



Improvement of out-of-plane thermal conductivity of composite laminate by electrostatic flocking

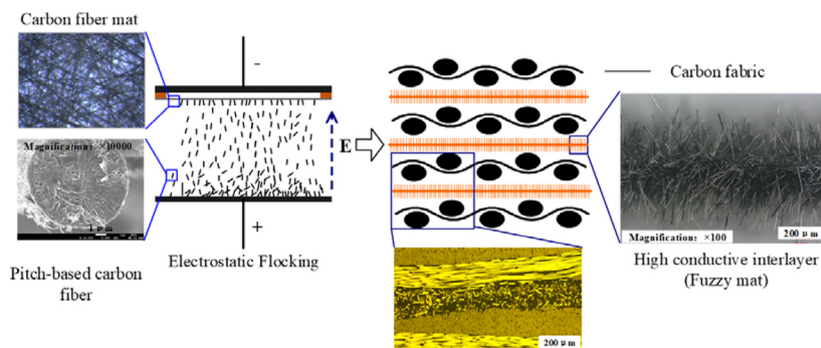
Yafei Sun, Shaokai Wang ^{*}, Min Li, Yizhuo Gu, Zuoguang Zhang

Key Laboratory of Aerospace Advanced Materials and Performance (Ministry of Education), School of Materials Science and Engineering, Beihang University, No. 37 Xueyuan Road, Haidian District, Beijing 100191, China

HIGHLIGHTS

- Electrostatic flocking method was adopted to successfully fabricate thermal conductive fuzzy mat.
- Fuzzy mat effectively improves out-of-plane thermal conductivity by bridging fabric plies of composite laminate.
- An out-of-plane thermal conductivity of 1.2 W/(mK) is achieved, which is increased by 216% compared with control sample.

GRAPHICAL ABSTRACT



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ABSTRACT

Increasing the out-of-plane thermal conductivity of polymer composite laminate is a challenge for applications that require heat dissipation. This paper adopted the electrostatic flocking method to embed different fillers in thin carbon fiber mat to produce a high thermal conductive ply, which was named fuzzy mat. The fuzzy mat was laminated alternatively with two-dimensional fabric to bridge the adjacent fabric plies. The effects of micro-structure and structural parameters of fuzzy mat, as well as the filler type, on out-of-plane thermal conductivity were investigated. The results showed that the fuzzy mat effectively filled the resin rich area in the carbon fabric composite and greatly improved thermal conductivity. Pre-cured and double surface implanted fuzzy mat was beneficial for the construction of thermal conduction path. The out-of-plane thermal conductivities of the composite laminates increased as the content and length of pitch-based carbon fiber fillers increased. The thermal conductivity of composite laminate by introducing 10 wt% fuzzy mat with 300- to 400- μm -long pitch-based carbon fiber filler was up to 1.2 W/(m·K), which was 216% higher than the thermal conductivity of the control sample, which was 0.38 W/(m·K). Moreover, the vapor-grown carbon fiber effectively improved both the out-of-plane thermal conductivity and the interlaminar shear strength.

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1. Introduction

Thermal management is a significant challenge in the fields of aerospace [1], electronic packaging [2–4], phase change energy storage [5], heat sinks [6,7], and so on. High-density integration of high-powered electronic devices requires efficient heat dissipation, and consequently,

^{*} Corresponding author.
 E-mail address: wsk@buaa.edu.cn (S. Wang).

thermal conductive materials are attracting more and more attention. For instance, fiber-reinforced polymer composites have been successfully used in various industrial fields due to being lightweight and exhibiting excellent mechanical performance. However, the low thermal conductivity of these composites limits their use in applications that require heat dissipation [8].

The thermal conductivity of fiber-reinforced polymer composite depends on its microstructure and the intrinsic properties of fiber and matrix [9,10]. The three-dimensional textile composite with through-thickness fibers may achieve out-of-plane thermal conductivity as high as $8.4 \text{ W}/(\text{m}\cdot\text{K})$ at a through-thickness fiber volume fraction of 5.5 vol% [11]. However, the widely used composite laminate has extremely low out-of-plane thermal conductivity because of the lack of a continuous thermal conductive path [12,13]. Moreover, the polymeric matrix possesses very low thermal conductivity and thus acts as a heat barrier in the composite laminate.

Many studies have been carried out to improve the out-of-plane thermal conductivity of composite laminate, including fiber treatment [14,15] and the addition of thermal conductive fillers. High thermal conductive fillers, such as copper [16], silver [17,18] and silicon carbide [19], have been plated on the surface of carbon fiber to construct a continuous heat conduction pathway. Mun et al. [20] constructed polyacrylonitrile (PAN)-based carbon fiber and silicon carbide core-shell structure in epoxy composite laminate, and a thermal conductivity of $0.75 \text{ W}/(\text{m}\cdot\text{K})$ was measured at a filler loading of 80 wt%. Various thermal conductive fillers were also introduced in polymer matrices including metals, ceramics, carbon, and carbides [21,22]. The effects of filler type, shape, contact mode with each other, and synergistic effect [23–25] of different fillers have been investigated. Wrosch et al. [26] applied copper nanoparticles into a polymer matrix, forming a continuous sintered network in the carbon fiber-reinforced polymer composites. Liu et al. [27] distributed graphene in the resin rich regions of carbon fiber laminates, achieving a thermal conductivity of $0.38 \text{ W}/(\text{m}\cdot\text{K})$ at a graphene loading of 0.7 wt%, which was 53.6% higher than that of carbon fiber-reinforced polyetheretherketone (PEEK) composite. Bhattacharyya et al. [28] dispersed carbon nanofiber (CNF) in a phenolic resin, and then impregnated carbon fabrics using this modified resin. The prepared multiscale composite with 1.5 wt% CNF obtained a thermal conductivity of $0.07 \text{ W}/(\text{m}\cdot\text{K})$, which was 36.5% higher than that of carbon/phenolic composite.

