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GRAPHITE NANOPLATELET-BASED EPOXY COMPOSITES AS ADHESIVES AND PADS FOR THERMAL INTERFACE APPLICATIONS

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INTRODUCTION

The ever-decreasing size of micro electronic devices has resulted in higher power densities in much smaller spaces, leading to high heat evolution from the devices, which if not effectively removed would result in reductions in performance (e.g. switching delays) and reliability (e.g. thermally stressed, and then cracked, solder joints). Thermal management strategies are thus essential for electronics cooling and key to this are thermal interface materials (TIMs) [1]. Commercially, TIMs are employed in the form of heat dissipation compounds which primarily consists of polymer matrix/carrier in which thermally conducting fillers such as silver, aluminium nitride, boron nitride or SiC are dispersed at loadings of 50-70 wt.%. Such polymer matrix compounds can be used as the thermal grease/paste or adhesives [2]. These TIMs reduce interfacial thermal contact resistance between mating surfaces and thus facilitate heat dissipation. Both thermal grease/paste or adhesive are mainly employed for thin gap filling applications. However, when gap between mating surfaces is significantly large TIMs in the form of thermal pads are used. Thermal pads are similar to thermal adhesives but the polymer matrix is very compliant (soft) in nature such as silicone.

The thermal transport ability of TIMs strongly depends on the thermal conductivity of filler [3]. Carbon nanomaterials such as graphene, graphite nanoplatelets and carbon fibres have significantly higher thermal conductivity (>1000 W/m.K) [4, 5] than other inorganic fillers (BN, AlN or SiC) and therefore these have been extensively researched for development of next-generation polymer-based TIMs [6, 7]. A graphite nanoplatelet (GNP) is a 2-dimensional nanomaterial which can have thickness of 10-100 nm [8] and lateral width of several microns. Since it comprises of multilayer graphene sheets, it could offer comparable thermal conductivities in TIMs to that of graphene. However, unlike graphene, GNPs are cheap fillers which can be produced easily in high quantities.

GNP-based epoxy and silicone composites have been reported for thermal interface applications on the basis of their thermal conductivities [7, 9]. The thermal contact resistance between a semiconductor die and a heat sink or spreader is a major hindrance in the dissipation of heat from the die to heat sink [3] but this has not been evaluated much for GNP-based TIMs. The present study reports GNP/rubbery epoxy composites produced by three-roll milling and quantifies their heat dissipating ability as thermal interface adhesives and thermal pads by measurement of thermal contact resistance according to ASTM D5470, which to some extent replicates conditions in which electronic devices operate [10].

METHODOLOGY

GNPs (ex. XG Sciences) of sizes 5 µm (GNP-5) and 15µm (GNP-15) were dispersed at a loading of 2-35 wt.% in a rubbery epoxy (RE) resin using three roll milling (model 80E from EXAKT GmbH). The same protocol was followed for the production of composites as was reported in [9]. The dispersion of GNPs in rubbery epoxy resin was examined by scanning electron microscopy (SEM). The thermal conductivity and compression properties of composites were measured using a hot disk thermal analyser (Hot Disk AB), and a universal testing machine (Instron), respectively. For thermal contact resistance measurement, composite dispersions were sandwiched (in uncured form) between copper cylinders and tested in a thermal contact resistance measurement rig in uncured form or cured in-situ according to the procedure described in [10]. The composites were also fabricated as thermal pads/sheets by curing the dispersions (120 °C for 3 h) in a custom-made mould under a pressure of 0.3 MPa.

RESULTS AND DISCUSSION

The thermal conductivity, compression properties and thermal contact resistance of the composites produced in this study are presented in Table 1.

Table 1. Thermal conductivity, Compression properties and thermal contact resistance of pure rubbery epoxy and GNP/rubbery epoxy composites produced by roll mill. Thermal contact resistance of commercial TIM adhesive and Paste is also presented for comparison.

Material	Thermal Conductivity W/m.K	Compressive modulus (at 20 % strain) MPa	Compressive strength at failure MPa	Compressive strain at failure (%)	Bond line thickness µm	Thermal Contact resistance
Pure rubbery epoxy (RE)	0.176 ± 0.001	7.8 ± 0.52	2.48 ± 0.68	27.5 ± 4.94	15	9.4×10^{-5}
8 wt.% GNP-15/RE	1.13 ± 0.024	12.84 ± 0.56	5.18 ± 0.2	37.2 ± 0.14	-	-
15 wt.% GNP-15/RE	1.75 ± 0.004	15.23 ± 0.09	6.29 ± 0.64	38.83 ± 1.13	55	4.7×10^{-5}
20 wt.% GNP-15/RE	3.29 ± 0.038	21.23 ± 1.23	6.61 ± 0.8	33.64 ± 5.23	54	5.4×10^{-5}
25 wt.% GNP-15/RE	3.17 ± 0.11	20.16 ± 1.81	5.3 ± 0.88	29.06 ± 1.61	-	-
25 wt.% GNP-5/RE	1.47 ± 0.001	23.42 ± 1.57	8.93 ± 0.89	34.01 ± 1.53	60	6.8×10^{-5}
35 wt.% GNP-5/RE	2.36 ± 0.003	32.3 ± 5.54	9.01 ± 3.1	27.45 ± 3.2		
EPM 2490 (Commercial silicone based TIM, a product of Nusil Ltd.)	1.4	18.72 ± 3.92	7.5 ± 1.4	51.61 ± 2.86	95	1.01×10^{-4}
Matrix II paste (commercial TIM) www.tim-consultants.com	-	-	-	-	10-20	4.6×10^{-6}

Table 2. The thermal contact resistances of Thermal Pads having thickness of 0.6-0.7 mm measured at temperature of ~40 °C and compressive stress of 0.16 MPa

