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Heterogeneously structured conductive carbon fiber composites by using multi-scale silver particles

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This paper reports a new approach to enhance the through-thickness thermal conductivity of laminated carbon fabric reinforced composites by using nanoscale and microscale silver particles in combination to create heterogeneously structured continuous through-thickness thermal conducting paths. High conductivity of 6.62 W/(m K) with a 5.1 v% silver volume fraction can be achieved by incorporating these nanoscale and microscale silver particles in EWC-300X/Epon862 composite. Silver flakes were distributed within the inter-tow area, while nanoscale silver particles penetrated into the fiber tows. The combination of different sizes of silver fillers is able to effectively form continuous through-thickness conduction paths penetrating fiber tows and bridging the large inter-tow resin rich areas. Positive hybrid effects to thermal conductivity were found in IM7/EWC300X/sliver particle hybrid composites. In addition, microscale fillers in resin rich areas showed less impact on tensile performance than nanoscale particles applied directly on fiber surface.

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1. Introduction

Advanced composite materials have been widely used as structural materials for aerospace, military, and industrial applications due to their high stiffness and strength to weight ratios. Currently, low thermal conductivity of composites restricts their ability to replace metallic structures involving thermal management functions; such as the leading edges of supersonic aircraft wings, the inlet or exhaust areas of gas turbine engines, lightweight heat exchangers, electronics packaging materials, hydraulic pump enclosures and electromagnetic interference (EMI) enclosures [1]. In many cases, internal heat needs to be effectively dissipated from these systems and high through-thickness thermal conductivity (TTTC) is essential for using advanced composite materials in these applications [2–4].

Carbon fiber is widely used reinforcement in advanced polymer composites due to its superior mechanical and physical performances. Polyacrylonitrile (PAN)-based fiber and pitch-based fiber are two of the types most commonly used. They have different mechanical and thermal properties which directly impact the performance of their composites [5]. PAN fibers possess high strength and relatively low modulus, while pitch-based fibers have higher modulus and lower tensile strength [6,7]. Pitch-based fibers have higher thermal conductivity than PAN-based fibers due to their higher degree of graphitization. Both types of carbon fibers possess higher thermal conductivity in its axial direction than the transverse direction due to long and continuous crystal structures existing along fiber axis direction to promote phonon transport. As a result, laminated composites show a higher in-plane thermal conductivity along fiber axial direction than through-thickness directions transverse to the fiber axis. The absence of fibers in the through-thickness direction and insulating resin rich areas between fiber tows and layers further result in low through-thickness thermal conductivity. Although Schuster et al. [8] and Sharp et al. [9] achieved a noticeable increase in through-thickness or out-of-plane thermal conductivity by using three-dimensional fiber reinforcements, enhancing the through-thickness thermal conductivity of widely used laminated composites is highly desired. Silver particles [10,11], copper particles [12], carbon black [13], carbon nanotubes [14–16], aluminum powder [17], aluminum nitride [18] or combinations of different particles [19,20] have been tested to improve the thermal conductivity of polymer materials, and some have also been applied in the fiber reinforced composite materials as fillers in matrix. The TTTCs of the composites produced with these fillers were no more than 3.0 W/(m K) in thermal conductivity. The interlaminar resin-rich layer has proven to be the major reason for low through-thickness thermal conductivity. Han

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et al. [3,13] enhanced TTTC by up to 60% by raising the curing pressure and various interlaminar interface modifications through the incorporation of fillers. Simon and Robitaille [21] suggested that transverse thermal conductivity was also influenced by the gap thickness of the inter-tow area which played an important role on composite conductivity prediction. A continuous through-thickness thermal conduction path with good connection between filler particles will improve TTTC of laminated composite effectively. Silver, aluminum, and copper all have high thermal conductivity. Nanoscale aluminum and copper show extremely high reactive activity and are explosive under certain circumstance. Silver was chosen due to its high thermal conductivity, sintering phenomenon and high security.

This research attempted to enhance the TTTC of laminated carbon fabric reinforced composites by utilizing a size synergy advantage of nanoscale and microscale silver particles. The goal is to develop heterogeneously structured and continuous conductive paths of the silver particles along through-thickness direction to effectively increase thermal conductivity as shown in Fig. 1. The effects of silver particle size, concentration, and fiber type on TTTC values and tensile performance were investigated.

