Effects of Surface Characteristics on Mechanical and Dielectric Properties of Adhesively Bonded Carbon Fiber Composites

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ABSTRACT

The rapid rise of fiber reinforced composite usage in aircraft, spacecraft and automobile industries made the proper comprehension of repair and joining of these materials a crucial aspect. Adhesive bonding is one of the most advantageous and desirable joining and repair technique for fiber reinforced composites. However, the heterogeneity of fiber reinforced composites and the complex interfacial nature of the adhesive bonds, makes most non-destructive evaluation and assessment techniques ineffective to assess the state of the bond. Different manufacturing and surface preparation techniques impart different surface characteristics to the adherends, hence proper understanding of the state of bonds is dependent upon the characteristics of the adherend surfaces. In this approach, carbon fiber composite adherends with controlled surface modifications were made into lap-shear test specimens using film adhesive. The effects of surface characteristics such as surface roughness and surface free energy of the adherend surfaces, on bond performance are studied. These surface characteristics are found to be an indicator of bond performance and can even be used to explain failure modes. Moreover, the performance of these bonds with varying surface characteristics are analyzed non-destructively by dielectric spectroscopy and compared with the mechanical performance of the bonds.

INTRODUCTION

Since the introduction of Fiber Reinforced Polymer (FRP) composites, more and

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more metallic structural components are being replaced with composites for lighter, more durable and sustainable structures. Most applications of composite materials are currently focused on aerospace, automobile and marine transportation vehicles. With the advantage of higher specific strength, composite materials introduce complexity into the structure when it comes to monitoring the structural health. This complexity is further amplified when it comes to investigating the joints and bonds of composite structural components non-destructively. The anisotropic nature of fiber reinforced material poses difficulty to assess them using conventional Non-Destructive Evaluation (NDE) techniques. The surface properties of the adherends play a significant role in bond strength. The roughness and wettability between the adherend and adhesive interfaces are the most important factors in determining bond strength [1][2]. The surface roughness ensures mechanical interlocking and the surface free energy ensures the compatibility of the surfaces to result in a strong chemical interaction. Hence proper understanding of bond strength demands an in depth understanding of the effects of such parameters on bond strength.

As bonded composite components have multiple material interfaces, it further results in signal attenuation and interference with common NDE techniques. Ehnhart et al. [3] reviewed viability of the common and advanced NDE techniques for measuring adhesive bond strength and found conventional NDE techniques to be non-reliable and advanced techniques like Shearography, Active Thermography and Laser Ultrasonics to be potential NDE techniques for bond assessments. Even though conventional ultrasonic methods were ineffective in weak bond detection, Yilmaz et al. [4] found focused, high frequency Acoustic Microscopy to be better suited for such task. However, Raihan et al. [5] observed that the dielectric nature of FRPs allows for the detection and assessment of damages accrued in the component using Broadband Dielectric Spectroscopy (BbDS). A state variable approach of assessing composites and bonded structures was described by Reifsnider et al. [6], that makes use of dielectric state variables as parameters of assessment. The method of using the dielectric characterization to assess composite adhesive bonds was also documented and patented by this investigating team [7]. Elenchezhian et al. [8] found a correlation between Dielectric Relaxation Strength (DRS) and composite adhesive bonds, where weaker bonds tend to have higher DRS.

**EXPERIMENTAL PROCEDURES**

For this study, carbon fiber reinforces polymer (CFRP) woven laminates were used as adherends and FM 300 film adhesive was used to form lap-shear bonds. Before forming the bonds the surfaces of the adherends were modified and the surface characteristics were measured. Following the bond formation, Broadband Dielectric Spectroscopy tests were carried out and the shear strength of the bonds were measured using the MTS testing machine. Each testing procedure is described in detail as following:
SAMPLE PREPARATION:

Carbon Fiber Reinforced Polymer (CFRP) laminates were made from 8ply BMS 8-276AE Type-40 12K woven prepgs cured at 350°F and 40psi pressure in Wabash compression molding oven. The laminates were cut into 4in by 1in adherend coupons in a water jet cutter. The adherends were washed, dried and stored in a humidity controlled chamber overnight to evaporate the moistures from the coupons. The adherends were bonded into lap-shear joints using 4 layer FM-300 film adhesive using a closed mold in a pressure and temperature controlled oven. Sample sets of 5 different type of surface characteristics were made. Bonded sample set with adherends made with peel ply were designated as ‘Control’, bonded sample set where adherend surfaces were sanded using P60 grit Aluminum Oxide sand paper were designated as ‘Sanded’, bonded sample set with adherend surfaces contaminated with 20% Polydimethylsiloxane (PDMS) - 80% Hexane mixture, were designated as ‘Contaminated’, bonded sample set where adherend surfaces were both sanded and contaminated were designated as ‘Sanded & Contaminated’, finally bonded sample set where adherends were made without using the peel ply were designated as ‘W/O Peel Ply’.

