



Thermal conductivity enhancement of epoxy adhesive using graphene sheets as additives



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ABSTRACT

Graphene-epoxy thermal conductive adhesive was obtained using few-layer graphene sheets as additives which were fabricated by a re-expansion and exfoliation method. The experimental results show that graphene sheets can effectively enhance the thermal conductivity of epoxy matrix, with $4.01 \text{ W m}^{-1} \text{ K}^{-1}$ for the maximum filling loading of weight 10.10%, which is enhanced by more than 22 times of the pure epoxy resin, about 2.2 times higher than that of the same resin with the maximum weight 16.81% filled with graphite nanoflakes, and approximately 2.4 times higher than that of the same resin with the maximum weight 44.3% filled with the natural graphite powder.

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1. Introduction

With the rapid development of electronic technology, the electronic components are gradually transformed from isolated to highly integrated [1] and modularized [2,3], which cause high heat flux for the electronic devices, and a great amount of heat is produced during the running [4,5]. According to the studies of Bar-Cohen et al. [6], the stability of the electronic devices will be depressed by 10% as their temperature rises by every 2°C . So, the heat cumulation of electronic components will directly depress the stability or shorten the electronic products life time, meanwhile lead to some serious consequences [7]. Therefore spreading the heat of the electronic devices effectively and efficiently has become an important issue on modern electronic encapsulations [8–11].

Cementation is a general technic in the high-integrated electronic encapsulation with thermal conductive adhesives [12–14]. Traditional thermal conductive adhesives are fabricated by filling the resin with high thermal conductivities fillers, such as carbon black [15], Aluminum oxide (Al_2O_3) powder [16], Aluminum nitride (AlN) powder [17], Zinc oxide (ZnO) powder [18], Boron nitride (BN) powder [19], diamond powder [20], copper powder (Cu) [21], Nickel (Ni) powder [22], Aluminum (Al) powder [23], Silver (Ag) powder [24] and so on. However, to improve the thermal conductivity of the adhesives ($1\text{--}5 \text{ W m}^{-1} \text{ K}^{-1}$), the above-mentioned traditional fillers must be large enough weight (wt) load

(fraction $\geq 50\%$), which would greatly affect their physical properties [25–27]. In recent years, carbon based materials, for example, carbon nanotubes (CNTs) [26], graphite nanoflake [28], and graphene oxide sheets [29] have been gradually used in thermal systems due to their low density. Whereas there are some factors to limit such carbon materials using in thermal adhesives for the material cost and inherent thermal conductivity and so on. Graphene as a single layer structure of two-dimensional new carbon material brings very unusual surprise [30], and has attracted tremendous interests particularly of the engineers and scientists due to its unique physical and chemical properties like strong mechanical robustness (Young's modulus of monolayer graphene is $1.0 \pm 0.1 \text{ Tpa}$) [31], excellent thermal conductivities (more than $5000 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature) [32] and large specific surface area ($\sim 3100 \text{ m}^2 \text{ g}^{-1}$ for activation of graphene) [33] and so on. As the highest thermal conductivity ever known up to now [32], meanwhile the layered structure is preferred to form the effective pathways for heat transfer, graphene sheet is considered to be a kind of good thermal conductive additive for thermal engineering applications [32,34]. The use of graphene sheet as a thermal filler to increase thermal conductivity of systems in thermal applications has been reported [36–40]. Some researchers pursued graphene sheet to improve the thermal conductivity of nanofluids. Tessa Theres Baby et al. [35] added 0.05% volume fraction (vol) of hydrogen exfoliated graphene (HEG) in deionized water (DI) and ethylene glycol (EG) based nanofluids, the enhancement in thermal conductivity of about 16% at 25°C and 75% at 50°C . Soujit Sen Gupta et al. [36] prepared water nanofluids at 0.2% concentration graphene nanosheets, the thermal conductivity enhancement

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obtained was 27%. Also some researchers exploited graphene sheet as a candidate to improve the thermal conductivity of polymer material. Raquel Verdejo et al. [37] filled 0.25 wt% functionalized graphene sheet in silicone foam, which enhanced the thermal conductivity of nanocomposite up to 8% than the original silicone foam. Khan M. F. Shahil et al. [38] mixed ~10 vol% graphene-multilayer graphene with epoxy revealed a record-high enhancement of thermal conductivity by 2300% than pristine epoxy. Xingyi Huang et al. [39] chose 50 vol% CNTs or 50 vol% graphene nanoplatelets to increase the epoxy resin thermal conductivity up to $7.30 \text{ W m}^{-1} \text{ K}^{-1}$ by synergetic effects.

In the present work, we report a re-expansion and exfoliation method to obtain mass few-layer graphene sheets using the commercially available graphite intercalation compounds (GIC) as starting material. Then the graphene-epoxy resin thermal conductive adhesive by filling the graphene sheets was fabricated by above mentioned method, subsequently we investigate the thermal conductivity of this kind of thermal adhesive with different weight filling loads. In addition, we chose natural graphite powder and graphite nanoflake as reference fillers with the same epoxy resin, and the thermal conductivities of all samples were characterized on Hot Disk TPS-2500 thermal constants analyzer.

