Predicting Adhesive Bond Performance Based on Initial Dielectric Properties

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ABSTRACT

In recent years, there has been a widespread growth in the application of composite materials particularly in the Aerospace and Automotive sectors. This is because composite structures are generally comparatively light in weight and provide corrosion and wear resistance as compared to metals or ceramics. Due to the strict fail- safe philosophy of the aerospace industry, the certification approach for current practice in joining composite materials is to thicken the joining areas and to use numerous fasteners which in turn increases the weight and stress concentrations in the structure. The use of adhesive bonding can improve the stress distribution between the composite materials / dissimilar materials and can contribute to a lighter structure. However, there much investigation is yet to be done in this discipline to predict the bond strength and performance using non-destructive evaluation methods.

This paper will focus on an approach to study the mechanical as well as the dielectric properties of an adhesive bond. The dielectric testing is done by using Broadband Dielectric Spectroscopy (BbDS), wherein the dielectric characteristics of the material are analyzed in a wide frequency spectrum. The data obtained by this technique are used to demonstrate the charge transport, the combined dipolar fluctuation, and the effects of polarization occurring between the boundaries of materials. The continuous modifications of the dielectric spectra are due to the changes in the electrical and structural interactions between the particles, shapes, and orientations of the constituent phases of the morphological structure of the material system. Information about the morphologies, impurities/contamination or interaction of the dissimilar surfaces of the pristine bond can be obtained from the initial BbDS properties. The dielectric properties for adhesively bonded composites with different surface adhesion properties have shown promising evidence of predicting the final mechanical performance of the bonded material system. The success and limitations of this approach will be discussed, and needs for continued investigation identified

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Figure 1. Experimental procedure

INTRODUCTION

A heterogeneous material system made of multiple phases such as fibers, defects, matrix and voids is called a composite material. These materials are more light-weight as well as more corrosion and wear resistant than ceramics, metals, or other materials. Currently, composite materials have been used in applications in various industries such as the automotive, medical devices, and primarily the aerospace industries. However, due to the strict fail-safe approach followed by the aerospace industry, it impedes the current breakthroughs in technology. Various bonding technologies have been developed that could improve the bond quality, and increase its lifetime. One of these methods uses fasteners to join two composite materials which increases the weight as well as the cost of the structure. Comparatively, adhesive bonding is an up-and-coming method that reduces the cost and weight of bonded composite structures. Adhesive bonding can also benefit composite systems by reducing the fiber cuts and stress concentrations developed by drilling holes for fasteners. These adhesives are also used to fill the gaps in flanges of an automobile which avoids corrosion thus proving to be useful in the automotive industry. However, it is still a very primitive concept and plenty of research needs to be done for predicting the strength, life, performance and durability of an adhesive bonded joint.

There are numerous factors that affect the mechanical and electric properties of an adhesive joint. These include the bond-line thickness, the time required to cure, the rate of spread or thickness of film, the number of coats applied, and the drying conditions for multiple coating. Standard procedures for testing and characterization should be followed to determine the specimens are consistent. These procedures are established based on the surface roughness, geometry, and the cure time of the specimen.

If any of the preceding factors of the adhesive or the adherent are altered, it sets up a weak adhesive bond between the composite structures. A weak interfacial bond, also known as ‘kissing bond’, between the two adherent surfaces is a type of

weak adhesive bond. This develops when the adherent surfaces are in contact with each other. The detection of such a weak bond has been problematic in the recent years. Additionally, the non-destructive methods (NDT) to observe such defects in an adhesive bond, notably a kissing bond, are very few.

Broadband Dielectric Spectroscopy (BbDS):

Broadband Dielectric Spectroscopy is the study of the interaction of electromagnetic waves with matter. This method has become a prominent tool for determining the dielectric properties of a material. However, there are several different factors that can change the dielectric spectra of a heterogeneous material such as the morphological homogeneity, the interactions of electrical and structural properties between the particles, and the shape as well as the orientation of the combining phases. The dielectric spectra go through various alterations from the moment it is manufactured to the end of its service life. However, during this period, the material experiences gradual damage accumulation.