Composite coating	Total thermal contact resistance $m^2.K/W$
25 wt.% GNP-15/RE produced by Roll mill	4.2×10^{-4}
EPM 2490	3.9×10^{-4}
Dow Corning Pad	3.5×10^{-4}

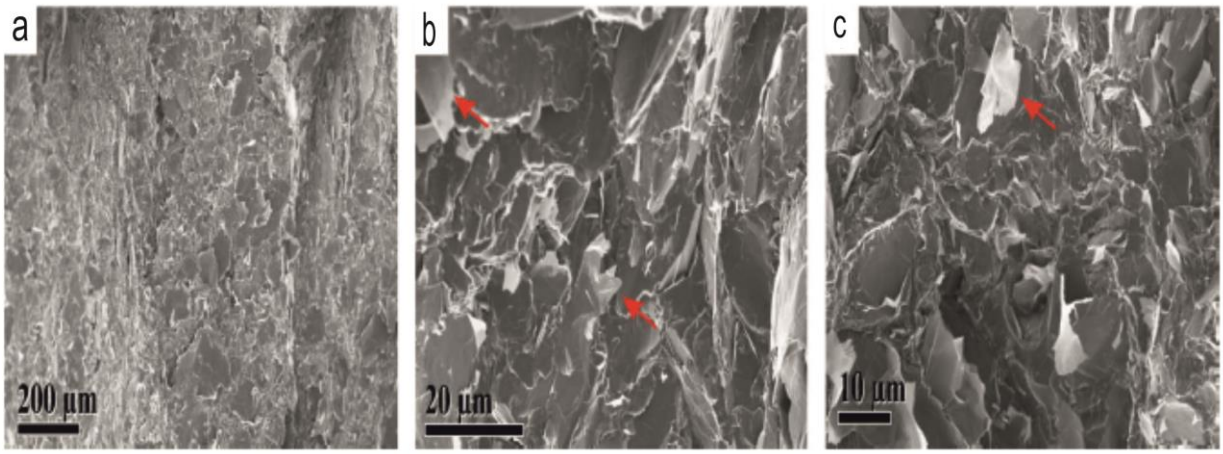

Fig. 1. SEM images of GNP-15/RE composite produced by roll mill at loading of (a &b) 25 wt.% and (c) 25 wt.% GNP-5/RE composite, arrows point towards GNPs in the matrix.

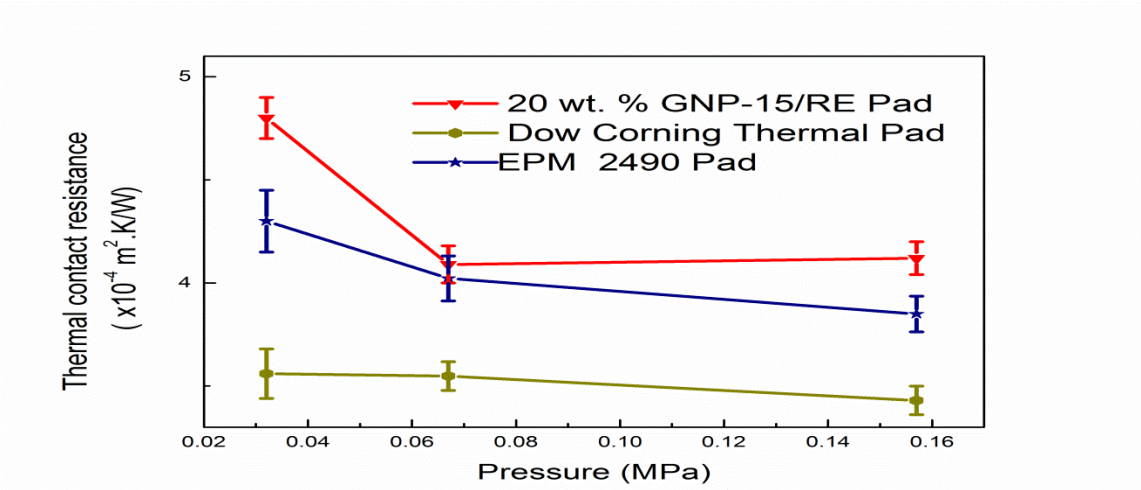

Fig. 2. Thermal contact resistance vs. applied pressure of GNP/rubbery epoxy and commercial pads measured at ~40 °C between smooth copper cylinders.

SEM revealed uniform dispersion of GNPs, regardless of loading, with less interplatelet alignment at low loadings (Fig. 1). The thermal conductivity of GNP/RE composites increased with GNP loading and size, as both factors favour improved thermal pathways. The thermal conductivities of 20wt. % GNP-15/RE (3.29 W/m.K) and 35 wt.% GNP-5/RE (2.36 W/m.K) were both significantly higher

than pure RE (0.17 W/m.K). GNP/RE retained good compliance, compressive moduli at 20 and 25 wt.% loading of GNP-15 being comparable to commercial BN/silicone TIM (Table 1). Although thermal contact resistance of GNP/RE was higher than for commercial paste (Table 1), its interfacial thermal transport outperformed GNP/silicone (due to RE's strongly adhesive nature) and, across thick bond lines, it outperformed previously reported GNP-pastes [11] and commercial silicone based TIM adhesive (Table 1). The thermal contact resistance of thermal pads decreases with increases of pressure (Fig. 2) as pressure improves their conformability with the mating surfaces. The 20 wt.% GNP-15/RE thermal pad had slightly higher thermal contact resistance than other thermal pads produced from commercial TIMs (Table 2). These results suggest that GNP/RE composites are promising candidates for thermal interface adhesives and pads.

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