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Relationship of dielectric property change to composite material state degradation

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ABSTRACT

Polymer matrix composites are widely used in many industries, i.e. aerospace, microelectronics, energy storage etc., because of their unique properties and performance. During their service life, changes of material state caused by deformation and damage accumulation under combined mechanical, thermal and electrical fields requires fundamental understanding to support design of those material systems. Heterogeneous material systems are inherently dielectric as determined by their complex morphology. Dielectric properties of such materials are altered by many factors, e.g., electrical and structural interactions of the particles, and the shape, orientation and distribution of the constituents of the material system. When damage occurs, new phases are created as micro-defects, and grow progressively, interact, and accumulate. The dielectric properties of the composite system also change in a manner that uniquely reflects those details. In the present work we report a non-invasive, in-operando technique to study changes in dielectric properties during progressive damage accumulation in composite materials subjected to mechanical loading.

1. Introduction

Applications of heterogeneous fiber reinforced polymer matrix composite materials require an understanding of the relationships that exist between the functional characteristics of material systems and the long-term behavior of those materials under combined mechanical, thermal, and electrical fields. In general, the heterogeneity of composite materials has been introduced and engineered to achieve specific properties and functions. These material systems are designed in such a way that they can withstand distributed micro damage, e.g., matrix micro-cracking, or multiple fiber breaks, even in the same fiber. Single micro-defects do not individually alter the strength of the composite materials, but when these defects accumulate and interact with each other during the service life, significant changes in the engineering properties of the material system can occur. The taxonomy of these changes as a function of the constituents and loading histories is still incomplete, especially as they relate to the development of final fracture events.

Degradation of composite materials is generally initiated and evolves by microdamage development events, especially matrix microcracking and crack growth, delamination, fiber fracture, fiber–matrix debonding, and microbuckling [1]. Progressive failure in composite materials can be considered to be the statistical accumulation, and subsequent interaction between microdefects. During dynamic tensile loading, acceleration of the damage accumulation can be rate dependent, and can alter the ultimate strength, strain to failure, and energy dissipation capabilities [2]. Hufner [3], Fig. 1, showed that the nonlinear stress–strain behavior of off-axis woven composite materials can be divided into four damage zones, as we will discuss below.

There are many Non-destructive techniques used to detect the complex damage in composite materials. Ultrasonic Testing (UT) and Acoustic Emission (AE) techniques are widely used NDT methods for composite defect detection [4–7]. In ultrasonic NDE, elastic waves propagate through the sample. Flaws/damage cause disturbances in the wave field which can be detected typically by utilizing one the following measurements: time of flight (TOF; wave transit or delay), path length, frequency, phase angle, amplitude, acoustic impedance, and angle of wave deflection (reflection and refraction). Scanning Acoustic Microscopy (SAM) defect detection techniques are high resolution ultrasonic imaging methods that apply a range of frequencies from 30 MHZ to 3 GHz. Figure 2.3 is the schematic of the SAM technique often used to detect delamination in composite laminates [8]. Both of these techniques have specific advantages and applicability; they also have some limitations such as the requirement of point to point inspection, limited penetration depth due to attenuation, limited interpretation due

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to a high variation of composite properties, low contrast due to high attenuation and scattering in composites, and slow methodology. Acoustic Emission (AE) is also a well-established NDE method [4], but due to the multiple reflections in composite materials, wave propagation speed dependence on direction, and attenuation, the "triangulating" of damage in composite materials using AE signals is difficult.

Most NDE techniques require sensors that contact the surface of the composites or are non-contact methods that must have a clear view of the sample surface. But in electrical methods, most of the time, the composite system itself is used as the sensor. Detecting damage using electrical resistivity is a well-known practice for damage detection in composite research [9–20]. Irving and Thiagarajan [20] have studied electrical resistance changes for both static and fatigue testing of CFRP. Although electrical resistance methods have the potential to characterize the damage state during the service life of composite materials, they do not give quantitative information about the defects, orientation of the flaws, crack density, and local morphology, or specific information about damage modes.

Broadband dielectric spectroscopy is used to study the interaction of electromagnetic waves with matter, usually in the frequency range from 10^{-6} to 10^{12} Hz. This dynamic range generally can provide information about the molecular and collective dipolar fluctuations, as well as about charge transport and polarization effects that occur at inner and outer boundaries in the form of different dielectric properties of the material under study. Fig. 2 shows the effect of different charge displacement mechanisms on dielectric response and their corresponding effective frequency ranges. Hence, broadband dielectric spectroscopy can also be used as an effective tool to detect and interpret the damage state of heterogeneous material systems.