The dielectric properties of a heterogeneous material are usually calculated in the frequency range of 10^{-6} Hz to 10^{12} Hz. The data generated in this range are related to the molecular and combined dipolar fluctuation, the effects of polarization, and the charge transport emerging inside the material boundaries (Figure 2). The interaction between electromagnetic waves and matter are characterized in detail using Maxwell's equations [2]. The material system is made of numerous polarization mechanisms such as dipolar (orientational), ionic (molecular), interfacial (Maxwell–Wagner–Sillars), electronic polarization and hopping charge polarization [3]. The figure below shows the various polarization types and their effect on the dielectric response as well as its corresponding effective frequency range.

Figure 2. Dielectric response of material constituents at broad band frequency ranges.

BbDS of Composite Materials System

A composite material system comprises of two parts, the matrix and the reinforced fiber material, which when combined will have varying electrical properties. Polarization, in the context of composites, can be defined as the buildup of charge on the material surface while it is being manufactured. Nevertheless, during its service life, the material interface is exposed to numerous variations induced by environmental changes. The charge storage usually occurs at the interfaces and defects of the material [4, 5]. Interfacial polarization occurs at the interface, resulting in the dielectric relaxation of the heterogeneous material which enables the determination of the structure and electrical properties of the material. The dielectric relaxation, is caused by numerous factors such as its orientation relative to the direction of the vector in the applied field, the shape of the inclusions, and the number of interfaces.

Reifsnider et al first proposed using BbDS as a non-destructive defect testing method, to study the changes arising in the dielectric response when defects are formed in the heterogeneous material due to mechanical loading [6,7], shown in Figure 3. The degradation of the composite materials occurs where there is the development and growth of microcracks in the matrix, delamination, fiber fracture, micro-buckling and decoupling of the fiber-matrix interface. Despite typically being used as a NDT method for defect detection, this approach was employed to observe the different modes of damage in the heterogeneous composite systems and establish their individual strength based on their dielectric spectra [8-12]. Figure 4 shows the changes that occur in the dielectric properties when a continuous glass fiber reinforced with plain weave coupons is loaded with a quasi-static tensile load.

Figure 3. The Bode plot becomes constant at lower frequencies with increase in damage

Figure 4. In-situ response of the dielectric property in different zones of damage progression of a composite coupon sample subjected to monotonic loading.

The interface of the adhesive and the adherent surface is subjected to pre-contamination which results in a weak adhesion bond in the bonded composite system. Surface treatment plays an important role in the dielectric response of the adhesive bond. Any change in the surface treatment of the composite will cause changes in the interfacial region of the bond. The focus of this paper is divided into two segments: the quality of adhesive bond based on their surface preparation, followed by the strength prediction of lap-shear bonded samples using BbDS.

EXPERIMENTAL PROCEDURE

Specimen Preparation

The composite laminates were prepared from TORAY T700S/G94 graphite epoxy prepreg. The laminate was 4-ply, with the ply orientation to be $[0]_{4s}$, and was manufactured with the compression molding process at the Institute of Predictive Performance Methodologies. The curing cycle was as provided by the manufacturer.

The laminates were cut into specimens and then bonded together. The thickness of the adhesive joint was maintained to be approximately 0.075 in. Henkel Hysol EA 9394, an aerospace grade adhesive was used for bonding the specimens, cured at the room temperature.

Small 1 inch x 1 inch button samples were prepared in such a way that, they could be tested in our Dielectric setup for their electrical characteristics and quality analysis, with the different electrodes. These button samples were prepared with no defects on one surface, water droplet on one surface, oil drop on one surface, sanded on one surface, and one sample was immersed in water before bonding.