2. Material and methods

2.1. Materials

All messages of materials used in the experiment are listed in Table 1.

2.2. Characterization methods

The Scanning electron microscopy (SEM) morphologies of all samples were characterized using FEI Quanta 400F electron microscope with thermal field emission 20 KV. Transmission electron microscopy (TEM) images were obtained using a FEI Tecnai G2 Spirit electron microscope with an accelerating voltage of 120 KV. Raman spectroscopy was performed by a Renishaw in Via Raman

microscope with 514 nm laser excitation at room temperature and with transferring the sample on a glass slide. X-ray photoelectron spectroscopy (XPS) was measured by ESCALab250. And the thermal conductivities of all samples were measured by Hot Disk TPS-2500 thermal constants analyzer at room temperature.

2.3. Fabrication of the graphene sheet

The detailed process of the graphene sheet fabrication was described in our previous work [40]. In briefly, GIC (a conventional expanded graphite based on H_2SO_4) was took into the muffle furnace, and kept the temperature about 900°C for 60 s, worm-like graphite was obtained, then the worm-like graphite was dispersed in ethanol for 5 h (h) by ultrasonic exfoliation, and we can get the graphite nanoflake after the filtration (the filter paper with $45 \mu\text{m}$ pore size) and vacuum drying process. Then, the graphite nanoflake was dispersed into the oleum for 5 h to weaken the graphite interlayer force, next we diluted the oleum by ice water and filtrated the solution, subsequently we shifted the treated graphite nanoflake in the H_2O_2 aqueous solution about 2 h with 5–10 wt%, again by filtration and vacuum drying treatment, then the graphite nanoflake were transferred in ethylene glycol solution for layers insertion, after filtration, then the intercalation graphite nanoflake were brought into the muffle furnace at 600°C about 120 min (mins) for the second thermal expansion. After the thermal expansion, the graphite nanoflake were put in a furnace with gas ($\text{H}_2:\text{N}_2 = 5:95$) for reduction about 2 h at 920°C . After the thermal reduction procedure, the reduced graphite nanoflake were shifted into the *N*-Methyl pyrrolidone (NMP) solution by ultrasonic exfoliation about 12 h with mechanical stirring about 200 rounds per minute, finally, the graphene sheet powder can be realized after the filtration and vacuum drying process. All the filter papers were used in this part with $40 \mu\text{m}$ pores.

2.4. Fabrication of thermal conductive adhesives

The epoxy resin and curing agent (1:1) were slowly mixed about 10 min in a beaker by mechanical mixer, then 5 wt% reactive diluent was added, after 5 min mixed, three fillers (natural graphite, reduced graphite nanoflakes, and graphene sheets) and some other additives (~1 wt% KH-550 and ~4 wt% hydroxy silicone oil) were added into the specifying beaker separately, after 30 min fast stirring by mechanical stirrer, the homogeneous mixture of thermal adhesives can be obtained, then put the prepared adhesives into the vacuum container application for de-aeration. Therewith the de-aeration adhesives were molded in Polytetrafluoroethene (PTFE) molds with the size of $\Phi \times H = 35 \text{ mm} \times 10 \text{ mm}$, the molds were transferred into the oven with constant temperature of 80°C and kept for 30 min, then the oven temperature was increased to 120°C and kept for 60 min, the cured adhesives can be obtained after the oven was turned off and its temperature was naturally cooled down at ambient environment.

3. Results and discussion

3.1. The morphologies of fillers

Fig. 1A shows the optical image of GIC of 100 meshes with metallic luster. Fig. 1B is the SEM image of the GIC, from which it shows that the GIC has layered structure, with the size of about $200 \mu\text{m}$. Fig. 1C is the SEM image of the worm-like graphite was obtained by expanding the GIC in the furnace of 920°C . It is obvious that the volume of the worm-like graphite is hundreds of times larger than that of the GIC, which is attributed to some substances (General chemical name: XCY) in the GIC interlayer decomposed

Table 1
Materials in the experiment.

Name of the material	Grade	Technical parameter	Manufacturer
Graphite intercalation compounds (GIC)	Analytical reagent (AR)	C > 98.0%	Qindao Huateng Graphite Product Co. Ltd
Graphite powders	AR	C > 98.0%	Qindao Yanxin Graphite Product Co., Ltd.
Graphite nanoflake	–	–	Self-prepared
Graphene sheet	–	–	Self-prepared
Bisphenol-A liquid epoxy resin (E-51)	–	Epoxide number 0.41–0.47 mol/100 g	Foshan Nanhai Baosheng Chemical Co., Ltd.
Cure agent: 650 polyamide	–	–	Foshan Nanhai Baosheng Chemical Co., Ltd.
Hydroxy silicone oil ($\text{HO}[(\text{CH}_3)_2\text{SiO}]_n\text{H}$)	AR	–	Guangdong BioMax Si&F New Material Co., Ltd;
Silane coupling agent: (3-aminopropyl) triethoxy-silan (KH550)	AR	98%	Aladdin Chemistry Co., Ltd.
Reactive diluent: propylene oxide-butyl ether	Industry grade	–	Yancheng Hehai Chemical Diluent Co., Ltd.
Deionized water	–	–	Self-prepared

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