Electromagnetic phenomena can be described by four equations constructed by Maxwell [24].

$$\nabla \cdot \vec{\boldsymbol{D}} = \rho \tag{1}$$

$$\nabla \times \vec{\boldsymbol{H}} = \vec{\boldsymbol{J}} + \frac{\partial \vec{\boldsymbol{D}}}{\partial t} \tag{2}$$

$$\nabla \times \vec{\boldsymbol{E}} + \frac{\partial \vec{\boldsymbol{B}}}{\partial t} = 0 \tag{3}$$

$$\nabla \cdot \vec{\boldsymbol{B}} = 0 \tag{4}$$

Here $\vec{\boldsymbol{D}}$ is the dielectric displacement, ρ is the charge density, $\vec{\boldsymbol{H}}$ the magnetic field, $\vec{\boldsymbol{E}}$ the electric field, $\vec{\boldsymbol{B}}$ is the magnetic induction, and $\vec{\boldsymbol{J}}$ is the ohmic current density. In addition to Maxwell's equations, the field must satisfy continuity equations based on the charge density ρ and current density $\vec{\boldsymbol{J}}$ which can be expressed as follows

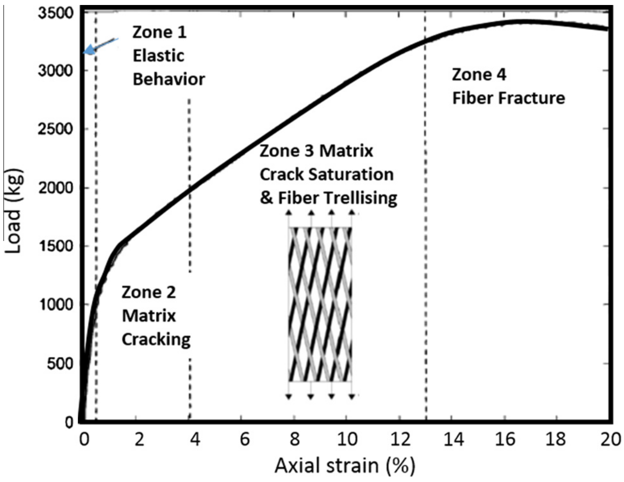

Fig. 1. Off-axis response of woven composite (45° tension) [13].

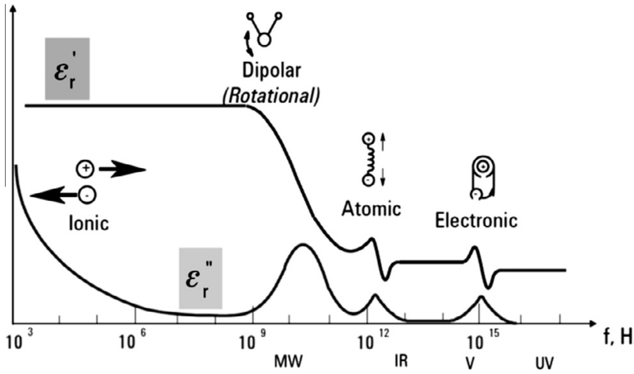

Fig. 2. Dielectric responses of material constituents at broad band frequency range [21].

$$\nabla \cdot \vec{\boldsymbol{J}} + \frac{\partial \rho}{\partial t} = 0 \tag{5}$$

For linear materials the interrelation between the dielectric displacement $\vec{\boldsymbol{D}}$ and electric field $\vec{\boldsymbol{E}}$ can be expressed as

$$\vec{\boldsymbol{D}} = \varepsilon_0 \vec{\boldsymbol{E}} + \vec{\boldsymbol{P}} \tag{6}$$

and polarization can be related to the charge density when no external source is present by the following equation

$$\nabla \cdot \vec{\boldsymbol{P}} = -\rho \tag{7}$$

For a linear relationship between the dielectric displacement $\vec{\boldsymbol{D}}$ and electric field $\vec{\boldsymbol{E}}$ the proportionality constant ε can be used to express

$$\vec{\boldsymbol{D}} = \varepsilon \varepsilon_0 \vec{\boldsymbol{E}} \tag{8}$$

Here the constant ε is called the permittivity of the material which describes the dielectric behavior, normalized by ε_o, the dielectric constant of vacuum. When the Polarization $\vec{\boldsymbol{P}}$ is taken into consideration using (6) and (8) we obtain

$$\vec{\boldsymbol{P}} = \chi \varepsilon_0 \vec{\boldsymbol{E}} \equiv \varepsilon_0 (\varepsilon - 1) \vec{\boldsymbol{E}} \tag{9}$$

Here χ is the polarization coefficient known as the dielectric susceptibility.

The dependence of the complex dielectric function $\varepsilon^*(\omega)$ on the angular frequency ω of the applied electric field and temperature can generally be attributed to the following

(1) Microscopic fluctuations of molecular dipoles.
(2) Propagation of mobile charge carriers by translational diffusion of electrons, holes or ions.
(3) The separation of charges at interfaces which gives rise to an additional polarization. The latter can take place at inner dielectric boundary layers (Maxwell/Wagner/Sillars-polarization) on a mesoscopic scale [25].

Each of the above mentioned processes have specific features in the frequency and temperature dependence of the real and imaginary part of the complex dielectric function.

Fazzino et al. studied polymer based composites and showed that when they develop micro cracking due to mechanical loading, their dielectric response changed dramatically and definitively [22]. They used end-loaded bending, which caused surface