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Recovery and improvement in low-velocity impact properties of e-glass/epoxy composites through novel self-healing technique

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We report the recovery and improvement in low-velocity impact properties of e-glass/epoxy composites achieved through embedding self-healing agent (SHA) filled hollow glass fibers (HGFs). At first, catalytic technique was used to fill bonded HGFs with SHA. The HGFs were then laid on e-glass fibers and the laminates were fabricated using vacuum assisted resin infusion molding (VARIM) process. Low-velocity impact tests at two different energy levels were conducted multiple times in the closest proximity to determine the healing efficiency. Results showed significant improvement and recovery in impact properties with 53.6% gain in peak load after second impact in SHA filled HGFs samples in comparison to control samples. A significant gain in energy to peak load was also found in SHA filled samples with 86.6% improvement over control samples. Optical microscopy images of SHA filled HGFs samples showed filling of cracks developed after impact. A distinct damage behavior was observed in control and HGFs embedded samples.

1. Introduction

Self-healing of materials, such as glass, polymers, and concrete, has been investigated in order to extend the service life of these structures [1–4]. In most of these investigations, the healing process involved human intervention and thus the materials were not able to heal autonomically. In recent years, polymer composites have been attractive candidate to introduce the autonomic healing concept into modern day engineering materials. Even though several methods have been utilized in self-healing of polymeric materials [5–9] and fiber reinforced polymers [10–15], the concept of repair by bleeding of enclosed functional agents remains a challenge and has gathered wide attention in the scientific community.

The first use of hollow glass fibers embedded in a composite laminate was suggested by Bleay et al. [11]. In their study, filled hollow fibers with a resin were released into the damaged area when the fiber was fractured. A two-part epoxy resin was used as the repair medium. The two components was diluted with solvent and infiltrated into different plies of a composite based on Hollex S2-glass fiber. Even though the method needed no manual intervention for healing damages, efficient recovery of matrix strength was observed only at elevated temperature. More recently, several self-healing unidirectional glass fiber composites have been developed [13–17]. Trask et al. [18] placed self-healing plies within both glass fiber/epoxy and carbon fiber/epoxy laminates. They investigated quasi-static and impact properties and confirmed self-healing. In the work performed by Jones et al. [19], it was shown that solid-state self-healing system was capable of healing transverse cracks and delaminations in a composite. Their system involved a thermoplastic healing agent dissolved in a conventional thermosetting epoxy resin. Through Charpy impact testing, cracks were developed in the resin system before it was self-healed. However, the healing was assisted by heating the fractured samples. Woldesenbet and Williams [20] showed that, a considerable portion of the tensile strength can be restored by using single hollow fiber filled with releasable healing agent DCPD in polymer matrix composite. When the crack was initiated and propagated through the composite breaking the hollow fiber, the healing agent flowed out and filled the gap. Polymerization was facilitated when the healing agent contacts the Grubb's catalyst that was coated on the outside surface of the hollow glass fiber. Even though different research groups around the world are engaged in developing self-healing materials, problems of localized healing and damage detection in fiber reinforced composites do still exist in most self-healing systems. Use of temperature or catalyst stayed highly impractical solution for localized self-healing. Moreover, the application of temperature to overall structure for self-healing is impossible particularly in case of undetected

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damages. Also the use of catalyst on the surface of hollow glass fibers may mismatch the mixing ratio that is required to achieve curing.

Low velocity impact testing of composites is a very crucial area for researchers. A major concern that limits the usage of composites is their susceptibility to damage due to impact loading. There are practical situations like tool drops, runway debris, bird strikes, hailstorms, and ballistic loading, which induce considerable damage to the composite structures. Composites are inherently weak in transverse direction, i.e., the stiffness and the strength in through the thickness direction are poor since no fibers are present in that direction. Low-velocity impact is considered potentially dangerous mainly because the damage might be left undetected, as the surface may appear to be undamaged. Understanding the causes for the formation of such damages and improving the damage resistance characteristics of composites are very important. When subjected to impact loading, the energy is absorbed in the form of creation of new surfaces. The failure mechanisms include indentation, matrix cracking, delamination, ply splitting, and fiber fracture [21–24]. This will considerably reduce the residual mechanical properties of the laminate. The worst scenario occurs when the damage is at subsurface levels. It is known that the residual compressive strength, which is the most affected mechanical property, is reduced to almost up to 50% [25–28]. In many situations, the level of impact at which visible damage is formed is much higher than the level at which substantial loss of residual properties occurs. Even when no visible impact damage is observed at the surface (energies below barely visible impact damage, BVID), matrix cracking can occur, and the load carrying capacity of the composite laminates is considerably reduced. Visible damage occurs if an impact is above a threshold impact energy, which depends on the laminate stiffness. Though there have been a number of studies on the impact response of composites, to the best of the authors' knowledge, there are no studies reported in the open literature on the use of SHA filled HGFs to recover/reduce the impact properties of glass/epoxy composites.