Among these high conductive materials, the ones with high aspect ratio, such as whisker, fibrous and lamellar fillers, were found to be the preferred fillers, which were beneficial for forming a continuous heat conduction pathway [29,30]. Therefore, some studies have focused on the resin rich area [31] and established a heterogeneous structure by using high aspect ratio fillers. Yong et al. [32] inserted random carbon nanotube mats between carbon fabric layers, and the transverse thermal conductivity increased from $0.50 \text{ W}/(\text{m}\cdot\text{K})$ to $1.39 \text{ W}/(\text{m}\cdot\text{K})$ at a CNT content of 1.06 wt%. Burger et al. [33] noted that filler alignment may achieve a higher thermal conductivity at a lower content. Wang

et al. [34] prepared a flexible composite film with 17.11 wt% carbon nanotube arrays and obtained an out-of-plane thermal conductivity of $8.2 \text{ W}/(\text{m}\cdot\text{K})$. The out-of-plane thermal conductivity of randomly oriented MWCNT film was reported to be only $1.52 \text{ W}/(\text{m}\cdot\text{K})$ [35]. These observations suggest that promoting filler orientation along the through-thickness direction may lead to higher out-of-plane thermal conductivity.

A great variety of innovative methods have been attempted to align conductive fillers to achieve high thermal conductivity in a certain direction for high conductive composites or thermal interface materials. Guo et al. [36] increased the thermal conductivity of poly(vinylidene fluoride) (PVDF) membrane by 84.5% after introducing and aligning 50 wt% polyaniline nanofibers under electric field. Du et al. [37] and Li et al. [38] utilized the magnetic properties of carbon nanotubes and graphene nanoplates (GNPs) to align them under external magnetic field during composite fabrication. The thermal conductivities of resultant composites were effectively improved. In addition, many approaches utilizing the orientation of conductive fillers have been developed [39]. Zhang et al. [40] constructed vertically aligned graphene nanocomposite by rolling graphene sheets to form cylinder and then slicing it into thin films, and a through-thickness thermal conductivity of $615 \text{ W}/(\text{m}\cdot\text{K})$ was measured. A similar method was adopted to successfully prepare boron nitride composite with excellent thermal conductivity [41]. Besides, Uetani et al. [42] fabricated a vertically aligned carbon fiber scaffold by electrostatic flocking followed by the impregnation of fluorinated rubber solution, and a thermal interface material with a through-thickness thermal conductivity of $23.3 \text{ W}/(\text{m}\cdot\text{K})$ was prepared. All these results proved the significant effect of filler orientation on thermal conductivity. However, effective approach for the manufacturing of fiber-reinforced composites still remains challenge.

In this paper, a new fuzzy mat was developed to improve the inter-laminar thermal conduction of composite laminate, which was prepared by depositing high thermal conductive fillers on a thin fiber mat by the electrostatic flocking method. The effects of fuzzy mat structure, structural parameters, and filler type on the out-of-plane thermal conductivity were investigated to disclose their influencing mechanisms.

2. Experimental

2.1. Materials

GW3031 carbon fiber twill fabric was supplied by Weihai Tuozhan Fiber Co., Ltd., which had an areal density of $240 \text{ g}/\text{m}^2$. The axial thermal conductivity of GW3031 was approximate $8 \text{ W}/(\text{m}\cdot\text{K})$, and its radial thermal conductivity was only $0.7 \text{ W}/(\text{m}\cdot\text{K})$. Thin carbon fiber mat with an areal density of $10 \text{ g}/\text{m}^2$ was chosen as a substrate for depositing high conductive fillers. This mat was purchased from Kaifeng Pengyuan Glass Fiber Products Co., Ltd. Three kinds of high thermal conductive fillers were used including mesophase pitch-based carbon fiber,

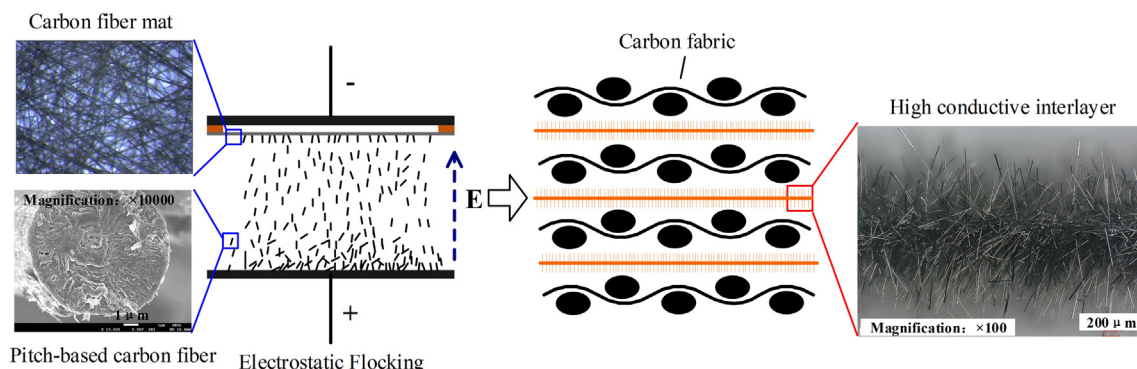


Fig. 1. Schematic of high conductive fuzzy mat by electrostatic flocking.

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