



Effect of boron nitride addition on properties of vapour grown carbon nanofiber/rubbery epoxy composites for thermal interface applications



M.A. Raza ^{a,*}, A.V.K. Westwood ^b, C. Stirling ^{c,1}, R. Ahmad ^a

^a Department of Metallurgy and Materials Engineering, CEET, University of the Punjab, Lahore, Pakistan

^b Institute for Materials Research, School of Chemical and Process Engineering, University of Leeds, LS2 9JT, UK

^c Morgan Advanced Materials, Swansea, SA6 8PP, UK

ARTICLE INFO

Article history:

Received 3 July 2015

Received in revised form

10 October 2015

Accepted 16 October 2015

Available online 19 October 2015

Keywords:

Hybrid composites

Electrical properties

Mechanical properties

Scanning electron microscopy

ABSTRACT

This work is focused on developing an epoxy-based hybrid composite using BN and VGCNF, with the main motivation of producing thermally conducting but electrically insulating composite thermal interface materials (TIMs). Various compositions of BN/VGCNF/rubbery epoxy hybrid composites were developed by 3-roll milling. The thermal conductivity of hybrid composites increases with increasing VGCNF content and electrical conductivity decreases with increasing BN content. SEM showed that BN inclusion inhibits VGCNF contacts resulting in more electrically insulating composites. Compression testing showed that BN inclusion produced stiffer composites than those produced with VGCNFs at equivalent loading. The thermal contact resistance of 6 wt.% BN/8 wt.% VGCNF/rubbery epoxy composite was $3.36 \times 10^{-5} \text{ m}^2 \cdot \text{K/W}$ at a bond line thickness of 18 μm . Thermal contact resistance measurements showed that hybrid composites can offer better interfacial thermal transport at thick bond lines and this improves with increasing VGCNF content due to increased thermal conductivity imparted by VGCNF.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

Thermal interface materials (TIMs) are vital for microelectronics packaging as they are responsible for improving interfacial thermal contacts between components, such as microprocessors, and heat sinks, thus ensuring sufficient heat removal from these components [1]. Conventional TIMs are made by dispersing inorganic fillers such as boron nitride (BN), aluminium nitride or silicon carbide in polymer matrices. These are primarily marketed as thermal pastes. Thermal pastes offer superior thermal interfacial contacts but they have issues of pump-out or dry out from interfaces when exposed to thermal and power cycle resulting in increase in thermal contact resistance which threatens microelectronic devices long term reliability [2]. Polymer composites are commonly used as adhesive TIMs since, as well as offering good thermal conductivity, their compliant nature suits gap-filling applications thereby improving contacts between the mating surfaces

and also binding the surfaces to improve mechanical stability [3]. High thermal conductivity and low thermal contact resistance are the most desirable characteristics of TIMs [4].

Recently, carbon nanofillers such as graphene, graphite nanoplatelets, carbon nanotubes and carbon nanofibres have been widely researched as fillers to produce heat dissipating polymer-based composites [5–8] and thermal pastes [4,9,10] that offer great potential for thermal interface applications due to their high thermal conductivity and low thermal contact resistance, respectively. However, high thermal conductivity alone cannot ensure good interfacial thermal conductance. The performance of TIMs depends on many factors such as concentration and morphology of filler and on the wettability, spreadability and adhesion of the resulting polymer composite dispersions on substrates/components, which improve thermal contacts between the mating surfaces [11]. Also, the potentially high electrical conductivity of carbon nanofiller-based polymer composites is considered a drawback in microelectronic packaging. For carbon black thermal pastes, Lin et al. [12] addressed this problem by incorporation of exfoliated clay and fumed alumina into carbon black-based thermal pastes.

Production of hybrid composites seems an easy solution for

* Corresponding author.

E-mail address: mohsin.ceet@pu.edu.pk (M.A. Raza).

¹ Current address: Haydale Ltd., Ammanford, SA18 3BL, UK.

further improving conducting and mechanical properties of carbon nanofiller-based composites. By dispersing different types of carbon nanofillers or combining carbon nanofillers with inorganic fillers in a polymer matrix, a composite with multifunctional properties can be developed. There are several reports in which researchers have reported that hybrid combination of fillers can produce synergetic effects on the thermal, electrical and mechanical properties of composites [13–16].

BN is the most popular filler material for TIMs mainly due to its high electrical resistivity, which is important for avoidance of short-circuiting in certain systems, and high thermal conductivity (280 W/m.K). Like carbon nanomaterials, BN can also have several nanostructures which make it an interesting filler material for polymer composites [17]. Few research articles report thermal conductivity of hybrid composites produced with BN and carbon nanomaterials. Teng et al. [18] produced hybrid filler composites by dispersing functionalized multiwalled carbon nanotubes (MWCNTs) and BN in epoxy resin. They reported that thermal conductivity of 1 vol.% MWCNT/epoxy composite increased with increasing vol.% of BN. A composite containing 30 vol.% BN and 1 vol.% MWCNT had thermal conductivity of 1.9 W/m.K which was ca. 8x higher than for neat epoxy. This increased thermal conductivity was attributed to better conductive networks formed by the hybrid fillers. Pak et al. [19] reported polyphenyl sulphide-based composites produced by dispersing a mixture of BN and MWCNT fillers. A maximum thermal conductivity of 1.74 W/m.K was obtained for a composite consisting of 1 wt.% MWCNT and 50 wt.% BN compared to 1.0 W/m.K for a composite having 50 wt.% BN particles only. The synergetic effect on thermal conductivity of composites was attributed to the formation of three-dimensional thermal transfer pathways between BN and MWCNTs. Ng et al. [20] reported that employing mixed BN and carbon fibre fillers in polybutylene terephthalate (PBT) composites did not enhance thermal conductivity and this was attributed to the low aspect ratio of the carbon fibres which could not help in formation of conducting networks. However, the authors found that incorporation of carbon fibres improved tensile properties and processability of BN/PBT composites.

