NOVEL THIN AND FLEXIBLE CARBON NANOTUBE BASED PIEZORESISTIVE SENSING SKINS

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Abstract

Carbon nanotubes (CNTs) are of particular interest for multiscale composites due to their exceptional mechanical and electrical properties combined with recent advances in processing science and characterization techniques. In this research, carbon nanotubes are systematically deposited onto a variety of fabrics such as aramid, polyester and nylon using scalable dip-coating method. The carbon nanotubes form a percolating conductive network, which is piezoresistive, i.e., the electrical resistance of the sensor changes with applied deformation. The change in resistance is much higher than the expected change due to the physical change in dimensions of the specimen (Poisson contraction and axial change in length). The conductivity of the carbon nanotube sensor mainly depends on the CNT-CNT contact resistance in the conducting network. The electrical tunneling gaps increase between the carbon nanotubes, resulting in the increase of electrical resistance. In this research, the ability to detect and distinguish different types of damage in adhesively-bonded hybrid composite-to-steel joints was evaluated through a novel concept of using carbon nanotube sensing skins embedded in the adhesive bond line.

1. Introduction

Advances in the processing science, automation in manufacturing and cost reduction of fiber reinforced polymer composites has increased their usage in a variety of fields such as automotive, aerospace and aviation and civil infrastructure. Increased use of composite materials in complex components combined with manufacturing constraints due to intricate geometries requires joining different composite parts to themselves and, in many cases, to other conventional materials like aluminum and steel. In the aviation industry, adhesives have been used for bonding face-skins to honeycomb core in sandwich constructions and for bonding primary and secondary structural assemblies. Adhesives play a crucial role when fiber composites are used for strengthening of steel structures such as bridges girders [1]. The effectiveness of such strengthening depends on the integrity of the adhesive bond between the steel and the fiber composite.

Adhesive bonding provides higher strength to weight ratios and have about three times higher shearing strength than riveted joints [2]. The joints are more durable because the stress is distributed across the bonded area. Adhesive bonds have superior fatigue resistance, improved visual appearance, and enhanced aerodynamic performance when compared to riveted joints. A critical shortcoming of this type of joint is its inability to be disassembled, which makes the inspection of hidden damage very difficult. Therefore, many researchers have characterized mechanical properties and failure modes of adhesively bonded joints [3]. Studies have also addressed the effect of different parameters such as adhesive selection, surface treatment, bonding pressure and adherend thickness [4].

The durability of a structure is a very important design parameter for many applications. Often, this is dependent on the joint design and how well the joint system is manufactured. Estimating the performance and durability of an adhesively bonded joint is further complicated by the inability to easily detect incipient damage in adhesive. Several non-destructive testing methods [5] such as conventional ultrasonic technique, lamb waves, acoustic emission, thermal and spectroscopic methods exist, but each of these techniques have their advantages and disadvantages and are not used widely.

Because of their unique mechanical and physical properties along with extraordinary thermal and electrical properties, carbon nanotubes are of particular interest in a variety of applications and have been used to create novel material systems [6]. The recent progress in the processing science of nanomaterials has allowed researchers to manipulate the carbon nanotubes at the nanoscale for tailoring the structural properties and functional requirements of multifunctional systems. Carbon nanotubes have been used for enhancing mechanical properties like fracture toughness [7], fatigue life of composites [8]. Baughman et al. [9] were one of the first to report the coupling between mechanical and electrical properties of carbon nanotubes. Li et al. [10] and Baughman et al. [11] highlighted the use of carbon nanotubes as sensors and actuators.

Carbon nanotubes have high thermal and electrical conductivity and a structure similar to that of graphite. Their high aspect ratios (length/diameter) allows to create electrically conductive polymer nanocomposites at very low volume fraction of carbon nanotubes [7]. Researchers have leveraged this capability to develop in situ sensing functionality for damage in fiber composites by creating an electrically percolating network in the matrix [12]. Carbon nanotubes have also been added to adhesives for monitoring the structural integrity and fatigue life evaluation of joints [13]. Recently, carbon nanotubes have been used to create thin skin-like structures for strain sensing and damage monitoring [14] Carbon nanotube coated non-woven fabrics with randomly oriented fibers have been applied for use as sensing skins for existing steel/concrete civil infrastructure [15-17].

