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Comparison of Thermal Interfacial Performance of Carbon Nanofiller-Based Polymer Composites

Mohsin Ali Raza*[1], Aidan Westwood[2], Chris Stirling[3], Rafiq Ahmad[1]

[1]College of Engineering and Emerging Technologies, University of the Punjab, Lahore, Pakistan
[2]Institute for Materials Research, SPEME, University of Leeds, LS2 9JT, UK
[3]Morgan AM&T, Swansea, SA6 8PP, UK
*Corresponding author email: mohsinengr@yahoo.com

Introduction

Thermal interface materials (TIMs) are essential components of microelectronics as they improve interfacial contacts between the microchips and heat sinks, thus ensuring sufficient electronic cooling [1]. Thermal interface adhesives are polymer-based composites which improve contacts between the mating surfaces, offer good thermal conductivity and also bind mating surfaces to improve mechanical integrity of the electronics packaging [2]. High thermal conductivity and low thermal contact resistance are desirable characteristics of TIMs [3]. Carbon nanofillers such as graphite nanoplatelets (GNP), carbon nanotubes and carbon nanofibres are being extensively researched as fillers for polymer composites due to their very high thermal conductivity [4, 5]. On the other hand, carbon black (CB)-based thermal pastes have been reported to offer very low thermal contact resistances [6]. Researchers have reported the potential of carbon nanofiller-based polymer composites for thermal interface applications due to their high thermal conductivity [4, 7]. However, high thermal conductivity alone cannot guarantee good TIM performance. The performance of TIMs mainly depends on wt.%, size, shape and orientation of the fillers and on the adhesion, wettability and spreadability of the resulting polymer composite dispersions, which improves thermal contacts between the mating surfaces [8]. The present work reports comparison of thermal interfacial performance of carbon nanofiller-based polymer composite adhesives, measured according to an ASTM standard, D5470, that mimics the conditions prevailing in electronics packages.

Experimental

GNP/rubbery epoxy, GNP/glassy epoxy, CB/rubbery epoxy, CB/silicone and CB/GNP/rubbery epoxy hybrid composite dispersions were produced by mechanical mixing. The details of the production and thermal and mechanical characterisation of these composites have been reported in [9-11]. These composites were tested, with cured matrix, as adhesives according to ASTM standard (ASTM D5470) on a thermal contact resistance measurement rig. The details of the rig and the testing procedure have been demonstrated previously [12]. Briefly, these composite pastes (uncured) were sandwiched between the copper substrates and cured at 125 ºC for 3 h and then placed in the rig for the measurement of thermal interfacial transport properties. The effect of GNP, hybrid combination of CB and GNP, types of polymer matrix and CB on the thermal interfacial performance of the composites was studied and is reported here. The effect of applied pressure, temperature and surface roughness of the substrate on the thermal interfacial performance of these composites is also reported.

Results and Discussion

The thermal contact resistance of GNP/rubbery epoxy composite (containing 25 wt.% GNPs with average lateral width of 5 µm) measured on smooth and rough surfaces and the thermal contact resistance of GNP/glassy epoxy composite measured on smooth surfaces, each as a function of coating thickness are presented in Fig. 1 and Fig. 2, respectively.