Figure 5. (a) Button specimens (b) Lap-shear specimens

Also, four different types of lap-shear specimen joints were prepared in such a way that the specimens had (i) no defects on one surface, (ii) a water drop on one surface, (iii) an oil drop on one surface, and (iv) small piece of a vacuum bagging film (VB film) on one surface before bonding. These were induced defects to study the dielectric parameters and the strength of the specimen. These were 7 inch x 1 inch, with an overlap of 1 inch.

The surface was prepared as per the ASTM D2093-03 (2011) standard and the lap joint specimen was prepared as per the ASTM D5868-01 (2014).

Electrical Characterization

The dielectric parameters for the adhesive bonds was obtained using the NovoControl Broadband Dielectric Spectroscopy, at the Institute of Predictive Performance Methodologies. The testing was carried out in a closed environmental setup at room temperature using a 0.5-inch electrode as shown in Figure 6. Five readings were observed for each adhesive bonded joint and the average was used for the calculation.

Figure 6. Dielectric testing of adhesive joint in BbDS

Figure 7. Lap-Shear testing of the adhesive bonded sample

Mechanical Testing

The lap shear strength was obtained as per the ASTM D5868 standard, using the MTS Testing machine as shown in Figure 7. The axial breaking load was noted for the specimens and the shear strength was calculated.

RESULTS AND DISCUSSIONS

Adherent surface quality assessment based on dielectric measurements

In general, there are many dielectric parameters such as the permittivity, electric modulus, conductivity and dielectric relaxation strength that are used by various researchers and industries for their analysis purposes. We have used the dielectric relaxation strength in this approach as it involves with the permittivity of the low and high frequency ranges.

Dielectric Relaxation Strength (DRS) is the absolute difference between the permittivity value at the lowest and highest frequencies. The DRS is a dielectric property of the material by which the interfacial polarization of the adhesive bond can be determined. The results prove that the interfacial polarization of the resin based material system is higher when the DRS value is higher.

For the button samples that were bonded together, the electrical characterization was carried out and the DRS values are obtained. The results are shown in Figure 8. From the results, it was observed that the DRS value for the button specimen A is much smaller than the button specimen F, which was contaminated with oil. Hence by this DRS value, the quality of the adhesive bond can be analyzed.

Figure 8. Normalized DRS of adhesively bonded button samples

Relationship of DRS with the Shear Strength

For more accurate results, the lap shear specimens were tested by electrical characterization methods and the mechanical testing methods.

From the Figures 9 and 10, we can observe that as the shear strength of the adhesive bond decreases, the DRS value of the specimen increases. Therefore, a higher DRS value indicates a lower shear (mechanical) strength and vice versa.

Figure 9. Mechanical vs Dielectrical characterization of the Lap-Shear Specimens

Figure 10. Normalized Mechanical vs Normalized DRS of Lap-Shear specimens

CONCLUSION

The surface quality of the adherent of an adhesive bond and the bond quality, lifetime prediction are various challenges that are commonly faced by the aerospace, automobile, marine industries and defense sectors. Hence a new methodology is proposed to identify the interfacial defects by using the non-destructive testing method, BbDS. By analyzing the initial dielectric properties of the material, the surface defects, bond quality and hence the life of the material based on the bond quality can be predicted.

Adhesive bonded composite button specimens are tested for their di-electrical properties. The pattern observed by the DRS values of these specimens indicates the level of contamination of the bond and its quality. Adhesive bonded composite lap- shear specimens are tested for their mechanical and electrical properties. The strength of the adhesive bond is found to be higher with a lower DRS value i.e., as the DRS value of a bond increases, there is decrease in its strength.

By comparing the strength and quality of the bond by DRS value obtained from the BbDS analysis, we can conclude that the BbDS is a promising technique that can be used by the industries and researchers to determine the characteristics of the bond. This phenomenon can also be used during its service life if any exterior damage occurs to the adhesive bond and the material surface.

