Applied Thermal Engineering 66 (2014) 493–498

Contents lists available at ScienceDirect

Applied Thermal Engineering

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

Thermal conductivity enhancement with different fillers for epoxy resin adhesives

Yuan-Xiang Fu, Zhuo-Xian He, Dong-Chuan Mo, Shu-Shen Lu*

School of Chemistry and Chemical Engineering, Sun Yat-sen University, Guangzhou 510275, China

HIGHLIGHTS

- Thermal conductive adhesives with 8 different fillers were tested.
- The layer-shape filler is beneficial to form the heat pathways.
- The sharp-corner-shape filler is most difficult to achieve the heat pathways.
- The adhesive filled with the natural graphite has higher thermal conductivity.

ARTICLE INFO

Article history:
Received 9 September 2013
Accepted 18 February 2014
Available online 28 February 2014

Keywords:
Filler
Thermal adhesive
Thermal conductivity
Powder shape
Filling load

ABSTRACT

Heat dissipation is an important issue for electronic devices. In the present work, we prepared eight kinds of thermal adhesives by filling the epoxy resin with natural graphite, copper, aluminum, zinc oxide, boron nitride, aluminum oxide, diamond and silver powders, and measured the thermal conductivity of all samples. The results show the eight fillers can efficiently improve the thermal conductivity of the epoxy resin. Meanwhile, we found the layer-shape filler is more favorable than the ball-shape filler and the sharp-corner-shape filler to enhance the thermal conductivity of epoxy resin, and the low price layer-shape natural graphite-epoxy adhesive had the highest thermal conductivity up to 1.68 W m^{-1} K^{-1} at weight 44.3% of the eight thermal adhesives. All the fillers and the cross sections of thermal adhesives morphologies images were characterized by scanning electron microscopy, and the thermal conductivities of all the samples were measured by Hot Disk TPS-2500 thermal constants analyzer.

1. Introduction

With the quick development of the micro-electronic technology, the electronic components are gradually transformed from isolated to highly integrated [1] and modularized [2,3], which cause high heat flux from the electronic devices, and a great amount of heat is produced [4,5] during the running of the electronic devices. According to the studies of Bar-Cohen et al. [6], the stability of the electronic devices will be depressed by 10% as their temperature rise by every 2 °C. Heat cumulation will directly affect the stability and lifetime of the electronic devices, or brings some others serious consequences [7]. At present, how to realize the effective and efficient heat dissipation from the electronic devices has become the focus of the electronic capsulation [8–11], and the study on the thermal conductive adhesive, an essential material of electronic capsulation, has become more and more important. With the rapid development of the material science and technology, many kinds of new materials like carbon nanotubes (CNTs) [12,13], carbon fiber [14], and graphene sheet [15,16] are utilized to improve the thermal conductivity of the thermal adhesives. However, there are also some factors such as the mass production and the high price to limit such new materials widely used in the electronic capsulation at the present time.

The conventional thermal conductive adhesives are usually prepared by filling the resins with one or several kinds of common fillers, such as graphite powders, carbon black [17], Aluminum oxide (Al_2O_3) [18], Aluminum nitride (AlN) [19], Zinc oxide (ZnO) [20], Boron nitride (BN) [21], and diamond powders [22], or metal powders such as Copper (Cu) powders [23], Nickel (Ni) powders [24], Aluminum (Al) powders [25], and Silver (Ag) powders [26]. Though a great amount (the proportion of the content ≥50%) is needed to improve the thermal conductivity of the adhesives (1–5 W m^{-1} K^{-1}) for the traditional fillers, the technique of the preparation for these fillers is easy and low-cost, therefore these

* Corresponding author.
E-mail address: lvshsh@mail.sysu.edu.cn (S.-S. Lu).

http://dx.doi.org/10.1016/j.applthermaleng.2014.02.044
1359-4311/

low price traditional fillers are also widely used in the thermal conductive adhesives which will be the main products used in the electronic capsulation currently [27].

Herein we prepared eight kinds thermal conductive adhesives by filling the epoxy resin (E-51) with conventional fillers such as natural graphite, Cu, Al, ZnO, BN, Al_2O_3, diamond, and Ag powders, respectively. We measured the thermal conductivities of all samples by Hot Disk TPS-2500 thermal constants analyzer, and demonstrated the variations and the enhancement of the thermal conductivities of these thermal conductive adhesives as the filling load of the fillers are increased. We comparatively study the effects of fillers morphologies on the thermal conductivities of the thermal conductive adhesives. Meanwhile, considering the prices of the fillers, we think the lamellar graphite is a kind of excellent thermal conductive adhesive filler.

