COMPARTMENTED FIBRES: THE CONCEPT OF MULTIPLE SELF-HEALING IN ADVANCED FIBRE COMPOSITES

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ABSTRACT

Polymers reinforced with high performance fibres are successfully replacing metal alloys in lightweight aircraft structures. A critical factor in structural design is the resistance of a structure to progressive damage which develops during its service time. The brittle nature of matrix cracking is the main source of composite failure initiating ply delamination and fibre to matrix debonding.

Bio-inspired autonomous healing systems, embedded in a polymer matrix, are being developed to improve the fracture resistance of structural composites [1, 2, 3]. An exemplary system can consist of fibres with individual compartments, i.e. compartmented fibres, which are filled with a healing liquid. The healing agent is distributed within the fibre in the form of long elongated compartments of ellipsoidal shape with high aspect ratio. The fibres are designed to release the liquid healing agent at multiple specific microcrack sites developed in the polymer matrix as a result of structural loading during its life-time.

The advantage of such a fibre as a healing agent carrier is obvious - compartmented fibres enable multiple local healing events. Neither is the whole healing agent consumed in the first damage/healing event (as for hollow fibres) nor is the functionality of remaining compartments affected by the healing event (compared to vascular networks).

Compartmented fibres were spun from an oil/water emulsion of a healing agent in an aqueous solution of sodium alginate. The retention of the liquid healing agent (ortho-dichlorobenzene) in a solid fibre was provided by the coagulation of the alginate polymer during fibre formation.

Spun fibres were embedded in polymethylmethacrylate, pre-cracked in 3 point bending and allowed a certain period of time to self-heal before they were loaded again. Computed micro-tomography was used to visualise damage and healing sites. Test samples with the new fibres located at the likely damage sites were able to recover, (in part) their mechanical properties.

The concept of multiple release of healing agent from a compartmented fibre into the surrounding polymer matrix was demonstrated successfully.
1. INTRODUCTION

Multi-compartmented fibres were spun from an oil in water emulsion of a healing agent in an aqueous solution of sodium alginate [4]. The emulsion was extruded through a spinneret into a coagulation bath where the fibre solidification and liquid retention took place as a result of the ion exchange reaction (sodium alginate is a natural polysaccharide able to cross-link with bivalent ions such as Ca$^{2+}$ and form a three-dimensional network). In this work alginate fibres containing 1,2-dichlorobenzene as a healing agent for polymethylmethacrylate (PMMA) resin were prepared. Spun fibres were embedded in a PMMA resin and compression moulded. Three-point bending test combined with computed µ-tomography was used to prove that compartmented fibres are able of distributed multiple-healing of the surrounding polymer matrix.

2. MATERIALS AND SAMPLE PREPARATION

2.1 Materials

Sodium alginate (ALG), α-dichlorobenzene (DCB), Poly(ethylene-maleic-alt-anhydride) (PEMA) and Calcium chloride hexahydrate were purchased from Sigma-Aldrich, The Netherlands.

2.2 Spin dope preparation and fibre spinning

Sodium alginate was dissolved in de-ionized water and a 6 wt.% solution was prepared. PEMA 2.5 wt.% solution was prepared by dissolving the copolymer in water at 70ºC/60 min. Appropriate amounts of PEMA solution were mixed with the sodium alginate solution using a high speed mixer at 2500 rpm for 2.5 min. After that the mixture was continuously stirred with a three-blade stirrer and the DCB was slowly injected. The emulsification took place at 300 rpm for 5 and 10 min, depending on the desired morphology of the emulsion. The composition of the final emulsion was as follows: the DCB/ALG ratio was 1/4 and the PEMA/DCB ratio was 1/50. Fibres with small and large compartments were spun using a plunger based lab scale spinning line in a conventional wet spinning process. A spinneret containing one capillary of 0.25 mm diameter and 2 mm length was used. The extrusion rate was 38 µl/min. and the take up speed was 1.3 m/min. The coagulation bath was 0.8 m long and contained a 0.45 M solution of CaCl$_2$.6H$_2$O. Fibres were wound onto a pre-heated (40ºC) plastic bobbin.

2.3 Composites

As spun compartmented fibres were unidirectionally aligned and embedded in a thin layer of castable polymethylmethacrylate (PMMA) on top of 3 mm thick extruded sheet of PMMA. The thickness of the layer containing the fibres was 300 µm. Beams for three point bending test were cut from the sheets. Compartmented fibres were placed on the tension side of the beam.
3. METHODS

3.1 Three point bending

Composite beams were tested in three-point bending \((off-axis \, geometry)\). Composites were pre-strained up to \(\varepsilon = 4\%\) to crack open the compartments and allowed to heal after the test. After the healing period the samples were bended again, this time in a different normal plane \((i.e. \, off-axis)\), and up to the strain of \(\varepsilon = 7\%\) (ultimate failure). X-ray micro-computed tomography was used to observe compartments crack opening and the release of the healing agent into the surrounding polymer matrix. Samples were X-ray scanned before bending, after the first and after the second bending test.

3.2 Computed \(\mu\)-tomography \((\mu\text{-CT})\)

Compartment crack opening and healing agent release was measured with Phoenix Nanotom X-ray tomography scanner, using 100 kV radiation. The contrast of the encapsulated healing agent was enhanced with 1,2-dibromobenzene (density of 1.956 g/cm\(^3\)). The samples were rotated along their longitudinal axis and three X-ray attenuation images were recorded and averaged every 0.5º. The image reconstruction was done using the Phoenix reconstruction software package. The resolution of the X-ray tomography scan was \((3\mu\text{m})^3\) per voxel. The final rendering of the 3D tomography of the cracked compartments was completed with Volume Graphics 2 software.

4. RESULTS

Computed \(\mu\text{-tomography}\) is a powerful non-destructive testing method which was used to qualitatively investigate the release of healing agent from compartmented fibres embedded in polymer matrix after the flexural test of polymer composites.

![Figure 1: Morphology of compartmented fibres embedded in a polymer matrix before and after successive flexural tests reconstructed with computed \(\mu\)-tomography.](image)

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Figure 1 shows the compartment opening and healing agent release into the polymer matrix before and after successive flexural tests. Release of healing agent is manifested from the formation of local cauliflower like structures. Pre-cracked composites were allowed to heal for one week prior to subsequent secondary off-axis loading. The images shown are viewed from the tension side of the composite beam. It can be seen from Figure 1 that new compartments are opened when the fibre is more strained and new healing agent is delivered into the neighbouring matrix. The compartmented fibre responds to the damage progressively.

![Figure 1: Compartment opening and healing agent release into the polymer matrix before and after successive flexural tests.](image)

5. CONCLUSIONS

Micro-computed tomography combined with off-axis flexural test proved that compartmented fibres enable multiple local healing in fibre reinforced polymer composites. All the tested model composites were able to partially recover their mechanical properties. The concept of multiple release of healing agent from a fibrous carrier was demonstrated successfully.

REFERENCES

