



Mechanical properties of mendable composites containing self-healing thermoplastic agents



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ABSTRACT

The tensile and compressive properties of mendable composites containing thermoplastic particles as self-healing agents are assessed in this paper. The effects of the type and concentration of thermoplastic healing agent on the mechanical properties of carbon fibre–epoxy composites were determined experimentally by mechanical testing and computationally using finite element analysis. The elastic modulus and failure stress of the mendable composites decreased at a linear rate with increasing weight fraction of thermoplastic agent in the epoxy matrix phase. Microstructural analysis revealed the thermoplastic agents thickened the polymer-rich layers between the plies, and this reduced the average fibre volume content of the mendable composite thereby lowering its mechanical properties. Restoration of the in-plane properties of mendable composites following healing of delamination damage using thermoplastic particles was also assessed. Healing partially restored the compressive modulus of the mendable composites, but had no healing effect on their compressive strength or tensile properties.

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1. Introduction

Self-healing of thermoset polymers and fibre reinforced-thermoset matrix composites can be achieved in several ways, including microcapsules or microvasculates containing liquid resin healing agent [1–4], polymers with reversible cross-links [5] or mechanically-responsive covalent bonds [6], and thermoplastic–thermoset solid solutions (in which the thermoplastic agent is fully dissolved within the solid thermoset matrix) [7]. An emerging self-healing method uses thermoplastic agents dispersed as discrete and solid particles, fibres or stitched filaments within the polymer matrix [8–22]. The thermoplastic resides in the thermoset matrix as an insoluble phase which is inert until the healing process is activated by heat. Thermoplastic agents used in high concentrations (typically above 5%) are effective at healing cracks in epoxy resin and delaminations in mendable carbon–epoxy composite. The most studied and best understood thermoplastic healing agent is the ionic copolymer poly(ethylene-co-methacrylic) acid (EMAA) [8–22]. Poly(ethylene-co-glycidyl methacrylate) (PEGMA) and ethylene vinyl acetate (EVA) are also effective thermoplastic healing agents [23,24], although less is known about their healing

performance. Except for recent research by Varley and colleagues [23,24], the healing properties of PEGMA and EVA have not been thoroughly investigated, although they are effective at repairing damage in thermoset matrix composites.

Not all thermoplastics can be used as a healing agent; as they must have several essential properties to be effective at repairing cracks. Firstly, the thermoplastic must have sufficiently low melting temperature and viscosity so it can flow easily into open cracks while the thermoset matrix remains solid during the elevated temperature healing process. Secondly, ionic copolymers such as EMAA and PEGMA are effective healing agents because they contain functional groups which are chemically reactive with amine groups in epoxy. Condensation reactions at elevated temperature generate by-products (mostly water) which phase separate into high pressure bubbles within the thermoplastic. This pressure forces the thermoplastic (which is molten at elevated temperature) into open cracks thereby filling the damage and healing the composite upon cooling. Another essential property is the thermoplastic must have good adhesive properties with epoxy to ensure strong bonding after filling the crack. Weak bonding may allow the crack to reform, and therefore strong non-covalent bonding such as hydrogen bonds is necessary between the thermoplastic and epoxy matrix to achieve good healing performance.

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Until now, research into thermoplastic healing agents has focussed solely on their healing efficiency in terms of restoration of interlaminar fracture toughness and interlaminar fatigue resistance as well as the delamination healing mechanism [8–24]. The effect of thermoplastics on other properties of mendable epoxy and fibre–epoxy composites has not been investigated. The high volume fraction of thermoplastic needed to heal large cracks may affect the in-plane mechanical properties and environmental durability of mendable composites. However to date, research into the impact of thermoplastic agents on the mechanical properties of composites before and after healing has not been investigated and so is necessary to determine the extent to which the thermoplastic agent is beneficial, detrimental or has no impact upon the properties of composites. It is worth noting that the influence of other self-healing methods such as microcapsules and microvasculates on the mechanical properties of polymers and polymer matrix composites has also not been extensively investigated, and only a few studies have been reported (e.g. [25–27]).

The study presented in this paper therefore investigates the effect of thermoplastic agents on the mechanical properties of mendable carbon–epoxy composites. The effects of the type and concentration of thermoplastic on the tensile and compressive properties of the composite are measured experimentally and computed using finite element analysis. The types of thermoplastic studied were EMAA, PEGMA and EVA and the weight percentage of healing agent in the epoxy matrix phase was between 5% and 15%. Changes to the mechanical properties caused by the thermoplastic agents are related to changes in the microstructure of the mendable composites. This study also aims to determine the healing efficiency of thermoplastic agents on the in-plane properties of damaged composite materials. The capacity of thermoplastic agents to heal delamination cracks and restore the mechanical properties of mendable composites is investigated. The recovery to the tensile and compressive properties following healing of delaminated composites using different types and concentrations of thermoplastic agent is determined.