This research study presents a novel concept of using carbon nanotube coated sensing skins embedded in the adhesive bond line for damage sensing of hybrid composite-steel joints. These novel sensing skins were fabricated by depositing carbon nanotubes on randomly oriented, non-woven aramid fabric. This non-woven fabric is a like a scrim carrier cloth with very low fiber volume. Modified single-lap joint configurations were prepared. Two kinds of specimens were manufactured: (1) Sensing layer was insulated from steel by a thin layer of adhesive, and (2) sensing layers insulated from steel using a non-woven scrim cloth. Damage sensing and failure modes of these specimens were evaluated using the piezoresistive property of carbon nanotube sensors, that is, mechanical deformation causes its resistance to change. The specimens were tested under quasistatic conditions and acoustic emission sensors were used to validate the in situ damage response.

2. Experimental

Modified single-lap shear joints were used as a model joint system to investigate in situ damage sensing. The model hybrid joints systems were manufactured using steel and carbon fiber composite using a 2-part epoxy adhesive. A carbon nanotube based sensing layer was then embedded in the bond line for monitoring the condition of the joint using. The carbon nanotube sensing layer needs to be electrically isolated from the steel and carbon fiber composite to prevent sensor shorting and galvanic corrosion. Specimens were fabricated with different insulation methods for the sensing layer which promoted different failure mechanisms. As reference, specimens were also manufactured without the integrated sensing layers.

Materials and Manufacturing

The carbon fiber composite was manufactured using T300 carbon fiber pre-impregnated with epoxy resin (Cytec Industries, Woodland Park, New Jersey) using an autoclave. Eight plies of the prepreg were assembled to make a 30.48 mm (12 inch) x 30.48 mm unidirectional laminate. The prepreg was cured in an autoclave at 200°C for 5 hours under a pressure of 0.5 MPa (80 psi). Once cured, the laminate was cut to avoid fiber breakage and delamination. 100 mm (4 inch) long x 25.4 mm (1 inch) wide specimens are made. Low carbon, precision ground steel was machined on a milling machine in 100 mm (4 inch) x 25.4mm (1 inch) to ensure that there were no burrs and all planes were perpendicular. A 2-part epoxy adhesive (Hysol 9309.3NA, Henkel) is used for joining the steel and the carbon fiber composite.

The carbon nanotube sensors were fabricated by depositing carbon nanotubes onto a non-woven aramid veil (20601 50g/m^2 from Technical Fiber Products Ltd.) with randomly oriented fibers, which acts as a carrier fabric for the conductive materials. The aramid veil is porous with low fiber volume fraction. The carbon nanotubes were deposited on the carrier fabric using a commercially available carbon nanotube sizing agent (SIZICYL® – Nanocyl, Belgium). The carbon nanotube sizing agent was diluted with ultra-pure water at a mass ratio of 1:2 (sizing: ultra-pure water). To disperse the carbon nanotubes uniformly throughout the solution, the diluted sizing was mixed in a centrifugal mixer (THINKY® ARM-310) at 2000 rpm for 150 seconds followed by sonication for 30 minutes using an ultrasonic bath (Branson® 1510). The dispersed solution was then transferred to a 1-liter flat-bottomed glass container. The carrier fabric was placed in the solution for 10 min, then flipped and placed back into the solution for another 10 min. After removing from the solution bath the fabric coated with carbon nanotubes was dried in an oven for 30 minutes at 150°C. The dried carrier fabric was allowed to cool and sensors were cut from the fabric to a size of 25.4 mm (1 inch) x 38.1 mm (1.5 inches). The carbon nanotubes coat the individual fibers in the fabric uniformly as shown in Figure 1, creating an electrically conductive network. This network of carbon nanotubes makes the sensor piezoresistive, which enables damage monitoring. To allow electrodes to be applied later in the process, 6 mm (0.25 in) region was masked using a high temperature tape at both ends leaving an area of 25.4 mm^2 (1 $inch^2$) in the bond line.

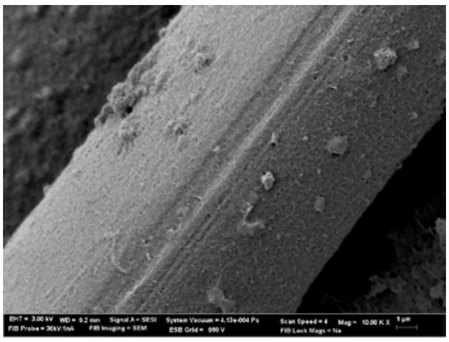


Figure 1. Scanning electron micrographs of a single fiber in the fabric coated uniformly using carbon nanotube sizing

