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Dependence of the electrical conductivity of graphene reinforced epoxy resin on the stress level

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ABSTRACT

This paper focuses on electrical conductivity of epoxy based nanocomposites containing graphene particles. We show that adding 0.5 wt% of nanoparticles decreases electrical resistivity of the material more than two orders of magnitude. This effect is modeled with good accuracy using ordinary micromechanical homogenization schemes. Combination of mechanical and electrical tests shows that electrical conductivity of the epoxy-graphene composite depends on the level of tensile stress in the material. To explain this effect we assumed formation of microcracks from the very beginning of the loading process due to very high stress concentration at the particles.

1. Introduction

Carbon reinforced composites have been used for more than half a century in applications where high strength to weight ratio is crucial. Due to the high cost and susceptibility to damage, non-destructive evaluation (NDE) techniques that identify damage within a composite structure are highly valued. One issue with many NDE techniques is that they require the material to be conductive. While the carbon fiber reinforcement is conductive, the matrix material holding the reinforcement material together generally is not conductive making many NDE techniques inapplicable. To the best of our knowledge, the idea to estimate strength reduction of a carbon fiber reinforced composites through electrical resistivity measurements has been proposed by Schulte and Baron (1989). The idea was further implemented in works of Wang and Chung (1997a,b; 1998) Wang, et al. (1999, 2006) Abry et al. (1999, 2001), Park, Okabe, Takeda, and Curtin (2002), and applied to carbon fiber yarn by Wentzel and Sevostianov (2013). AC and DC electrical methods have been extensively studied since then and have been used to study a variety of damage mechanisms, e.g. delamination or matrix cracking, under static and dynamic loading conditions.

Flandin, Cavaillé, Bréchet, and Dendievel (1999) first used nanoscopic conductive fillers with different aspect ratios in a thermoplastic matrix to monitor the applied macroscopic mechanical strain and the damage evolution during loading. Nanoscaled carbon black particles (Kupke et al., 2001) as well as microscaled carbon black particles (Muto et al., 2001) have also been used to modify the matrix of glass-fiber reinforced thermosets. It was shown that external stress as well as apparent mechanical damage can be detected in these multiphase composites via electrical conductivity methods.

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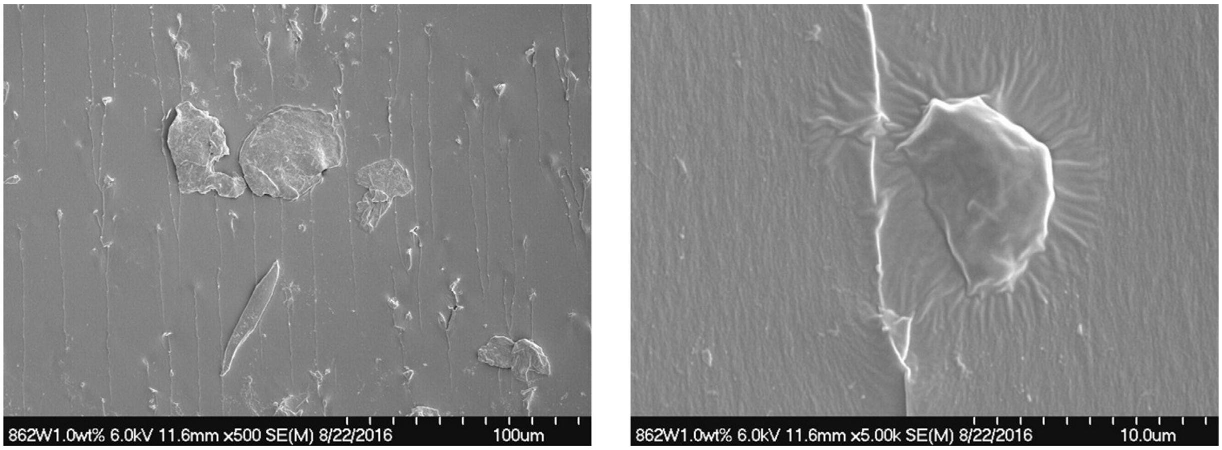

Fig. 1. SEM images of epoxy reinforced with graphene particles.