2. Experiment

2.1. Materials

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

2.2. Characterization method

The scanning electron microscope (SEM) morphologies of all samples were characterized using FEI Quanta 400F with thermal field emission 20 kV; Thermal conductivities of all the samples were measured by the Hot Disk TPS-2500 thermal constants analyzer at room temperature.

Table 1
Materials in the experiment.

Name of the material	Grade	Technical parameter	Manufacturer
Graphite powders	Analytical reagent (AR)	$C > 98.0\%$	Qindao Yanxin Graphite Product Co., Ltd.
Cu powders	AR	_	Tianjing Kemiou Chemical reagent Co., Ltd.
Al powders	AR	_	Tianjing Kemiou Chemical reagent Co., Ltd.
Ag powders	AR	_	Sinopharm Chemical reagent Co., Ltd.
ZnO powders	Industry grade	_	Wuxi zehui Chemical co., Ltd.
BN powders	AR	_	Liaoning Yingkou Liaobin Meticulous Chemical Co., Ltd.
Al_2O_3 powders	AR	_	Tianjin Kemiou Chemical Reagent Co., Ltd.
Diamond powders	_	_	Tianjin Chanyu Superhard Sci-Tech Co., Ltd.
Epoxy resin (E-51)	_	Epoxide number 0.41–0.47 mol/100 g	Foshan Nanhai Baosheng Chemical Co., Ltd.
Cure agent: 650 polyamide	_	Amide value 200 ± 20 mg KOH/g	Foshan Nanhai Baosheng Chemical 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

2.3. Surface treatment of fillers

To facilitate the fillers better dispersion in the epoxy resin matrix, surface modification is essential and the process is as following: ethanol solution with deionized water 10% was prepared firstly, and the potential of hydrogen (pH) of solution about 3.5–5.5 was regulated by acetic acid, then a certain amount of fillers powders were weighed up and put in the numbered beakers separately, then weight (wt) 5% of the filler KH-550 solution was added into the beakers separately, after that, put the beakers into the ultrasonic cleaner for 30 min by mild ultrasonic dispersion, then hydrolyzed for 2 h at 50 °C in the thermostat, filtered and washed them until neutral with deionized water and ethanol several times, transferred the treated fillers into the vacuum oven at 60 °C about 12 h. Owing to ZnO can hydrolyze in the acidic condition, and Cu powders will be oxidized at the hydrolysis process, we canceled the two type fillers surface treatment operation.

2.4. Preparation of the adhesives

E-51 and curing agent (1:1) mixed for 10 min (mins) by mechanical stirrer firstly, then wt 5% reactive diluent was added, after 5 min mixed, eight treated fillers were added the specifying beaker separately, homogeneous mixture of the thermal adhesives can be obtained after 30 min stirring. Put the prepared adhesives into the vacuum chamber for de-aeration, and moulded the adhesives in the polytetrafluoroethylene (PTFE) moulds (35 mm in diameter and 10 mm in thickness), and transferred the moulds into oven with the constant temperature of 80 °C for 30 min, then increased the temperature to 120 °C and kept the temperature for 60 min, the adhesives can be cured with the temperature naturally cooling down with the oven finally.

3. Results and discussion

3.1. Morphology of the filler

The morphologies of the eight fillers samples were characterized by SEM after spray-gold treatment in Fig. 1. Fig. 1a shows that the diameter of ZnO powders are about 0.5–1 μm and no aggregations is found in the figure, and the sample is with particles and cylindrical shape mostly. Fig. 1b is the fluffy structure powders of BN, wherein the particle diameter is observed to be 400 nm and with some aggregations in the figure, which may be caused by the imperfect grind after the surface treatment. The Al_2O_3 powders are nearly ball-like with the diameter in the range of 20–40 μm with some crackles on the surface is observed in Fig. 1c. Fig. 1d is the SEM image of the natural graphite powder, which has good layer structure of about 70–100 μm, and some broken graphite flake with smaller size. Fig. 1e shows the Al powders with ball shape and well dispersed, and most of the diameters are about 20 μm. Fig. 1f is the image of the Cu powders with laminate-shape structure and some with branch shape, which may be favorable to construct the heat pathways in the resin. From the image (Fig. 1g), it can be seen that the diamond powders with sharp-corner shape, and the size is about 40 μm. The Ag powders after surface treatment is well dispersed and aggregations can also be observed in Fig. 1h, which is probably due to the smaller size of Ag powders particle with about 1 μm.