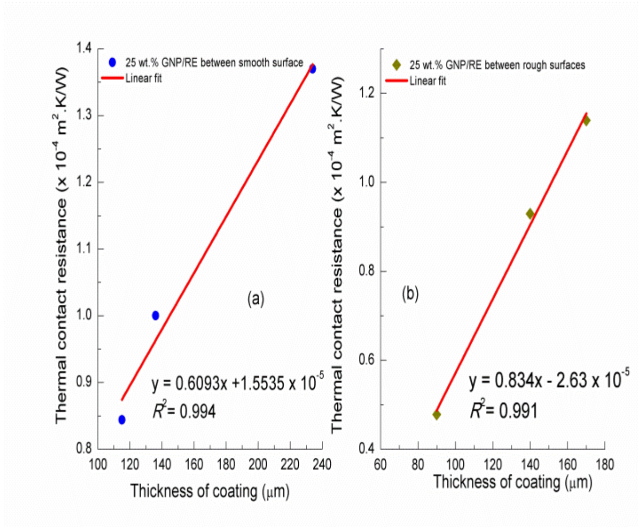

Figure. 1. Thermal contact resistance of 25 wt.% GNP/rubbery epoxy (RE) composite (cured between copper substrates) as a function of coating thickness measured between (a) smooth surfaces (b) rough surfaces at 25 ºC and under a pressure of 0.032 MPa.

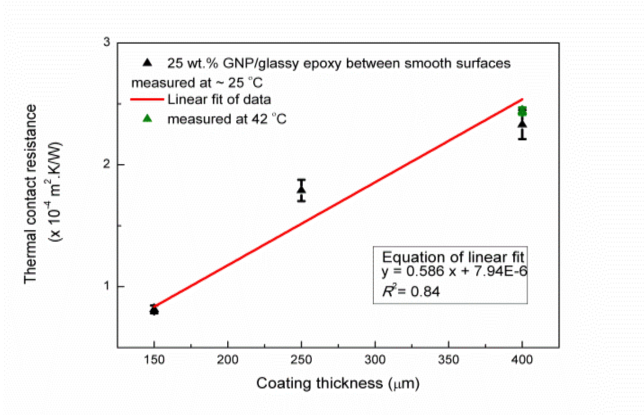

Figure. 2. Thermal contact resistance of 25 wt.% GNP/glassy epoxy (GE) composite (cured between copper substrates) as a function of coating thickness measured between smooth surfaces at 25 and 42 ºC and under a pressure of 0.032 MPa.

The thermal contact resistance of both GNP/rubbery epoxy and GNP/glassy epoxy at a coating thickness of ~150 µm is approximately 1 x 10^{-4} m^2/K.W. The glassy epoxy-based composite, due to its high crosslinking, forms much stronger bonds with the copper substrates compared to the rubbery epoxy (lightly cross-linked). Despite this difference in bonding strength, the thermal transport behaviour of the two composites is similar. However, the GNP/glassy epoxy dispersions could not be applied as thin bond lines due to their very high viscosity compared to GNP/rubbery epoxy [9]. The GNP/rubbery epoxy could give a thermal contact resistance as low as ~ 0.2 x 10^{-4} m^2.K/W at bond line thickness of 25 µm. Fig. 1 also shows that the thermal contact resistance of GNP/rubbery epoxy is much lower on rough surface than smooth surface [11] at bond line thicknesses < ca. 150 µm .

The thermal contact resistance of CB/rubbery epoxy is presented in Fig. 3.

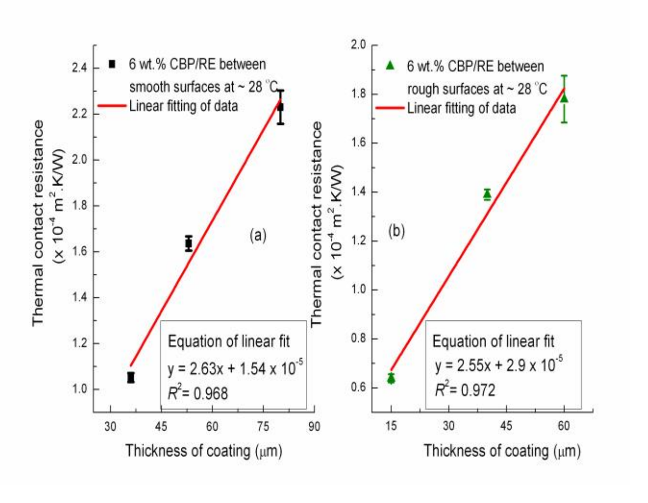

Figure. 3. Thermal contact resistance of 6 wt.% CB/rubbery epoxy (RE) composite (cured between copper substrates) as a function of coating thickness measured between (a) smooth copper surfaces (Ra= 0.03 µm) & (b) rough surfaces (Ra= 0.45 µm) at ~30 °C and under a pressure of 0.032 MPa.