SURFACE METROLOGY:

Surface features of the adherends have an active contribution to the bond strength via mechanical interlocking between the adherends and the adhesive. Before bonding, the adherend surfaces were profiled on an Ambios 200 stylus profilometer. A 10mg stylus force was used to map a 3mm × 3mm area on each adherend surfaces. The mapped surface was visualized using an open source Scanning Probe Microscopy (SPM) visualization and Analysis tool called Gwyddion [9]. The tool allowed to visualize the 3D surface topography and to measure the mean surface roughness (Sa) of the adherends. The mean surface roughness (Sa) was used as the roughness parameter to compare each of the surface modifications.

SURFACE FREE ENERGY:

The Surface Free Energy (SFE) is used to describe the excess energy available on the surface to form interactions with a different material. The SFE of the adherend surfaces were measured using a Krüss Mobile Surface Analyzer. These measurements were done by measuring the contact angles formed by a polar liquid (water) and a non-polar liquid (diiodomethane) using Double Sessile Drop technique. The contact angle relates the surface free energy of the solid to the surface tension of the liquid, as described by the Young equation [10]:

\[ \gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos \theta \]  

(1)

Where, \( \gamma_{SV} \) represents the surface free energy of the solid, \( \gamma_{SL} \) represents surface energy at the solid liquid interface and \( \gamma_{LV} \) represents the surface tension of the liquid. Using the Double Sessile Drop technique, two liquid of known surface tension is used
to measure the contact angles on the solid surface, then a mathematical method is used to solve equation (1) and calculate SFE of the solid. In this study calculations of SFE were carried out following the OWRK model [11].

BROADBAND DIELECTRIC SPECTROSCOPY:

The bonded lap-shear samples were placed in a faraday cage to carry out a Broadband Dielectric Spectroscopy (BbDS) test on the bonded region of the samples. The BbDS of the bonded samples were carried out by applying a 1V AC electric field through the bonded region of the samples and collecting the material response over 1MHz to 0.01Hz frequency range. Due to the dielectric nature of fiber reinforced composites, upon placement of such material in an electrical field, the material becomes polarized by moving unbounded electrons, ions and re-orienting the natural and induced dipoles with the direction of applied electric field. Each of this polarization mechanisms induces distinctive peaks on dielectric spectra between $10^{-6}$ to $10^{12}$ frequency ranges. The interactions between the applied electric field and the material is governed by the Maxwell’s equations [12]:

$$\nabla \cdot \vec{D} = \rho \tag{2}$$

$$\nabla \times \vec{H} = \vec{j} + \frac{\partial \vec{D}}{\partial t} \tag{3}$$

$$\nabla \times \vec{E} + \frac{\partial \vec{B}}{\partial t} = 0 \tag{4}$$

$$\nabla \cdot \vec{B} = 0 \tag{5}$$

Where, $\vec{D}$ represents dielectric displacement, $\rho$ represents charge density, $\vec{H}$ represents the magnetic field, $\vec{E}$ represents the electric field, $\vec{B}$ represents magnetic induction and $\vec{j}$ is current density.

The dielectric displacement $\vec{D}$ of a material relates the applied electric field $\vec{E}$ via the dielectric constant of the material $\varepsilon_r$:

$$\vec{D} = \varepsilon_r \varepsilon_0 \vec{E} \tag{6}$$

Where, $\varepsilon_0$ is permittivity of free space and $\varepsilon_r$ is the relative permittivity, which is expresses as a complex parameter composed of real permittivity $\varepsilon'_r$ and imaginary permittivity $\varepsilon''_r$:

$$\varepsilon_r = \varepsilon'_r - j \varepsilon''_r \tag{7}$$

The real permittivity represents the energy stored in the polarized material under applied electric field and the imaginary permittivity represents the losses in the polarized material under the same external electric field. Dielectric Relaxation Strength is a convenient dielectric parameter that has been used as a dielectric state
variable for inspecting and monitoring structural components. This variable is defined by the difference of the dielectric variable at the highest and lowest frequency, i.e

\[ DRS = \varepsilon_{r_{low}} - \varepsilon_{r_{high}} \]  

(8)

SHEAR STRENGTH TEST:

The bonded lap-shear specimens were tested for determining the shear strength of the adhesives in an MTS load frame. The tensile shear strength tests were carried out following the ASTM D5868 test standard [13]. The lap-shear specimens had a dimension of 7in × 1in with a 1sq.in bonded area. The bondline thickness of the manufactured bonded specimens were between 0.7mm to 0.8mm. Alignment tabs of 1sq.in were added to each ends of the lap-shear specimens. The alignment tabs were cut from the same laminate as the adherends. The gage length of the assembled test specimens were 5in. The shear strength tests were carried out in displacement control loading at a rate of 13mm/min.