Model Joint System Preparation

A modified version of single-lap joint specimens specified in (ASTM D5868) were made using the carbon fiber composite and the machined steel. The hybrid fiber composite-metal joints were assembled by clamping the composite substrate to a flat surface with the metal substrate clamped above it. A carpenter vice was used along with custom machined spacers for maintaining a bond line thickness of 2 mm. For each specimen configuration, the epoxy adhesive and the corresponding hardener were mixed in a centrifugal mixer at 2000 rpm for 150 seconds. Silicon carbide sandpaper (320 grit) was used to prepare the 25.4 mm^2 (1 inch^2) bonding area followed by cleaning with acetone. The steel was abraded to remove the oxide layer and roughen the surface using a sandblaster with the nozzle at a distance of approximately 50 mm (2 inches) from the steel followed by degreasing with acetone.

Insulation by adhesive: The first step for these specimens was to apply a very thin layer of adhesive on the steel surface and cure it in the oven for 75 minutes at 85°C. Once the adhesive layer was cured, the steel was clamped on one side of carpenter's vice and fiber composite on the other. The sensing layer was placed on the steel specimen and the adhesive was spread uniformly over a 25.4 mm^2 (1 inch^2) bonding region within the spacers and cured at 85°C for 75 min. The vice was tightened just enough such that a consistent bond line of 2 mm was achieved for all specimens.

Insulation by non-woven scrim fabric: An aramid veil (17g/m^3) was used to insulate the sensor. The veil was cut to 25.4 mm^2 (1 inch^2) squares to cover the bonding region. The composite and steel were bonded using the adhesive, with the non-woven scrim fabric and the carbon nanotube sensing layer embedded within the bond line. The scrim fabric was placed near the steel to electrically insulate the sensor. Spacers were used to maintain uniform bond line thickness. The adhesive was cured at 85°C for 75 minutes. Figure 2 shows the schematic of the cross-section of both kinds of specimens.

For both adhesive insulation and non-woven scrim insulation specimens, conductive silver paint (Flash-Dry 04999-AB, SPI. Supplies) was applied at the end regions (previously masked with high temperature tape in the bonding process) of the CNT sensor to minimize contact resistance. Electrical lead wires were then attached to the painted regions of the sensor using a sliver epoxy (40-3900, Epoxies Etc.). Specimens without carbon nanotube sensing layer were also prepared, using the epoxy adhesive to bond the composite directly to the steel. The same custom spacers were again used to maintain a similar bond line thickness.

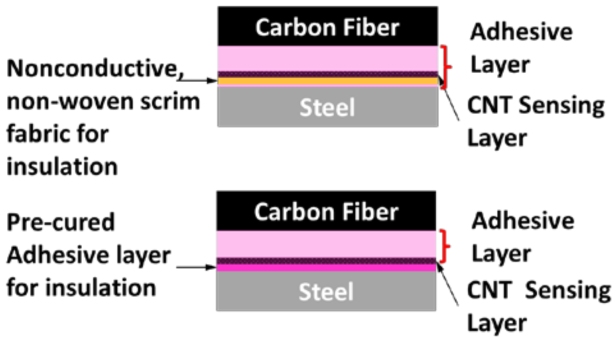


Figure 2. Schematic diagram of the cross-section of the different kind of specimens

Mechanical and Electrical Characterization

A displacement-controlled load frame (Instron 5567) was used to load the specimens under quasi-static tension at a crosshead displacement rate of 6.35 mm/min. A photograph of test configuration is shown in Figure 3. The specimens were tested under monotonically increasing loading (0.25 inch/min), and global deformation was measured using displacement of the crosshead. A Keithley 6430 sub femtoamp remote sourcemeter was used with a constant source voltage of 20 V to measure the electrical response of the sensors during both the monotonic and cyclic tensile experiments. An acoustic emission system (MISTRAS Group, Inc., Princeton Junction, NJ) was also used to record acoustic events during each experiment following the procedure described by Gao *et al.* [18]