In this work, we addressed these issues by using self-healing technique by embedding SHA filled HGFs in e-glass/epoxy composites. Self-healing agent was stored in hollow glass fibers (HGFs) that were embedded within e-glass/epoxy reinforced plastics. During a damage event some of these hollow fibers may fracture, thus, initiating the recovery of properties by 'healing' whereby a repair agent passes from within any broken hollow fibers to infiltrate the damage zone and acts to ameliorate the critical effects of matrix cracking and delamination between plies and, most importantly, prevent further damage propagation. This release of repair agent mimics the bleeding mechanism in biological organisms. Experimental efforts was focused on low velocity impact at time interval of 0 h (initial testing), and after 48–96 h (evaluating localized self-healing) respectively. One of the main aim of this work was to achieve the self-healing without using any catalyst and external triggering source such as temperature or ultraviolet radiation.

2. Materials, manufacturing and experimental procedure

2.1. Materials

2.1.1. Self-healing agent (SHA)

A single component room temperature cure Envirez 70301 resin supplied by Ashland Inc. was used as SHA. Luperox™ DDM-9 MEKP and cobalt octate (cobalt (II) 2-ethylhexanoate, 65 wt.%) supplied by Sigma–Aldrich were used as initiator and accelerator respectively. Envirez resin is a commercially available unsaturated polyester resin containing 70% extracts from renewable materials. The SHA and MEKP mix ratio was 66.6:33.3. Before infusing in HGFs, the SHA was mixed with 1–5% cobalt octate. The mixture was then added to MEKP and left to cure. Samples with 5% cobalt octate was found to cure completely after 4 h whereas the samples with lower

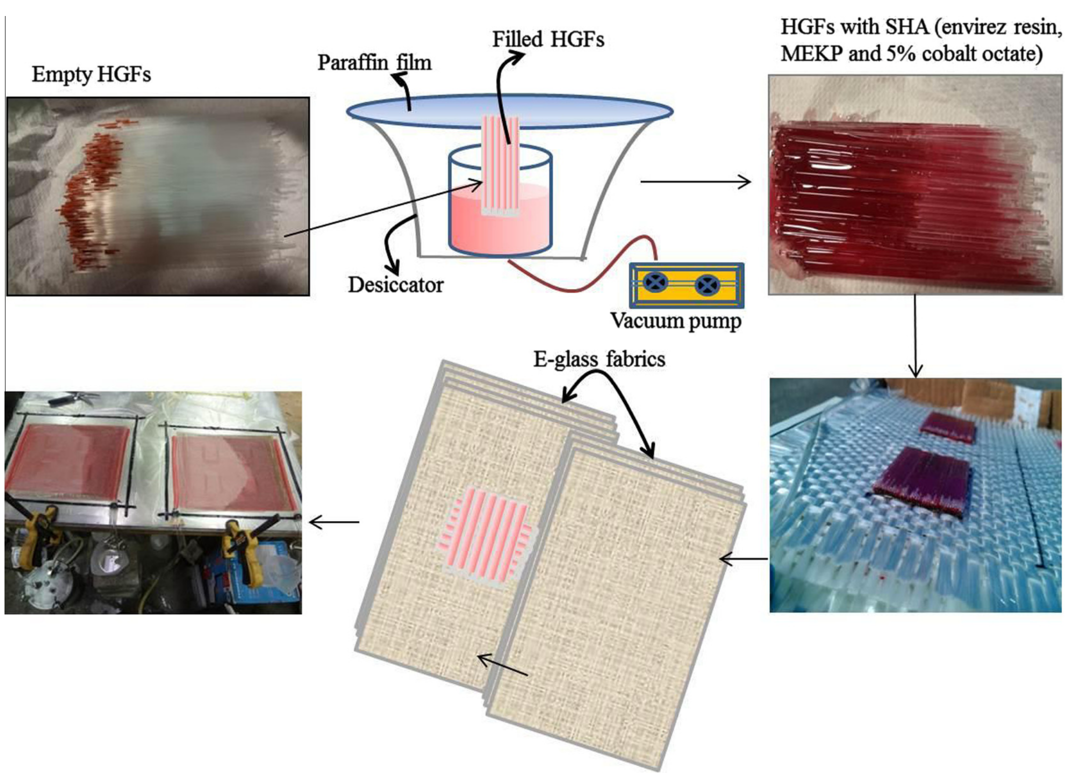

Fig. 1. Filling of HGFs and fabrication of e-glass/epoxy composite.