The CB/rubbery epoxy composites can be applied as thin bond lines of ~15 µm. Despite this very low bond line thickness, the thermal transport performance of CB/rubbery epoxy coating is much inferior to that of GNP/rubbery epoxy composite, attributed to ~4 times lower thermal conductivity of the former than the latter. However, the thermal contact resistance of CB/silicone composite (1.18 x 10^{-4} m^2.K/W) as an adhesive was 2x higher than for CB/rubbery epoxy (6.2 x 10^{-5} m^2.K/W) composite at equivalent bond line thickness of 20 µm. Perhaps, the more highly adhesive nature of rubbery epoxy composite on the copper surface contributed to its enhanced thermal interfacial transport.

The thermal contact resistance of 4 wt.% CB/12 wt.% GNP/rubbery epoxy hybrid composite measured between smooth and rough copper surfaces is presented in Fig. 4. The hybrid composite coating displays higher thermal contact resistance than GNP/rubbery epoxy composite coating at equivalent bond line thickness, suggesting no significant benefit for addition of CB in terms of the thermal interfacial performance of the GNP/rubbery epoxy composite adhesive coating. Conversely, the data also suggest that the addition of GNPs into CB/rubbery epoxy composite improves the performance of CB/rubbery epoxy composite. Thus, addition of more thermally conducting filler plays an important role in improving thermal transport performance of adhesives at the interfaces. The thermal contact resistance of commercial 65 wt.% BN/silicone based TIM (EPM 2490, a product of Nusil) was 27 % higher than that of 25 wt.% GNP/rubbery epoxy.

The thermal contact resistance of adhesives was not affected by the application of pressure in the range of 0.032-0.1 MPa, suggesting that thermal interface adhesives can give better longer term performance without risks of leakage compared to commercial thermal pastes.

Conclusions

The thermal interfacial performance of various carbon nanofiller-based polymer composites was studied to explore their potential as thermal interface adhesives for electronics thermal management. The comparative study suggests that GNPs offer potential as fillers for enhancing the thermal interfacial performance of polymer composite adhesives and that thermal interfacial performance of the adhesives depends on having a good combination of their thermal conductivity and their interfacial substrate contact resistance.

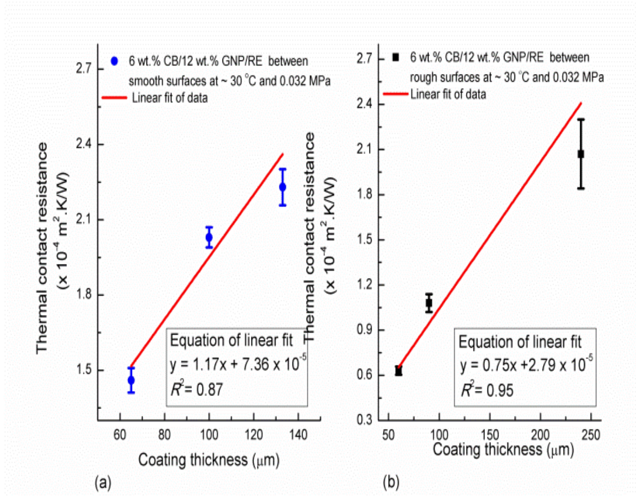

Figure. 4. Total thermal contact resistance versus coating thickness of 6 wt.% CB/12 wt.% GNP-5/rubbery epoxy hybrid composite (produced by mechanical mixing) measured between (a) smooth copper surfaces & (b) rough surfaces at ~30 °C and under a pressure of 0.032 MPa.

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