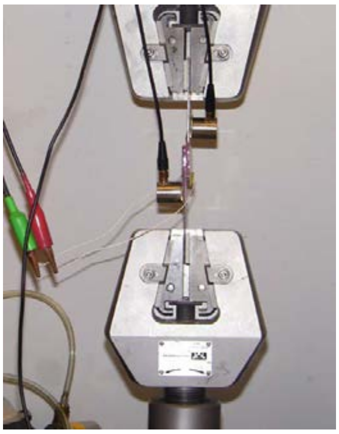


Figure 3. A photograph of the experimental setup

3. Results and Discussion

Figure 4 shows the joint shear strength for all the specimens. The specimens without carbon nanotube sensing layers have average joint shear strength of 14.4 MPa. The specimens insulated with non-woven scrim cloth have no reduction in average joint shear strength as compared to the specimen without the carbon nanotube sensing layer, although there is slightly more experimental scatter. The specimens with the carbon nanotube sensing layer insulated with a thin adhesive layer has an average joint shear strength of 9.7 MPa which is about 31% lower than the specimen without carbon nanotube sensing layer. Ahmed et al. [16] also reported a 23% decrease in ultimate strength for lap shear specimens with sensing layer and an insulative adhesive layer. This could be because of the reduced number of chemical bonds at the adhesive-sensing layer-adhesive interface as compared to that of the rest of the bond line due to partial curing of the adhesive on the steel surface. The interface is in a state of 'weak crosslinking'. Wang et al. [19] reported a significant decrease in tensile, bending and impact properties of specimens with epoxy-epoxy interface where both epoxy layers were cured separately.

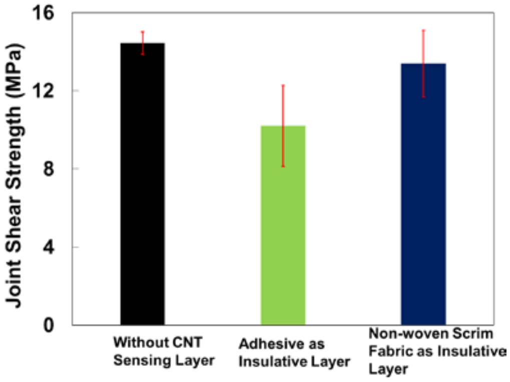


Figure 4. Joint shear strength for specimens with and without carbon nanotube sensing layer

Figure 5(a) shows the shear stress-displacement, resistance and acoustic emission response of the specimens with adhesive as the insulating layer under quasistatic tensile loading to failure. All specimens showed failure at the adhesive-sensing layer-steel interface. Initially, there is very little acoustic activity. The resistance change is linear in this region and is due to the deformation of the sensor. Sharp jumps in the resistance curve correspond with increased acoustic activity at the same time. This is likely due to damage to the carbon nanotube sensing layer. Before failure, the rate of change of resistance of increases. The resistance response has the ability to detect the onset of damage much before the eventual failure of the specimen. Figure 6 shows a photograph of the failed surface of the specimen. The overall change in resistance for these specimens is significantly higher than the non-woven scrim specimens as the sensing layer is subjected to damage.

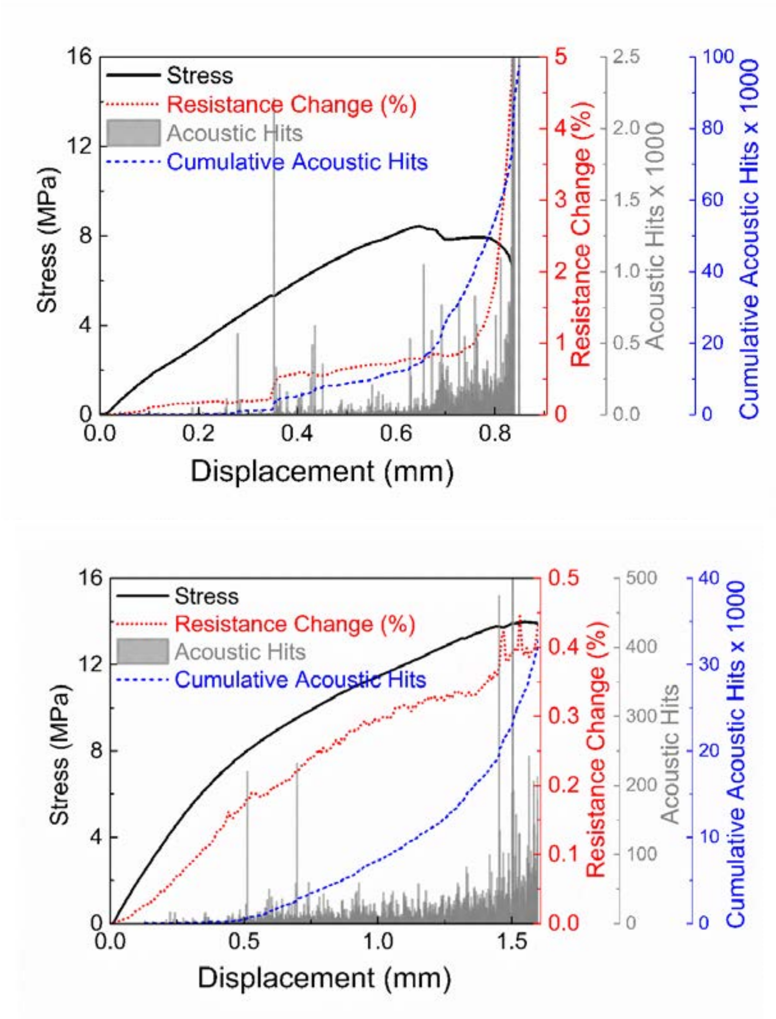
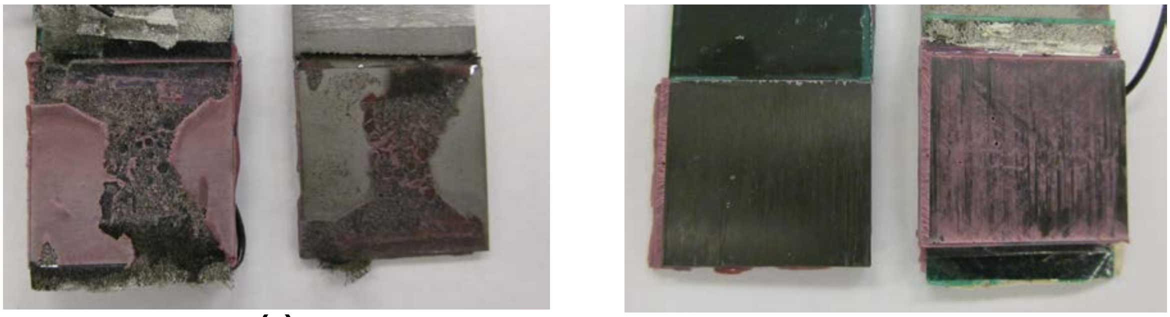