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REFERENCES

- 1 Kremer F., Schonhals A., *Broadband Dielectric Spectroscopy*, in Springer-Verlag, 2002.
- 2 Huray, Paul G. *Maxwell's equations*. John Wiley & Sons, 2011.
- 3 Tuncer, E., Serdyuk, Y. V., & Gubanski, S. M. (2001). "Dielectric mixtures--electrical properties and modeling." *arXiv preprint cond-mat/0111254*.
- 4 Liu Q. and Reifsnider K. "Heterogeneous mixtures of elliptical particles: directly resolving local and global properties and responses," *Journal of Computational Physics* (2012)
- 5 Baker, J. Adkins J. M., Rabbi F., Liu Q., Reifsnider K. and Raihan R., "Meso-design of heterogeneous dielectric material systems: Structure property relationships," *Journal of Advanced Dielectrics*, Vol. 4, No. 2 (2014), DOI:10.1142/S2010135X14500088
- 6 Fazzino, P. D., Reifsnider, K. L., & Majumdar, P. (2009). Impedance spectroscopy for progressive damage analysis in woven composites. *Composites Science and Technology*, 69(11), 2008-2014
- 7 Reifsnider, K. L., Fazzino, P., Majumdar, P. K., & Xing, L. (2009). Material state changes as a basis for prognosis in aeronautical structures. *Aeronautical Journal*, 113(1150), 789-798 (Silver Award, 2010, Royal Aeronautical Society).
- 8 Raihan, R., Reifsnider, K., Rabbi, F., Baker, J., Adkins, J.M., Liu, Q. "A Dielectric Spectroscopy Method of Directly Assessing Composite Micro-State Evolution During Manufacturing and Deformation" *Society for the Advancement of Material and Process Engineering (SAMPE)*, Long Beach 2013
- 9 Raihan, R., Adkins, J. M., Baker, J., Rabbi, F., & Reifsnider, K. "Relationship of dielectric property change to composite material state degradation." *Composites Science and Technology* 105 (2014): 160-165.
- 10 Raihan, R., Liu, Q., Reifsnider, K. L., & Rabbi, F. "Nano-mechanics Foundations and Experimental Methodologies for Multiphysics Prognosis of Functional Behaviour in Heterogeneous Functional Materials (HeteroFoAM)." *Procedia IUTAM* 10 (2014): 285-293.
- 11 Raihan, R., Reifsnider, K., Rabbi, F., Baker, J., Adkins, J.M., Liu, Q. "A Dielectric Spectroscopy Method of Directly Assessing Composite Micro-State Evolution During Manufacturing and Deformation" *Society for the Advancement of Material and Process Engineering (SAMPE)*, Long Beach 2013
- 12 Raihan, R., Reifsnider, K., Cacuci, D., & Liu, Q. (2015). "Dielectric signatures and interpretive analysis for changes of state in composite materials". *ZAMM-Journal of Applied Mathematics and Mechanics/Zeitschrift für Angewandte Mathematik und Mechanik*, 95(10), 1037-1045.
- 13 Elenchzhian, M., Vadlamudi, V., Banerjee, P.K., Dave, C., Mahmood, A., Raihan, R., Reifsnider, K., (2017). "Quality Assessment of Adhesive Bond based on Dielectric Properties" *Society for the Advancement of Material and Process Engineering (SAMPE)*, Seattle 2017
- 14 Tsangaris, G.M., Psarras, G.C., Kouloumbi, N., "Electric modulus and interfacial polarization in composite polymeric systems," *Journal of Materials Science* 33(1998) 2027-2037
- 15 Marty, P. N., Desai, N., & Andersson, J. (2004). "NDT of kissing bond in aeronautical structures". *In 16th World Conference on NDT*.
- 16 Psarras, G. C., Manolakaki, E., & Tsangaris, G. M. (2002). "Electrical relaxations in polymeric particulate composites of epoxy resin and metal particles". *Composites Part A: Applied Science and Manufacturing*, 33(3), 375-384.
- 17 Psarras, G. C., Manolakaki, E., & Tsangaris, G. M. (2003). "Dielectric dispersion and ac conductivity in—Iron particles loaded—polymer composites". *Composites Part A: Applied Science and Manufacturing*, 34(12), 1187-1198.

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