2. Materials and experimental testing

2.1. Mendable carbon–epoxy composites

Carbon fibre composites were made using unmodified epoxy or epoxy modified with thermoplastic healing agent. The epoxy resin was diglycidyl ether of bisphenol A (DGEBA, D.E.R 331 from Dow Chemical Company, USA), and it was cured using triethylene tetramine (TETA, DEH24 from Dow Plastics Australia). The thermoplastic healing agents were EMAA (Nucrel 2940, DuPont Polymers, USA), PEGMA (Sigma–Aldrich, Australia) and EVA (Evathene UE633, USI). The as-received thermoplastic pellets were cryogenically ground into particles which were then passed through sieves to collect particles in the size range of 245–425 μm . The epoxy resin was heated to 65 $^{\circ}\text{C}$ to lower the viscosity followed by addition of the thermoplastic particles which were then evenly

dispersed throughout the liquid resin via vigorous stirring with a propeller blade rotating at 300 rpm for 30 min. EMAA particles were added to the resin at weight fractions of 5%, 10% or 15% whereas the weight fractions of PEGMA and EVA were 10%. After stirring, the blended mixture was degassed for 25 min in a vacuum oven at 50 $^{\circ}\text{C}$, cooled to room temperature, and then TETA hardener was added at a ratio of 13 parts to 100 parts of DGEBA to attain a 1:1 stoichiometric network following polymerisation.

The unmodified and thermoplastic modified liquid epoxy/amine formulations were applied to plain woven carbon fabric using a brush and roller, and then ten plies were stacked in a cross-ply orientation. The carbon fabric contained an equal fraction of warp (0°) and weft (90°) tows. The warp tows of the fabric plies were aligned in the same orientation and this resulted in a cross-ply laminate. The composites were cured and consolidated at 70 $^{\circ}\text{C}$ and 2 MPa for one hour and then post-cured at 150 $^{\circ}\text{C}$ for 30 min. It is important to note that the thermoplastic particles remained immiscible in the epoxy network throughout the consolidation and cure process. In addition to this, the reactivity of the TETA hardener at 70 $^{\circ}\text{C}$ ensured that the thermoplastics did not melt prior to vitrification of the network as their melting points are all above 80 $^{\circ}\text{C}$. Any chemical interaction between the thermoplastic and the residual functional groups on the epoxy network which originates the pressure delivery mechanism, therefore would necessarily occur either while the thermoplastics are in the solid state at 70 $^{\circ}\text{C}$, or when physically locked into a three-dimensional cross-linked intractable network post vitrification of the network during post-cure at 150 $^{\circ}\text{C}$.

The final thickness and carbon fibre content of the unmodified and mendable composites are given in Table 1. The fibre content was measured using the resin burn-off test according to ASTM D2548-11 specifications. Composite specimens were heated to 565 $^{\circ}\text{C}$ for two hours in air to decompose the epoxy and thermoplastic particles and to oxidise any residual char. These conditions caused very little oxidation and mass loss to the carbon fibres [28]. The mass loss was used to determine the volume fraction of carbon fibre in the composites.

Table 1 shows the thermoplastic particles increased the thickness of the composite. The thermoplastic did not replace the space occupied by the epoxy (which remained approximately constant), but added to the volume fraction of the organic matrix phase. Fig. 1 shows that the thermoplastic particles are located between the ply layers. The particles, which were initially round in shape, were flattened due to pressure applied on the composite during the cure and consolidation process. The particles, which are between 245 and 425 μm in diameter, are too large to fit between the fibres within the woven plies. Their large size confined the thermoplastic particles to the polymer-rich interply layers which caused them to thicken. This thickening of the interply layers accounts for the increase in total thickness of the mendable composites with the weight fraction of healing agent.

Table 1 also shows the increase in composite thickness was not dependent on the type of thermoplastic. At the weight content of

Table 1
Physical and thermal properties of the composite materials.

Composite	Healing agent content in epoxy (wt%)	Healing agent content in composite (vol%)	Thickness (mm)	Percentage increase in thickness (%)	Carbon fibre volume content (%)	Glass transition temperature, T_g ($^{\circ}\text{C}$)
Unmodified	–	–	2.00 \pm 0.07	–	0.39 \pm 0.05	90
EMAA	5	4.7	2.36 \pm 0.12	18	0.32 \pm 0.03	
EMAA	10	9.2	2.71 \pm 0.12	36	0.26 \pm 0.03	97
EMAA	15	14.3	2.96 \pm 0.19	48	0.21 \pm 0.05	
EVA	10	9.0	2.73 \pm 0.03	37	0.29 \pm 0.02	97
PEGMA	10	8.7	2.73 \pm 0.12	37	0.29 \pm 0.03	83

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