Figure 5. Stress-displacement, resistance behavior and acoustic emission response for a) a specimen insulated with adhesive and b) specimen insulated with non-woven scrim fabric under quasistatic tensile test.

Figure 5(b) shows the response of specimen with a non-woven scrim layer as an insulating layer under monotonic tensile loading to failure. The change in resistance is proportional to the applied stress throughout the loading cycle right until the end where the specimen fails. This increase in resistance is because the sensing layer is subjected to tensile loads. The conductivity of the carbon nanotube sensing layer depends on the nanotube-nanotube contact resistance in the conducting network. The electrical tunneling gaps between the carbon nanotubes increases when the carbon nanotube sensing layer is strained under tension. This increases the contact resistance between carbon nanotubes, resulting in increase of electrical resistance. There is initial debonding at the steel-adhesive interface which is followed by complete failure at composite-adhesive interface, similar to the specimens with-out carbon nanotube based sensing layer and the specimens in [16] without sensing layer. Clearly, the single curing of the bond line, without any interface in the middle of bond line improves the bonding and prevents a premature failure at the adhesive-sensing layer-adhesive interface. The acoustic emission data shows good correlation with resistance change and applied stress curves for all specimens. The spikes in the resistance graph, are likely due to debonding at the steel-adhesive interface correspond accurately with increased acoustic activity.



(a) (b)

Figure 6. Photograph of the specimen after failure (a) at the adhesive-sensing layer-steel interface for the specimen insulated with adhesive layer and (b) at the composite-adhesive interface for specimen with non-woven scrim

4. Conclusion

This study demonstrates a novel method for detecting damage and identifying different failure modes in adhesively bonded hybrid steel-fiber composite joints using novel carbon nano-tube based sensing layer. The carbon nanotube sensors were used for damage monitoring without compromising on the mechanical properties of the adhesive bond. Acoustic emission sensors validate the results from carbon nanotube sensors. Different techniques for electrical insulation of the carbon nanotube sensors using adhesive layer and non-woven scrim cloth were discussed which induce unique failure modes. The adhesive insulation specimens had 31% lower joint shear strength as compared to non-woven scrim specimens and specimens without carbon nanotube sensing later. The adhesive layer specimens had sharp jumps in the resistance behavior and a lot more acoustic activity then the specimen with non-woven scrim. Every different failure mode has a corresponding resistance behavior which can be used to identify the type of damage in the adhesive bond. An innovative, low cost, non-invasive method has been successfully demonstrated for monitoring the condition of adhesively bonded joints. This approach has broad potential for use in future structural health monitoring and for structural sensing.

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