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Journal of Materials Research and Technology
www.jmrt.com.br

Original Article

Self-healing epoxy composites: preparation, characterization and healing performance☆

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ARTICLE INFO

Article history:
Received 6 June 2014
Accepted 22 October 2014
Available online 28 November 2014

Keywords:
Microcapsules
Synthesis
Low velocity impact
Thermography
Healing efficiency

ABSTRACT

Low velocity impact damage is common in fiber reinforced composites, which leads to micro-crack and interfacial debonding, where damage is microscopic and invisible. The concept of self-healing composites can be a way of overcoming this limitation and extending the life expectancy while expanding their usage in structural applications. In the current study, extrinsic self-healing concept was adopted using urea-formaldehyde microcapsules containing room temperature curing epoxy resin system (SC-15) as the healing agent prepared by in situ polymerization. Microcapsules were characterized using Fourier transform infrared spectroscopy (FTIR) for structural analysis. Size and shape of microcapsules were studied using optical microscopy and scanning electron microscopy (SEM). Size of the microcapsules was between 30 and 100 μm. Thermal characterization was carried out using thermogravimetric analysis. Microcapsules were thermally stable till 210 °C without any significant decomposition. Fiber reinforced composite fabrication was carried out in three different steps. In the first step, epoxy resin was encapsulated in urea-formaldehyde shell material, which was confirmed by FTIR analysis. In the next step, encapsulation of amine hardener was achieved by vacuum infiltration method. These two different microcapsules were added with epoxy at 10:3 ratio and composite fabrication was done with hand layup method. Finally, healing performance was measured in terms of low velocity impact test and thermoscopy analysis. Low velocity impact test with 30 J and 45 J impact loads confirmed the delamination and micro-crack in composite materials and subsequent healing recovery observed in terms of damaged area reduction and restoration of mechanical properties.

☆ Paper presented in the form of an abstract as part of the proceedings of the Pan American Materials Conference, São Paulo, Brazil, July 21st to 25th 2014.

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http://dx.doi.org/10.1016/j.jmrt.2014.10.016

1. Introduction

Thermoset polymers are widely used as matrices in most FRP composites used in structural applications due to their ease of processing, low cost and good wettability. They also offer good mechanical properties, as well as good corrosion resistance compared to metals and other engineering materials. They are subjected to different types of stresses during service life, and due to their uniqueness, they have several unique failure mechanisms compared to metals. Crack detection and repair in these types of materials are difficult and therefore self-healing techniques have been explored in a variety of ways to overcome this limitation. Another important issue is concerned with durability of these materials as they are exposed to different service loading due to their viscoelastic properties [1,2].

Dry et al. [3] first introduced capillary glass filled healing agent for self-repairing in cement and then in polymeric composites during fabrication. In a separate work, White et al. [4] used microencapsulation method as self-healing mechanism and showed it to be a better process over capillary glass tube. In their study, they used dicyclopentadiene (DCPD) as healing agent in the presence of urea-formaldehyde microcapsules. Results of the study showed a 60% healing efficiency in polymeric system. Similarly, Blaiszik et al. [5] used PUF microcapsules containing dicyclopentadiene and Grubbs' catalyst in E-glass fiber and EPON 828/EPIKURE 3274 composites, where they reported an average of 44% bond strength recovery of fiber/matrix interfacial bond for this composite system. Brown et al. [6] also did a study on effective healing efficiency of microcapsules based on various diameters for different agitation rate and catalyst concentration.

Epoxy resins are widely used in industrial applications due to their excellent chemical, physical and mechanical properties. Epoxy can be one of the more effective materials as a healing agent over other available healing compound in respect of cost and healing efficiency for self-healing composites. Damage recovery in large scale composite should be effective and have high restoration efficiency. Several research groups successfully synthesized epoxy contained microcapsules [7,8].

Low velocity impact damage is one of the significant concerns for composite materials. Composite materials show sufficient resistance if the applied load is in the fiber direction, but low resistance to impact loading which is in the transverse direction. Matrix-fiber debonding, delamination, matrix micro-cracking and fiber pull out are the common failure modes in composites under impact loading. Yin et al. [9] studied low velocity impact on the glass fiber reinforced composites with microcapsules. They used epoxy loaded microcapsules with latent hardener $CuBr_2(2\text{-MeIm})_4$ (the complex of $CuBr_2$ and 2-methylimidazole) as a healing system. They measured compression after impact for the measurement of mechanical performance after matrix recovery by crack repairing. They also used pressure and elevated temperature for the healing of large damaged areas. Patel et al. [10] also performed similar experiments for glass fiber reinforced composite with dicyclopentadiene (DCPD) loaded microcapsules and paraffin wax microspheres containing 10 wt% Grubbs' catalysts. About 51% of crack length was reduced as healing efficiency after low velocity impact. Williams et al. [11] investigated the healing efficiency for carbon fiber reinforced composite. They used hollow glass fibers as healing agent containers and applied low velocity impact testing for the measurement of healing efficiency by compression after impact. They reported that 90% recovery was possible for this system. Zainuddin et al. [12] studied the effect of the healing recovery for glass fiber reinforced composite. They used SHA in hollow glass tube for the improvement of peak load in low velocity impact testing. The LVI experiment was performed at 56 J for all samples and they showed that 53.6% improvement after second impact for SHA loaded composite in comparison to control sample.

In the current study, microcapsules were synthesized from urea-formaldehyde and characterized. Subsequently, these synthesized microcapsules were encapsulated with a commercially available diglycidyl ether of bisphenol A (DGEBA) epoxy resin; SC-15 part A and part B separately and characterized to study viability of these microcapsules as healing agents for self-healing in fiber reinforced composites. 10% microcapsules contained fiber reinforced composite samples were prepared for the investigation of healing efficiency. After composite fabrication with hand layup, samples were impacted at different energy levels and damaged area investigated with thermography. After 48 h, second impact was introduced at sample position with 10 J higher energy compared to first impact for the evaluation of healing performance.

2. Experimental

2.1. Materials

Materials used in the current study for microcapsule synthesis were urea, formaldehyde, styrene, hydrochloric acid (HCl), sodium hydroxide (NaOH), ethylene maleic anhydride (EMA), ammonium chloride (NH_4Cl) and resorcinol, which were all purchased from Sigma–Aldrich. DGEBA epoxy resin SC-15 part A was used as healing agent and acquired from Applied Poleramics Inc. Commercially available plain woven E-glass fabric (oriented in two directions, warp at 0° and fill at 90° respectively) was used as the reinforcement in composite and purchased from Fiber Glast Development Corporation. Density of the E-glass fabric was 2.58 g/cm^3 with a single fiber diameter of 14–16 μm. Fiber surface was sized with silane agent for a better compatibility and better adhesion between matrix and fiber.

2.2. Synthesis of microcapsules

Custom designed reaction vessel was used in the synthesis of empty and epoxy-filled microcapsules. The reaction vessel consists of two reaction chambers, one contained monomeric solution with 63.5 mm diameter low-shear mixing propeller and the other contained healing agent during synthesis. Fig. 1 shows an image of the reaction vessel. Synthesis of microcapsules used in the study was done by employing the method used in a study by Brown et al. [6] with slight modifications. At room temperature, 2.5% EMA solution was prepared by using