Böger et al. (2008) developed glass fiber reinforced composites with epoxy matrix containing carbon nanotubes and showed that this material has reasonably high electrical conductivity. Bellucci et al. (2011) studied electrical conductivity of epoxy resin as matrix (Epon 828) containing 0.1%, 0.25% and 0.5% wt carbon black nanofillers or carbon nanotubes. They reported two orders of magnitude decrease of electrical resistivity of such composites as compare to the matrix material. They also showed that epoxy matrix containing carbon nanotubes yields higher values of electrical conductivity than the same matrix containing carbon black particles. Ferreira, Nóvoa, and Marques (2016) provided a review of the state-of-the-art in the area of conducting polymer matrix composites.

Macías, D' Alessandro, Castro-Triguero, Pérez-Mira, and Ubertini (2017) proposed a micromechanical model to explain the conductivity of the polymers containing conducting nanofibers through formation of conducting networks (percolation effect). They assumed that the matrix material is a perfect insulator and did not consider the charge transport across the matric material. However, polymers have low electrical conductivity but it is not zero. Recently, Kang and Snyder (2016) proposed a model for charge transport phenomena in polymers.

In the present paper we focus on electrical conductivity of epoxy filled with graphene particles. We show that adding 1.0 wt% of this filler increase the conductivity of the epoxy more than two orders of magnitude. To explain this phenomena, we use ordinary micromechanical models accounting for the strongly oblate shape of the graphene particles. We also observed dependence of the electrical resistivity on the strain level. This phenomenon can be explained from the point of view of microcracks formation and partial debonding between the matrix and particles due to very high stress concentration at the particles tips. The quantitative explanation of the phenomena, however requires more detailed analysis.

2. Materials and methods

For this study, a two-part epoxy and electrically conductive graphene particles were used. 0.5 wt% and 1.0 wt% conductive filler were used. Epoxy resin, Epon 862, and 'W' amine curing agent were procured from Hexion. Surface functionalized expanded graphite (Miller et al., 2007), TG679, was procured from Adherent Technologies, Albuquerque, NM. All materials were used as received. Resin plaques of baseline Epon 862/W were prepared by mixing 40 g of 862 epoxy resin with 10.56 g of W curing agent. Nanocomposites were prepared through dispersion of either 0.5% or 1.0 wt% of the expanded graphite into graphene layers throughout the base epoxy mixture. Nano-particle dispersion was initiated via a THINKY mixer profile which followed a 4 minute mix/ 4 minutes degas cycle. The mixture was then sonicated for 5 minutes. This process was twice repeated, followed by a cycle in the THINKY mixer, to ensure a final degas. The mixture was poured into a $3'' \times 8''$ Teflon mold and cured 125 °C for 2 hrs followed by a 177 °C hold for an additional 2 hr.

A scanning electron microscope (SEM) was used to observe how well dispersed the conductive filler was. Dispersion of graphene appeared to be uniform. Fig. 1 illustrates the SEM images of epoxy reinforced with graphene particles.

The size of the panels was 75×125 mm. From these panels, ASTM D638 Type IV dog bone specimens were machined. The length of the specimens deviated from ASTM D638 guidelines; however, all samples were machined to consistent dimensions. Fig. 2a shows the specimens.

Resistance measurements were made using a 4-point method with kelvin clip leads. The leads were spaced 10 mm apart and attached to the center region of the gage section. A calibrated IET Labs Model 1693 RLC Digibridge was used to make the resistance measurements. The basic accuracy of the unit is reported as $\pm 0.02\%$. Prior to testing, the unit was tested against a known resistance standard to verify calibration and procedures were followed to zero the unit. This unit was controlled by a custom graphical user interface (GUI) which allowed resistance measurements to be recorded to an Excel spreadsheet. Resistance was measured at a sample rate of 5 Hz.