Vapour grown carbon nanofibres (VGCNF) are very similar to MWCNT in morphology but have larger diameters than MWCNTs. Their low cost and ease of dispersion in polymers compared to carbon nanotubes make them suitable filler for producing heat dissipating polymer composites (TIMs). A large number of publications on VGCNF/polymer composites have appeared in the last decade. Review articles [21,22] give a good overview of fabrication, properties and applications of VGCNF/polymer composites. In our prior work [23], we reported that VGCNF/rubbery epoxy composites gave thermal conductivity of 1.31 W/m.K and 1.8 W/m.K at 15 and 26 vol. % of VGCNF, corresponding to 7-fold and 10-fold increases, respectively, over neat rubbery epoxy. We also reported that these composites, even at a high loading of 26 vol.% of fibres, are very compliant (having low compressive modulus), have good ease of processing, low viscosity before curing and good spreadability as coating or adhesive, which makes them suitable candidates for thermal interface applications. However, these composites have high electrical conductivity which undermines their usefulness in thermal interface applications. The incorporation of inorganic fillers such as BN can decrease the electrical conductivity of VGCNF/rubbery epoxy composites and at the same time can produce synergetic improvements in the thermal conductivity of these composites by developing efficient conducting networks as reported by others [18,19].

The present work reports novel hybrid rubbery epoxy composites produced by co-dispersing an inorganic filler, BN, and a carbon nanofiller, vapour grown carbon nanofibres (VGCNF), in the

polymer matrix with the aim of producing TIMs with much better properties than conventional TIMs by exploiting the ability of BN as a co-filler to decrease electrical conductivity and enhance thermal conductivity. BN/VGCNF/rubbery epoxy composites were produced by 3-roll milling at various loadings of BN and VGCNF. The morphology, thermal conductivity, electrical conductivity and mechanical compression properties of the resulting composites were studied. To evaluate the performance of these composite adhesives as heat dissipating materials i.e., TIMs, selected hybrid composite coatings were studied as thermal interface adhesives according to ASTM D5470.

2. Materials and methods

Hexagonal BN particles with average particle size of 5–10 μm and VGCNF (Applied Sciences, Inc) with diameters of 70–200 nm and lengths from 50 to 100 μm were used as composite fillers in this study (Figs. 1 (a–c)). Further details on VGCNF can be seen in Ref. [5]. Both BN and VGCNF were used as-received. Rubbery epoxy resin (RE) was used as a matrix, details of which can be seen in Ref. [24]. Briefly, rubbery epoxy is formed by mixing polyetheramine, a difunctional primary amine with repeating oxypropylene units in the backbone and a molecular weight of about 2000 (Jeffamine D2000, ex Huntsman corporation) with epoxy resin (Epikote 828 ex Hexion Chemicals) at a ratio of 75/25. Jeffamine D2000 is an amine curing agent, however, it should be noted that in rubbery epoxy the Epikote 828 epoxy resin effectively crosslinks the Jeffamine D2000 because Jeffamine D2000 is present in 3 \times higher quantity than the Epikote 828. The resulting material could therefore be logically termed “crosslinked polyether” but it is commonly known as “rubbery epoxy” because it has a glass transition temperature below normal ambient temperature [25] and significantly lower modulus than highly crosslinked glassy epoxy [24].

The composites were produced using a 3-roll mill by dispersing BN and VGCNF in RE according to procedures described in our previous publication [26]. Briefly, a weighed amount of resin, BN and VGCNF was first mixed with mechanical stirrer at 1000 rpm for 5 min. The resulting dispersion was mixed in the roll mill according to the procedure described in Ref. [26]. Composites were produced with BN content fixed at 6 wt.% while VGCNF content was varied from 8 to 12 wt.%. These total filler contents from 14 to 18 wt.% were selected to produce hybrid composites because dispersions with higher loadings could not be easily coated as thin bond lines between substrates. A composite was also produced with 2 wt.% BN and 12 wt.% VGCNF. Finally, since commercial TIMs are comprised of BN fillers with a loading greater than 40 wt.%, a BN/RE composite was developed as a reference material with a loading of 45 wt.% using the roll mill.

The morphology of the freeze-fractured samples, sputter coated with thin layer of Pd/Pt alloy, was studied in secondary electron imaging mode using a LEO 1530 field emission gun scanning electron microscope operated at 3 kV. The thermal conductivity was measured by hot disk thermal constant analyser (Hot Disk® AB) with a sensor (radius 3.180 mm) which was placed between the two halves of each sample, having thickness of 8–10 mm and cross section of 20 \times 20 mm². The electrical conductivity was measured by placing cuboidal composite samples (~5 \times 5 \times 2 mm³) between two copper electrodes which were connected to a multimeter (Agilent 34401A) for the measurement of resistance. Compression testing of samples was carried out on cuboidal samples using an Instron Universal testing machine (Model No. 3382, load cell 100 kN) at a strain rate of 0.5 mm min⁻¹ [26,27]. These composites were also tested, with cured matrix, as adhesives according to ASTM standard D5470 on a thermal contact resistance measurement rig to quantify their performance as TIMs. The details of the

Download English Version:

<https://daneshyari.com/en/article/820029>

Download Persian Version:

<https://daneshyari.com/article/820029>

[Daneshyari.com](https://daneshyari.com)