International Journal of Heat and Mass Transfer 118 (2018) 510–517

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Vertically aligned graphene film/epoxy composites as heat dissipating materials

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ARTICLE INFO

Article history:
Received 21 March 2017
Received in revised form 20 May 2017
Accepted 4 November 2017

ABSTRACT

Efficient removal of heat has become one of the most critical challenges in the development of modern microelectronic devices. However, it is hard to dramatically improve thermal conductivity of composite materials even with highly loaded thermally conductive fillers due to the lack of efficient heat transfer paths. In this article, both horizontal and vertical graphene film/epoxy (GF/E) composites were designed and fabricated and their thermal and mechanical properties were studied. Vertically aligned structure constructed of continuous graphene films forms channels for heat removal. The thermal conductivity of vertical GF/E composites reaches 384.9 W m^{-1} K^{-1} at 44 vol% graphene, i.e. a dramatic enhancement of 3570% per vol% compared to the pure epoxy, and representing the highest value of all epoxy based composites. The cooling performance of light-emitting diode (LED) package with the application of GF/E composites is enhanced by reducing the temperature of LED lamp about 20.7 °C compared with the application of pure epoxy. The vertical GF/E composite proved to be a potential material for heat management of LED or other electronic devices.

1. Introduction

LEDs are two-lead semiconductor p-n junction light sources that emit light when activated. However, LEDs convert 75–85% of their electrical power into heat, which reduces their efficiency and makes them less reliable [1,2]. The problem is more serious for high power LEDs, which can be driven at currents from hundreds of milliamperes to more than an ampere [3].

Therefore, it is important to design an efficient thermal system to transfer heat fast from the LEDs to environment. Much work has been done to increase the efficiency by optimizing the heat sink [3–5], adjusting the refrigerant in heat pipes [6] and reducing the heat resistance between the LED chips and heat sinks [7]. Among these methods, improving the thermal conductivity of heat sinks is seen as an inexpensive and simple way which can be used in large-scale industrial production [8]. In the practical application of high-power LEDs, LED chip modules are arranged in arrays and are welded on a specialized substrate. Then the substrate is attached to the heat sink, whose heat dissipation performance influences the lifetime of LEDs.

In recent years, the growing availability of nanoscale carbon fillers with extraordinary thermal properties and low densities has yielded lightweight composites with enhanced thermal conductivity at low filler loading [9,10]. Traditionally, the target thermal conductivity values (>1 W m^{-1} K^{-1}) were met by dispersing high loadings (50–80 vol%) of thermally conductive fillers into polymers. Specifically, graphene and carbon nanotubes (CNTs) exhibit extraordinary mechanical, electrical and thermal characteristics, making them excellent fillers for improving the performance of composite materials. For example, the elastic modulus of epoxy can be improved by several orders of magnitude by filling the CNTs or graphene [11].

Having the ultrahigh thermal conductivity [12,13], these carbon nanomaterials appear to be ideal thermally conducting fillers in polymer. However, the reported thermal conductivities of those carbon fillers/polymer nanocomposites are far lower than the required values [14,15]. Strong phonon scattering at the filler/polymer interface is the main reason, which is due to the weak bonding and phonon mismatch between the carbon nanofillers and the polymer matrix [10]. Conventional one or two-dimensional thermal fillers such as CNTs, graphene and graphite can increase the effective thermal conductivity of composite if their dispersion is good. However, the improvement is strongly hindered by poor filler dispersion for the reason of lack of heat transfer path [16]. So the emphasis lies on making thermal fillers continuous and well-aligned to form path for heat transfer.

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https://doi.org/10.1016/j.ijheatmasstransfer.2017.11.014
0017-9310/

Due to its integrated three dimensional (3D) structure and superior properties, graphene foam was chosen as a promising filler to greatly improve the performance of polymer composites, such as the mechanical, electrical and thermal properties [17–21]. The room temperature thermal conductivity of graphene foam consisting of few-layer graphene and ultrathin graphite was increased from 0.26 to 1.70 W m^{-1} K^{-1} by using different etchants [22]. Thermal interfacial resistance of graphene at Si–Al interface was as low as 0.04 cm^2 kW^{-1}, which is an order of magnitude lower than those achieved with conventional thermal grease and thermal paste based thermal interface materials (TIMs) [23]. By introducing electrostatic flocking to make vertically aligned, high-density arrays of carbon fibers on a planar substrate, thermal conductivity of 23.3 W m^{-1} K^{-1} was achieved with a filler loading of 13.2 wt% [24]. Assembled aligned multilayer graphene/epoxy composite through vacuum-filtration achieved thermal conductivity of 33.54 W/m^{-1} K^{-1} [25]. Unidirectional freezing and drying is another method to make filler continuous and aligned in space [26]. If thermally conducting fillers can be vertically stacked in the polymer matrix, the thermal conductivity of the composites would be greatly improved along the vertical direction [27]. The finite element calculation proved the dependence of heat flux on the orientation of graphene sheets [28]. Due to the flexibility and small size of fillers, vertical structure is hard to maintain. One of solutions is to make vertically aligned graphene film [29], then infiltrated with liquid polymer.