2. Experimental

2.1. Materials

Two kinds of the reinforcement fabric were used: IM7 GP-6k plain weave fabric with an areal density of 190 g/m^2 produced by Textile Products Inc., and pitch-based carbon fabric EWC-300X with an areal density of 610 g/m^2 supplied by Cytec. Epon862 with curing agent epicure-W was produced by Momentive Specialty Chemicals Inc. Microscale and nanoscale silver particles were used as fillers. Silver flakes and silver suspension with silver nanoparticles were purchased from Alfa Aesar and Cabot Corporation, respectively. Silver flakes less than 20 μm and silver nanoparticles smaller than 50 nm were used. Silver suspension contains approximately 50 wt% nanoscale silver particles, ethylene glycol, polyvinylpyrrolidone and water. The nanoscale silver particles could be sintered at a temperature higher than 150 °C to increase thermal conductivity.

2.2. Sample design and preparation

The high conductivity composite samples were fabricated by introducing silver particles in several methods to realize inter-tow and intra-tow penetration in carbon fabrics. The silver suspension comprising of nanoscale particles and a solvent was sprayed directly to dry carbon fabrics. The suspension infiltrated the fiber tows by capillary force. After the fabric was impregnated with the required amount silver suspension, the impregnated fabric was further pressed to promote uniform distribution of nanoscale silver particles. The solvent was volatilized at 150 °C under a vacuum for 1 h. The nanoscale silver particles deposited on the surface of each filament fully coating them; and the nanoparticles were sintered at the volatilization temperature providing better connectivity and conductivity.

Silver flakes were evenly dispersed in the Epon862 resin system, and then applied to impregnate the carbon fabric layer by layer by hand lay-up process. The resin permeated into the fiber tows while microscale silver flakes were blocked from impregnation between the fiber tows due to their larger size than the pores between fiber filaments. The flakes mainly distributed at the inter-tow space. Three configurations of silver filled composites were fabricated: microscale silver flakes filled composite (ms-CFRP), silver nanoparticle filled composite (ns-CFRP) and a composite by applying both nanoscale and microscale silver particles in combination (mns-CFRP) via the aforementioned methods. Fig. 2 illustrates the proposed microstructures of the different composite designs. All samples were fabricated using a hot-press process. The samples were cured at 190 °C for 3 h.

2.3. Property characterization

Netzsch laser flash diffusivity tester (Netzsch LFA 457) was used to measure the thermal diffusivity (α). The measurements followed ASTM E1461 guidelines. The samples, measuring 10×10 mm^2, were coated with graphite. A graphite sample with known specific heat and thermal diffusivity was used as a reference. The measured specific heat by LFA457 had a systematic error of approximately 10%. Through-thickness thermal conductivity k can be calculated by:

$$k = \alpha \cdot C_p \cdot \rho \quad (1)$$

where C_p is the specific heat and ρ is the specimen density [22,23].

Tensile property tests were conducted following ASTM D 3039, by using a MTS Landmark machine with a non-contact video extensometer for strain recording. The specimens were measured at crosshead speed of 1 mm/min. E-glass fiber reinforced epoxy tabs were used.

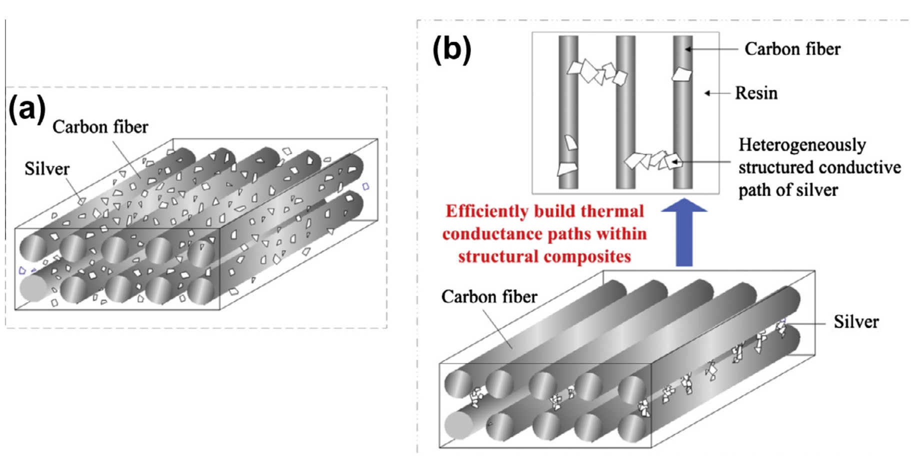

Fig. 1. (a) Conventional homogeneously structured filler-enhanced CFRP; (b) heterogeneously structured continuous conductive paths to improve the through-thickness thermal conductivity of structural composites.