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Electrical conductivity of unidirectional carbon fiber composites with epoxy-graphene matrix

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ABSTRACT

This paper aims at establishing connections between electrical resistivity and stresses in carbon fiber reinforced epoxy-graphene composites. To avoid the interconnection of multiple parameters, we examined composite specimens consisting of a single strand or tow (12,000 carbon fiber filaments) embedded in an epoxy matrix containing up to 1.0 wt% of graphene (TG679) particles. The specimens show resistance increase with the tensile stress. The characteristic of the dependence varies with the content of graphene particles, tending to be linear as the percentage of graphene content increases. We did not observe any significant changes in the ultimate strength of the composite with an increase of the graphene particles concentration.

1. Introduction

In this paper, we will focus on the electrical resistivity of unidirectional carbon fiber reinforced epoxy-graphene composites, and the dependence of this parameter on the level of tensile stresses. This work builds on the previous results of the present authors on dependence of the electric conductivity on tensile stress in neat carbon tows (Wentzel & Sevostianov, 2013) and epoxy-graphene composites (Wentzel, Miller, & Sevostianov, 2017).

Carbon reinforced composites have been used historically in applications where high strength-to-weight ratio is crucial. As the technology of composite materials has advanced, so has the utility of these materials. Carbon composites are now being used for more than just their mechanical strength: they are also being used for their electrical properties, since carbon fibers and graphite particles are good electrical conductors. Due to that, many results have shown that it is possible to use such composite materials as self-diagnostic materials.

Some of the first research performed was to monitor damage and stress level in continuous carbon fibers (see, for example, Wang & Chung, 1997a, 1998). 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, Schulte, & Schüler, 2002) as well as microscaled carbon black particles (Muto et al., 2001) have also been used to modify the matrix of glass-fiber reinforced thermosets; this research also showed that external stress as well as apparent mechanical damage can be detected in these multiphase composites via electrical conductivity methods. Böger, Wichmann, Mayer, and Schulte (2008) developed glass fiber reinforced composites with epoxy matrix containing carbon nanotubes (CNTs), and showed that this material has reasonably

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high electrical conductivity. More recently, graphene, carbon black, and short carbon nanofibers attracted attention of many researchers in this area. It has been shown, for example, that when graphene fills the insulating polymer matrix, a conductive polymer composite results (Kuilla et al., 2010; Bellucci, Coderoni, Micciulla, Rinaldi and Sacco, 2011; Galpaya et al., 2012). Such composite materials generally exhibit a non-linear increase of the electrical conductivity as a function of the filler concentration. A theoretical study by Xie, Liu, and Li (2008) predicted that graphene is more effective for conductivity improvement than competing nanofillers such as CNTs because of their large specific surface area. These findings were subsequently confirmed by experimental measurements of Bellucci et al. (2011). Ferreira, Nóvoa, and Marques (2016) and Khanam, Ponnamma, and Al-Madeed (2015) provided reviews of the state-of-the-art in the area of conducting polymer matrix composites.

Electrical conductivity of graphene/polymer composites is usually explained by low percolation threshold, when the conductive filler forms a network leading to a sudden rise in the electrical conductivity of the composites. Very often, the conclusions about percolation are made based on electrical conductivity measurements. Stankovich et al. (2006), for example, synthesized graphene/polystyrene composites and, based on electrical conductivity measurements, reported a low percolation threshold at 0.1 vol% of graphene. However, they were unable to observe it by microscopy analysis (similarly to Wentzel et al., 2017).

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. While polymers do have low electrical conductivity, it is not zero. Recently, Kang and Snyder (2016) proposed a model for charge transport phenomena in polymers. It was shown that adding 1.0 wt% of graphene filler increase the conductivity of the epoxy more than two orders of magnitude. This phenomena was quantitatively described using mean-field homogenization models, which accounted for the strongly oblate shape of the graphene particles; that study also observed that the electrical resistivity depends on the strain level. It allows one to use ordinary micromechanical homogenization technique to properly predict overall electrical conductivity in epoxy-graphene composites (Wentzel et al., 2017). This model was then validated by Govorov, Wentzel, Miller, Kanaan, and Sevostianov (2018) on specimens subjected to compressive stresses: the researchers observed reversible changes in conductivity only at the very beginning of the loading process, followed by a period of minimal resistance changes with increasing compressive loading.

In the present paper, we will focus on electrical conductivity of a composite consisting of uniaxial carbon fibers embedded in an epoxy-graphene matrix (three-phase composites). We will demonstrate that adding 0.5 wt% to1.0 wt% of graphene filler increases the conductivity of the epoxy-carbon fiber composites while not changing the ultimate tensile strength. This shows potential to allow for electrical change methods to be applied to monitor stresses in the composite material, without affecting that material's strength. We will also propose a simple model to estimate the filament stress at failure and to estimate the number of surviving filaments prior to the failure event.

2. Materials and methods

Samples were made using TORAY T1000 continuous carbon fiber filament and a two-part epoxy matrix. The nominal filament diameter of T1000 carbon fiber is 5 µm, the electrical resistivity is $1.4 \times 10{-}3$ Ω*cm, and the ultimate tensile is 3,040 MPa (Composite) and 6370 MPa (Filament). The test was designed to study one factor, conductive filler percentage by weight. The levels of conductive filler were; no conductive filler, 0.5 wt% TG679 and 1.0 wt% TG679. Epoxy resin, Epon 862, and 'W' amine curing agent were procured from Hexion. Surface functionalized expanded graphite, TG679, was procured from Adherent Technologies, Albuquerque, NM. All materials were used as received. Nano-particles were dispersed as graphene layers into the matrix material. To control how well dispersed the conductive filler was, the cured graphene/epoxy specimens were examined using scanning electron microscope. We did not observe any clustering or percolation of the graphene flakes (Fig. 1).

T1000 material was loaded onto a stand-alone tensioner set at 22.25 N. The tow was fed through a heated resin bath and drawn through a die set to a specific diameter. Once through the die, an additional layer of resin was added to the outside of the strand and fed through a length of shrink tube. Grips were placed on both sides of the strand, and weight was attached to one grip. Tension was released; then the strand was cut and hung on a rack by one of the grips, with the weight allowed to freely hang on the opposite end. A heat gun was used along of the shrink tube and excess resin was removed. The strand was then hung and cured in an oven. Once cured, the shrink tube was removed from the stand. Fiber volume fraction was controlled between 50%−60%. Test articles are show in Fig. 2.

The tensile load was applied to the specimen using a precision electromechanical drive system consisting of the stepper motor, gear reduction, and linear actuator drive assembly. A DC stepper motor with 200 steps per revolution, in conjunction with 10:1 gear reducer driving a precision bearing linear actuator, allowed for the discrete control of the tensile force down to 0.02 N per step. The applied tensile load was recorded with an S-Type load cell with a 0 – 4500 N range. The load frame uses a model SM-1000 load cell manufactured by Interface Inc. A typical load profile is illustrated in Fig. 3.

Samples were loaded into a load frame using a grip adapter. The grip adapter's function was to transfer the axial tensile load to the strand coupon while minimizing any lateral load transfer stemming from vertical misalignment between the upper and lower ends of the strand coupon. Load transfer was accomplished using a universal-joint grip adaptor designed