In this work, the graphene film was arranged into vertically aligned structure and then infiltrated with epoxy resin. The composite exhibits excellent thermal properties with the highest thermal conductivity of 384.9 W m^{-1} K^{-1}, which is much beneficial for heat removal of electronics. The composite is also applied for LEDs heat dissipation and satisfactory results are obtained.

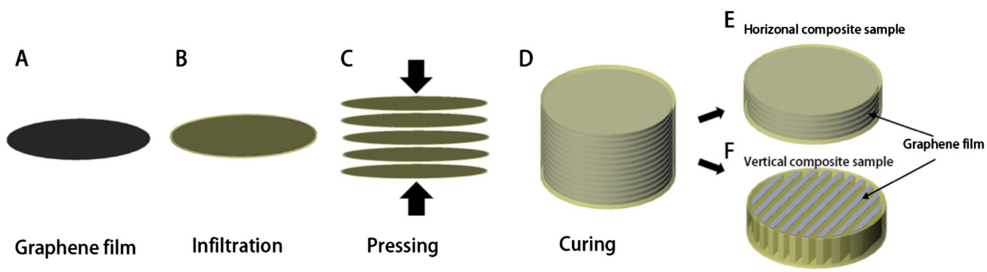

Fig. 1. Schematic of the fabrication procedure of graphene film/epoxy composite.

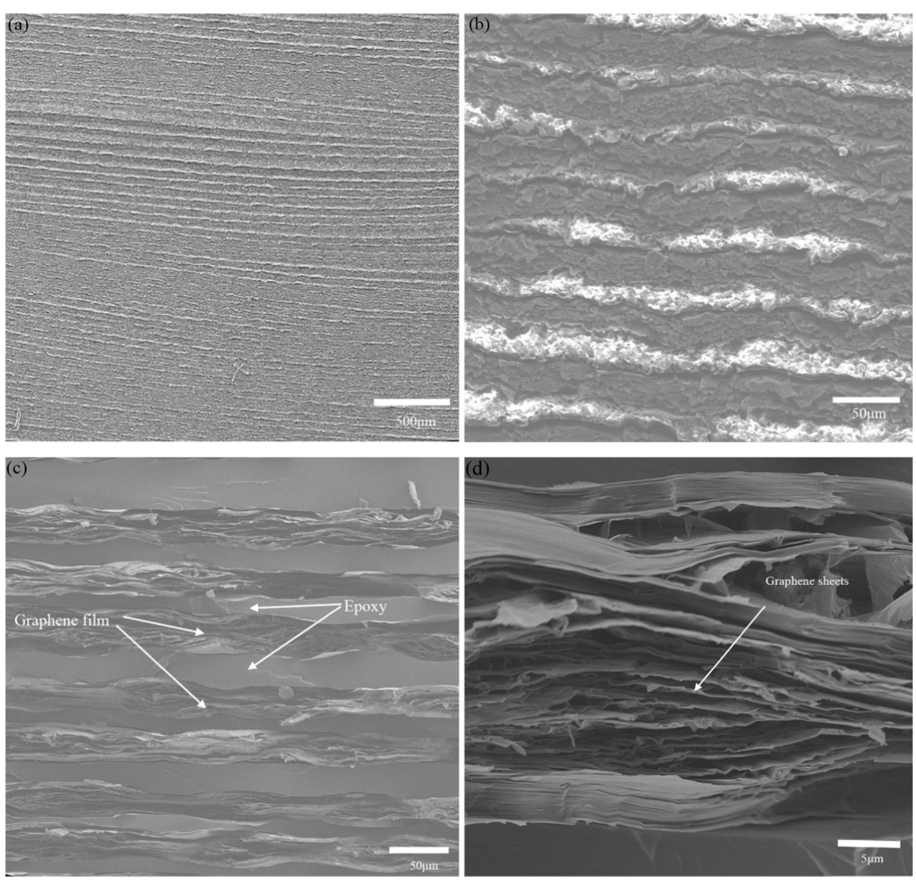

Fig. 2. Morphology of cross-sections of composites after mechanical cutting (a, b) and brittle fracture (c, d).