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Enhancing fatigue resistance and damage characterisation in adhesively-bonded composite joints by carbon nanofibres

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ABSTRACT

In the present work we report on the use of carbon nanofibres (CNFs) to simultaneously improve the cyclic fatigue resistance and the detectability of disbonding in adhesively-bonded structures made of carbon-fibre reinforced-plastic (CFRP) composites. The effects of the concentration of the CNFs (i.e. 0.4, 0.7 and 1.0 wt%) and their orientation (i.e. random versus aligned) in the epoxy-adhesive layer between two CFRP substrates are investigated. The results show that increasing the concentration of randomly-oriented CNFs (a) improves greatly the mode I fatigue resistance of the adhesive layer, including raising the crack growth threshold of the cyclic strain-energy release-rate, and (b) increases the quasi-static fracture toughness. Further improvements in the fatigue resistance occur when the CNFs are aligned perpendicular to the plane of the joint, i.e. normal to the crack plane, as opposed to being randomly-oriented in the adhesive layer. In addition, the CNFs form a conductive network that makes it possible to detect and characterise fatigue-induced disbonding using an electrical-resistance technique. A simple model is developed for the relationship between the disbond (i.e. crack size) and the electrical resistance of a bonded joint with conductive substrates. Finite element analyses are carried out to quantify the applicability of this model as a function of the conductivity of the adhesive from 10^{-4} S/m to 1 S/m. The results confirm that the proposed simple model is highly accurate for joints where the composite substrates have a through-thickness electrical conductivity exceeding a hundred times that of the adhesive. This research paves the way for new multi-functional adhesives with greatly enhanced fatigue resistance and disbond detection capability.

1. Introduction

Fibre-reinforced polymeric composites, and adhesively-bonded components using such composites, have a relatively low mechanical strength in the through-thickness direction. As a result, these structures are susceptible to damage such as delamination and disbonding, especially when subjected to cyclic fatigue loading [1,2]. Furthermore, non-destructive detection of delamination cracks in composites, and disbonds in adhesively-bonded structures, are important requirements for the safe operation of these advanced structures. The electric potential-drop and eddy-current techniques that have been developed for metallic structures are not applicable for composite structures due to the low through-thickness electrical conductivity as a result of the relatively very low dielectric constant of the polymeric materials [3]. Ultrasound techniques, which are most commonly used for non-destructive evaluation of composites, are not effective for bonded structures because the high acoustic impedance mismatch at substrate-adhesive interface tends to overshadow any disbonds. Therefore, there is a strong need for improving the damage tolerance and damage detection of composites and adhesively-bonded composite structures.

Epoxy polymers are widely used as the matrix materials for fibre-reinforced composites and as adhesives for joining structural composite components. Despite having many desirable properties, epoxies typically exhibit low electrical conductivity and a low resistance to fracture and fatigue crack growth [1]. Recent studies

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have shown that conductive carbon nano-reinforcements such as carbon nanotubes (CNTs) [4,5], carbon nanofibres (CNFs) [6,7], and graphene [8,9] can simultaneously improve the fracture resistance and electrical conductivity of an epoxy. In addition, carbon nano-reinforcements improve the delamination resistance of fibre-epoxy composites [10–15]. Several other studies have reported the use of conductive networks of carbon nano-modifiers for detecting delamination damage in fibre-reinforced composites [14,16–21]. However, very few studies have investigated the prospects of damage detection in adhesively-bonded composite joints using carbon nano-reinforcements. Two relevant studies [22,23] have focused on the electrical response of CNT networks in single-lap shear joints. The study by Lim et al. [22] examined the use of CNT networks to detect damage in adhesively-bonded hybrid composite/steel lap joints subjected to quasi-static and cyclic tensile loading. Under quasi-static loading, the electrical resistance of the single-lap shear joints was reported to increase by up to 10–30% as the shear stress increased to ~50–80% of the joint strength. During cyclic loading, the electrical resistance increased monotonically with cycling by ~5–20% prior to ultimate failure of the joints. This increase in the electrical resistance was attributed to the accumulation of damage in the adhesive layer, which was detected using an acoustic emission technique. Although the increase in electrical resistance correlated with the number of acoustic emission events, Lim et al. found that it was not possible to quantitatively relate the damage size with an increase in the resistance due to limitations in the acoustic detection technique. Mactabi et al. [23] studied the effects of CNTs on the electrical response and fatigue life of adhesively-bonded aluminium lap joints subjected to cyclic loading, and reported that the electrical resistance of the joints increased slightly (by ~10%) or remained unchanged throughout a large portion of the fatigue life. However there was a large increase in the electrical resistance during the final phase of the fatigue life, just prior to ultimate failure, possibly due to crack growth within the joint. However, they did not report any correlation between the size of any crack and the changes in electrical resistance.

