre/epoxy material, which is transparent and can potentially show damage evolution inside the laminate if sufficiently strong backlight is applied. We also decided to use another type of interfacial modification at the mid-plane of the laminate. Instead of toughening the interface with PA12 film, we inserted solid and laser-perforated PTFE release film pieces in the middle of the panels, which acted as an interfacial weakening element to control the onset of damage and localise delamination initiation to the modified interface. This could be a successful approach to minimise delamination at other interfaces and keep two thick blocks of the laminate almost intact, which can take a higher load in the subsequent compression after indentation test than a laminate which contains damage at multiple interfaces. Indentation tests were executed while the damage evolution was recorded with a digital camera originally supplied with our video extensometer. The test panels having a complete layer of perforated release film accumulated significantly more damage at the modified interface than the ones having 40 or 49 mm diameter perforated release film discs embedded (see fig. 3.).

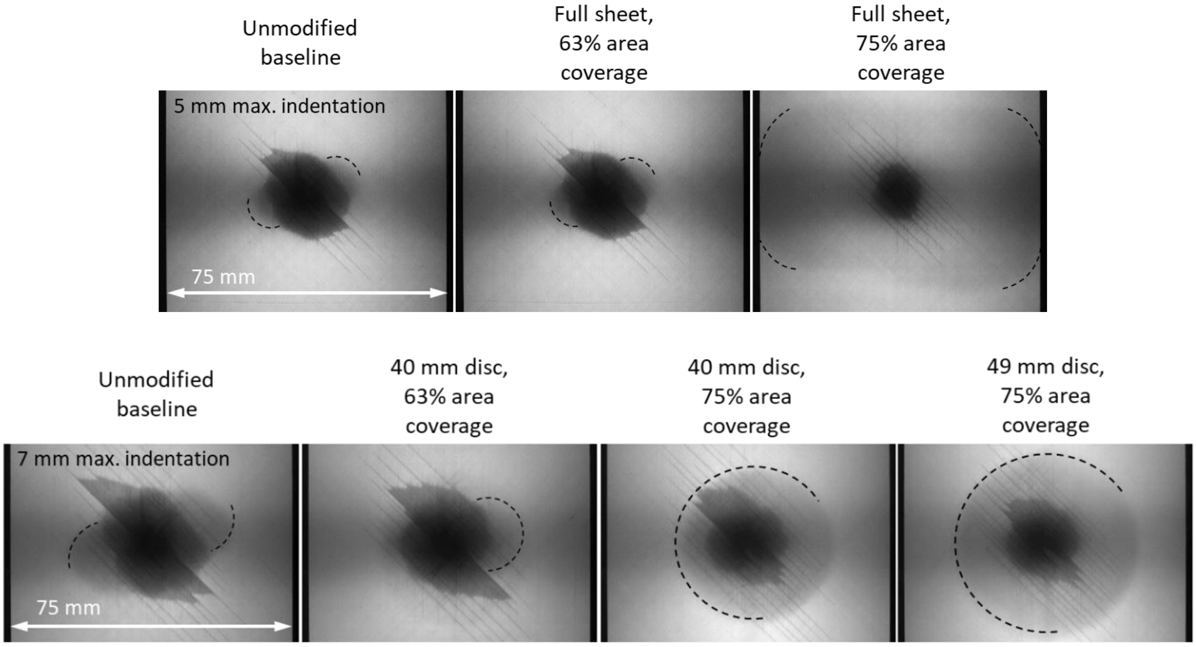

Figure 3. Delamination at multiple interfaces in 4 mm thick glass fibre/epoxy test panels with different modification elements at their mid-plane after 5 and 7 mm indentation (backlight images, dashed arcs indicate the approximate boundaries of mid-plane delamination)

In the subsequent in-plane compression test phase, the modified panels showed a more gradual response than the pristine baseline panels, but their damage accumulation started, and final failure took place at lower stresses and strains. Less catastrophic failure mode was achieved at the price of lower compressive failure strain and stress. We plan to publish our results in a journal paper.

2.5 Research activities beyond the work plan of the research programme

A number of research studies were pursued during the period of the research programme, which were not included in the original work plan but are within the broader scope of the grant.

The PI had a unique opportunity to join a world-wide team of top experts to develop new specimen concepts for more accurate tensile testing of unidirectional carbon fibre/epoxy composites than is possible with current standard coupons. Since this is a hot topic even decades after the introduction of carbon fibre/epoxy in the aerospace industry, we decided to take part, although it was not included in the initial work plan of the research programme. A round-robin exercise involving our sandwich hybrid composite specimen concept [11] and up to 7 well-reputed research laboratories was initiated by KU Leuven with the final goal of standardising the best approaches. We prepared seven 300x300 mm sandwich composite plates and sandpaper tabs for the joint effort, which were cut into coupons centrally at the KU Leuven and sent out to the participants across Europe. Five different types of unidirectional carbon/epoxy specimens were delivered to us for mechanical testing, which we executed with a BSc student with full accuracy. The longitudinal strains were measured with a double-camera 3D video-extensometer system on the front face and electronic strain gauges on the back face of the specimens. The sandwich specimens developed by our group performed best at our lab, exhibiting the highest failure strains due to the continuous glass/epoxy protective layers along the carbon/epoxy layer, eliminating stress concentrations near the gripped regions and assuring gauge section failures in most cases. The results of this extensive experimental study are planned to be published in a journal paper in due course as a joint effort of the participants. The PI also submitted a separate journal paper [12] about the development of the layer-joining technique of sandwich composite coupons.

The PI was involved in a project investigating the effect of transverse compressive stresses on the tensile failure strain of carbon fibre/epoxy with his PhD student in collaboration with the University of Bristol. The core of the concept was to use angle-ply layers with carefully designed orientation to induce compressive stress in the adjacent 0° carbon/epoxy plies and analyse its effect on the tensile failure strain of the 0° layer. One specimen type had glass fibre/epoxy protective plies on its outer surfaces, similar to the ones we proposed for accurate tensile testing of unidirectional carbon fibre/epoxy [11]. The results were published in a journal paper [13].

The PI worked on a project with one of his MSc students where they developed bi-directionally reinforced pseudo-ductile composites that had not been presented in the literature before. The results were published in a journal paper [14].

The results of our systematic study of the environmental effects on fragmenting and delaminating pseudo-ductile hybrid composites were published in a journal paper [15]. An MSc student is a co-author of the paper.

The PI contributed to the preparation of a review paper [16] on the results generated in the field of pseudo-ductile composites in the last twelve years, together with former colleagues from Bristol University and Imperial College London. The paper presents his results with pseudo-ductile hybrid composites in a separate chapter.

3. References

(The entry marked with * was not published with the support of the research grant.)

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