International Journal of Heat and Mass Transfer 94 (2016) 20–28

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

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

Thermal conductivity enhancement of platelets aligned composites with volume fraction from 10% to 20%

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ARTICLE INFO

Article history:
Received 6 October 2015
Received in revised form 13 November 2015
Accepted 17 November 2015
Available online 28 November 2015

Keywords:
Thermal conductivity
Hexagonal boron nitride
Polymer composites
Magnetic alignment
Mechanical vibration

ABSTRACT

Hexagonal boron nitride (hBN)-filled composites are widely used in electronics for thermal management. In order to enhance the materials heat transport capability, the hBN platelets are expected to be assembled into well-ordered structure. Such structure has been achieved in practice by the magnetic alignment approach. However, this approach is limited to the composites loaded with low volume fraction of platelets (<10%). In this paper, we report the use of combined mechanical and magnetic stimuli to fabricate the well-aligned composites at the volume fraction from 10% to 20%. The platelets in the resulting composites exhibit a high degree of alignment. For instance, in the 10 vol.% composite, the angle of 95.3% of platelets is greater than 45°, only ~5% of platelets falls into the horizontal direction. Thermal conductivity of the composites was investigated experimentally. It exhibited strong correlation with the platelets alignment. The measured thermal conductivity of 10 vol.% aligned composite is 74% higher than that of unaligned composite. Thermal conductivity were also analyzed by a theoretical model. Thermal boundary resistance (R_b), arising at the platelets–matrix interface, was extracted by fitting the measured thermal conductivity to model prediction. R_b is found to decrease with the increase of alignment degree. This study suggests that assembling the platelets into well-ordered structure can greatly enhance the heat transport capability due to the formation of conductive networks and the reduction of R_b.

1. Introduction

Excellent electrically insulating and thermally conductive properties have made polymer-based composites widely used in electronics for thermal management [1–4]. The polymeric composites are predominantly assembled with highly thermally conductive fillers, such as ceramics, metals or metal oxides [5–9]. Among various particles, hexagonal boron nitride (hBN) has been frequently chosen as the reinforcing filler. As schematically shown in Fig. 1a, hBN is a platelet-shaped, high aspect ratio (D/t) particle and possesses high in-plane thermal conductivity (~390 W m^{-1} K^{-1}) [10]. In most thermal management applications, as shown in Fig. 1b, the hBN-filled composite is expected to provide efficient heat flow (Q) from the heat source to heat sink in the direction normal to the composite layer. So the ideal orientation of hBN platelets in polymeric matrix is parallel to the direction of Q (Fig. 1b (I)). Such well-ordered structures can make the composite take full advantage of platelets high in-plane thermal conductivity. On the contrary, the isotropic (Fig. 1b (II)) orientation is much less favorable. Therefore, control of the platelets orientation can enable the enhancement of composites heat transport capability.

The well-ordered structures has been achieved in practice by several approaches, including tape-cast [11], spin-cast [12], shear alignment [13], electrical alignment [14], and magnetic alignment [15–17]. Among these approaches, magnetic alignment has been recently highlighted due to the remote control of fillers orientation and possibility of aligning fillers at arbitrary directions [17,18]. This approach relies on coating the hBN platelets with magnetic nanoparticles, such as Ni, Fe, Co [16] and Fe_3O_4 [15,17]. These modified hBN (mhBN) platelets exhibit a high magnetic response which enables the alignment of them in low-viscosity suspending fluids under linear, uniform magnetic field [15–17]. Then the magnetically-imposed alignment can be fixed by consolidating the suspending fluids.

However, this approach is limited to the composites loaded with low volume fraction (f) of mhBN platelets. When f increases above a percolation threshold, the steric interactions between the platelets will dominate, which hinders the platelets alignment. For instance, applying such approach, Lin et al. [17] prepared the aligned composites at f from 3% to 28%. When f was below 13%, the platelets could be assembled into highly ordered structures in composites. While, as f increases above 13%, significant amount

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http://dx.doi.org/10.1016/j.ijheatmasstransfer.2015.11.045
0017-9310/

Nomenclature

C_p	heat capacity, J kg^{-1} K^{-1}
D	diameter of platelet, μm
d	diameter of iron oxide nanoparticles, μm
f	platelet volume fraction, %
H_0	intensity of the magnetic field, Gs
I	peak intensity of X-ray diffraction
k^*_{33}	effective through-plane thermal conductivity of composites, W m^{-1} K^{-1}
k^c_{ii}	equivalent thermal conductivity along the ii symmetric axis of the composite unit cell, W m^{-1} K^{-1}
k_m	thermal conductivity of matrix, W m^{-1} K^{-1}
k_p	thermal conductivity of platelet, W m^{-1} K^{-1}
k_T	measured through-plane thermal conductivity of composites, W m^{-1} K^{-1}
L_{ii}	geometrical factor
P	population-fraction of the platelets to be above 45°, %
p	inverse of particle aspect ratio
Q	heat flow, W m^{-2}
R_b	thermal boundary resistance at the platelet–matrix interface, m^2 W K^{-1}
t	thickness of platelet, nm
w	rotating frequency, Hz
w_c	critical frequency, Hz