While the studies by Lim et al. [22] and Mactabi et al. [23] demonstrated the effectiveness of CNT networks for detecting the onset of damage in the adhesive layer of single-lap joints, they did not investigate whether the change in the electrical resistance could be used to characterise the damage size, or monitor its growth. A short communication by Zhang et al. [5] reported the use of CNT networks to detect and monitor the size of fatigue cracks in bulk epoxy nanocomposites. The epoxy nanocomposites, containing 0.5 wt% MWCNTs, were subjected to cyclic fatigue loading using the compact tension test. Zhang et al. [5] showed that changes in the electrical resistance could be used to determine accurately the fatigue crack size. However, it is not clear how this technique can be used to detect and monitor fatigue cracking in adhesively-bonded composite joints where the electrical resistance of the joint is also influenced by the conductivity of the composite substrates. Moreover, the result from the study by Zhang et al. [5] was confined to a single concentration of nano-reinforcement in the epoxy nanocomposites. The effects of the concentrations and orientation of the nano-reinforcement in the epoxy nanocomposites on the damage detectability using the approach demonstrated by Zhang et al. [5] remains unknown. Although the aforementioned studies [5,22,23] investigated the use of CNTs for detecting damage in adhesively-bonded joints, to the best of our knowledge, no studies have reported the use of CNFs for simultaneously improving fatigue resistance and enabling damage detection in adhesively-bonded joints. It is worth noting that the electrical conductivity of adhesives can be further enhanced by an electric field to align carbon nano-reinforcements in the through-thickness direction of the bond layer [4,7,8,24].

Considering that CNFs may be an excellent alternative to CNTs due to their wide availability and lower cost [25], the present study focuses on using CNFs for simultaneously improving the fatigue resistance and enabling crack-growth monitoring in adhesively-bonded composite structures. The effects of the alignment and weight concentration of CNFs on the fatigue resistance and electrical conductivity of the epoxy adhesive layer are investigated. Alignment of the CNFs is accomplished by applying an external alternating current (AC) electric field whilst the adhesive is in its liquid state, prior to cross-linking. The electrical response of the adhesively-bonded composite joints as a function of the orientation and concentration of the CNFs in the adhesive layer is measured *in-situ* during mode I cyclic loading using a direct-current (DC) potential drop technique. A simple analytical model that accounts for the conductivity of the adhesive and composite substrates has been developed. This model extends the resistivity model for nanocomposites, proposed by Zhang et al. [5], to determine the size of the fatigue cracks from the *in-situ* measurements. Finite element (FE) analyses are performed to verify the range of applicability of the proposed resistivity model by characterising the effects of the conductivity of the adhesive and the composite substrates on the accuracy of the analytical model.

2. Materials and experimental details

2.1. Materials

The epoxy resin used was a liquid blend of bisphenol A and bisphenol F (‘105’ from West System) and the hardener (‘206’ from West System) was a blend of aliphatic amines and aliphatic amine adducts based on diethylenetriamine and triethylenetetramine. Commercially available vapour-grown carbon nanofibres, Pyrograf® - III PR-24-HHT, supplied by Applied Sciences Inc., USA, were employed as the nano-reinforcements. The CNFs had a diameter in the range of about 70–300 nm and a length in the range of 30–200 μm in their as-supplied form. The composite substrates were prepared using 12 plies of unidirectional T700 carbon fibre/epoxy prepreg (VTM 264 supplied by Applied Composites Group). A CFRP panel, with dimensions of 300 mm × 230 mm × 2.5 mm, was cured and consolidated in an autoclave at 120 °C for 1 h, in accordance with the manufacturer's recommended cure process. The surface of the cured CFRP panel to be bonded was then abraded using 320 grit aluminium oxide abrasive paper, cleaned under running tap water for about 2 min, degreased with acetone, and finally cleaned with distilled water to remove any surface contamination.

2.2. Composite joint manufacture

A three-roll mill (Dermamill 100) was used to disperse the CNFs in the liquid epoxy resin. The details of the three-roll mill dispersion technique and the preparation of the adhesive joints are given in our earlier studies [7,24]. Essentially, the amine-based curing agent was added to the dispersed CNF/epoxy resin mixture. The CNF-modified epoxy resin mixture was then used as an adhesive layer, 2 mm in thickness, to bond the CFRP substrates. The double cantilever beam (DCB) adhesively-bonded joints were used for the fracture and fatigue tests and were 230 × 20 mm in size. The DCB joints used for characterizing the electrical resistance response during fatigue crack growth were 100 × 20 mm in size. The adhesive was cured for 48 h at room temperature (i.e. 25 °C), in accordance with the resin supplier's recommendations. The adhesive layer was prepared using three different concentrations of CNFs, i.e. 0.4, 0.7 and 1.0 wt%. In addition, an adhesive without CNFs