![Figure 1. Lap-shear test specimen schematic according to ASTM D5868.](image)

RESULTS

Among the five sample sets, only three different surface topographies were observed. The ‘Control’ sample set manufactured with a polyester based peel ply had the highest surface roughness and exhibited repeated surface features. The ‘Sanded’ sample set exhibited significantly lower surface roughness with irregular surface
features. The contaminated sample set have the same surface topography and roughness as the ‘Control’. The ‘Sanded and Contaminated’ sample set also had the same surface topography and roughness as the ‘Sanded’ sample set. While ‘W/O Peel Ply’ sample set exhibited the lowest surface roughness and had similar repetitive surface features, as shown in Figure 2.

Based on the measured surface free energy of the adherends with different surface modifications, it is observed that SFE of the ‘Sanded’ sample surface were lower than ‘Control’ samples. Significant drop in SFE were observed with ‘Contaminated’, ‘Sanded and Contaminated’ and ‘W/O Peel Ply’ sample sets, depicted in Figure 3. The standard deviations on the sanded sample sets were higher compared to the manufactured sample surfaces.
The frequency sweep of the dielectric properties of the bonded specimens revealed that the ‘W/O Peel Ply’ sample set had the highest real and imaginary permittivity magnitude, while it was lowest for the ‘Sanded & Contaminated’ sample set. However, for sample sets ‘Control’, ‘Sanded’ and ‘Contaminated’, the dielectric properties appeared to be inconsistent.

Figure 4. Real and Imaginary Permittivity of bonds with different surface modifications.

Figure 5. Shear strength of lap-shear bonds with different surface modifications.
Following the lap-shear strength test (Figure 5), the ‘W/O Peel Ply’ sample set found to be the weakest bond, with only 30% of the ‘Control’ sample set that had no surface modifications. Shear strength test of the ‘Contaminated’ sample set resulted in the highest bond strength.

DISCUSSION

From the investigation of the profiled surfaces of the ‘Control’ sample set, it was observed that peeling off the peel ply following manufacturing, removes the top layer of the epoxy resin system and exposes the fiber surfaces. Hence, the profiled surfaces exhibited the fiber weave topography and the manufactured surfaces had repeated regular features. Similar topography was also observed with ‘W/O Peel Ply’ sample set and due to the absence of peel ply the surface remained covered with epoxy resin and resulted in much smoother surface.

![Figure 6. Mean Roughness vs. Shear Strength.](image-url)
The comparison between the surface roughness and bond strength shows that sanded sample sets had higher bond strength compared to ‘Control’. Indicating improved mechanical interlocking between adhesive and adherends. Similar conclusion is drawn with ‘W/O Peel Ply’ sample set which lacked in proper interlocking due to reduced mean surface roughness.

The SFE measurements shows that sample set with the lowest SFE had the lowest bond strength. However, the highest bond strength was not observed with the adherends of highest SFE, it was observed with the ‘Contaminated’ sample set. This higher bond strength is due to the fact that PDMS contamination reduced surface friction between the adhesive and the adherends, which resulted in improved wetting and proper mechanical interlocking, leading to higher bond strength.

![Surface Free Energy vs. Shear Strength](image)

Figure 7. Surface Free Energy vs. Shear Strength.

The DRS is observed to be highest with the lowest bond strength, where the samples were made without using a peel ply. Hence the conductive fibers were separated from the electrode by a layer of insulative resin system, creating an interface of highly conductive and highly insulative material, leading to electrode polarization. The case of electrode polarization is confirmed by plotting the conductive contribution with respect to frequency, as it is seen in Figure 9, the conductive contribution to the imaginary permittivity is comparatively higher for the ‘W/O Peel Ply’ sample set. The inconsistency observed in Figure 8, between DRS and bond shear strength can be attributed to the improper contact between electrode and the bonded sample due to different roughness of the sample surfaces.
Figure 8. Dielectric Relaxation Strength (DRS) vs. Shear Strength.

Figure 9. Conductive Contribution to Imaginary Permittivity vs. Frequency.
The surfaces of the failed bonds revealed that, bonds with higher strength tends to have cohesive failure and bonds with lower strength tends to have adhesive failure as revealed by the Figure 10. The failure mode of bonds with relatively higher bond strength was partial cohesive-adhesive failure. Upon closer inspection of the highest bond strength adherend surfaces revealed slight fiber tear of the adherends, indicated by the black fibers stuck on the failed adhesive surfaces.

![Figure 10. Failure modes of bonds with different surface modifications.](image)

**CONCLUSION**

From the observations made in this study, it is evident that bond strength depends on both physical and chemical properties of the adherend surfaces. The study also infers that with further study these pre-bond properties can be used as a proper indicator of a good bond and a weak bond. Previously dielectric state variables were found to be a relevant parameter for assessing bond strength, however the inconsistency with different surface modifications stipulates that further investigations are required to have a proper understanding of the dielectric variables for bonded composite materials.

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REFERENCES