Greek symbols

α	thermal diffusivity, m^2 s^{-1}
μ_0	magnetic permeability of free space, m kg s^{-2} A^{-2}
θ	angle between the composite axis and the local particle symmetric axis, degree
ρ	density, kg m^{-3}
$\rho(\theta)$	distribution function describing the orientation of particles
η	viscosity of matrix, Pa s
χ_{ps}	magnetic susceptibility of the platelet shell

Subscripts

002	peaks of horizontally oriented hexagonal boron nitride
100	peaks of vertically oriented hexagonal boron nitride
11	in-plane direction
33	through-plane direction

of unaligned mhBN platelets were found in composites. In addition, in our previous [15] and Boussaad [16] work, the examples of well-aligned composites obtained by this approach have a maximum f of 9.14% and 10%, respectively.

To overcome the strong steric hindrances between platelets and thus achieve high degree of alignment, one possible method is to provide additional energy to platelets during magnetic alignment. Mechanical vibration is such a candidate as the additional energy, which is often applied for the arrangement of disordered particles [19–21]. Furthermore, the former investigation [22] has shown the possibility in assembling the high concentrations of alumina platelets into ordered architectures using magnetic fields and mechanical vibration. The results show that the mechanical properties of the alumina-reinforced composites can be tuned deliberately. Therefore, it is possible to tailor the thermal property of the platelets-reinforced composites with such strategy.

In this work, we investigated the thermal conductivity of polymer-based composites containing high concentrations of aligned hBN platelets. The outline of this work is as follows. In the beginning, the aligned composites were fabricated using combined mechanical and magnetic stimuli. For the purpose of comparison, the composites were also prepared with the former magnetic approach [15,17]. Then, the cross section of those composites was imaged by scanning electron microscopy (SEM) and the degree of platelets alignment were quantitatively examined by measuring the platelets angles (θ) with the SEM images. After that, X-ray diffraction (XRD) analysis was carried out to further evaluate the degree of alignment. Finally, thermal conductivities of those composites were compared and analyzed in light of platelets alignment degree.

2. Experimental section

2.1. Materials

hBN platelets (AC6041), with an averaged diameter (D) of 5 μm, were purchased from Momentive. The thickness (t) of platelets is estimated to be 250 nm [17], leading to an average D/t of 20. Epoxy resin, supplied by Huntsman, was selected as the polymer matrix. It composes of a low viscosity bisphenol-A based liquid resin (Araldite LY1564) and an amine based hardener (Aradur 3487). The mix ratio is 100:34 by weight. The initial viscosity (η) of the mixture is about 0.27 Pa s at 25 °C. The aqueous-based EMG-605 ferrofluid used to magnetize the hBN platelets was kindly supplied by Ferrotec. It contained 3.9 vol.% iron oxide nanoparticles coated with a cationic surfactant. The average diameter (d) of nanoparticles is 20 nm [15].

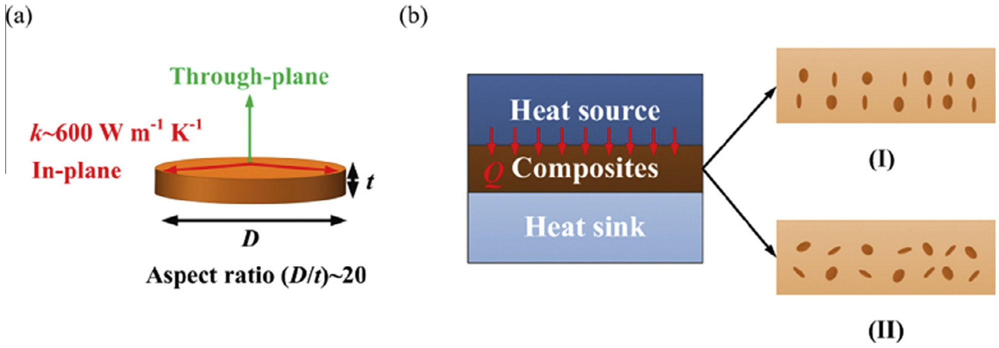

Fig. 1. (a) Representation of hBN platelet geometry and thermal property. (b) Schematic showing the utilization of hBN-filled composite for the heat dissipation from heat source to heat sink. Orientations of hBN platelets in the composite layer: (I) through-plane and (II) isotropic.