Materials Chemistry and Physics 162 (2015) 182–187

Contents lists available at ScienceDirect

Materials Chemistry and Physics

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

Heat conduction models of interfacial effects for $TiB_2-Al_2O_3$/epoxy composites

Yicheng Wu, Zhiqiang Yu*, Yannan He, Xiaolei Yang

Department of Materials Science, Fudan University, 200433 Shanghai, China

HIGHLIGHTS

- Epoxy composites reinforced by surface modified $TiB_2-Al_2O_3$ composite fillers were prepared.
- Thermal conductivity of composites increased as the filler volume fraction increased.
- Finite element models in 3-D are in reasonable agreement with experimental data.
- The vector diagram of thermal flux illustrates the heat is mainly conducted by the effective interfacial layers.

ARTICLE INFO

Article history:
Received 6 January 2015
Received in revised form
19 May 2015
Accepted 24 May 2015
Available online 27 May 2015

Keywords:
Composite materials
Thermal conductivity
Computer modeling and simulation
Interfaces

ABSTRACT

The heat conduction of $TiB_2-Al_2O_3$/epoxy composites was discussed by comparisons of the finite element model (FEM), the self-consistent model, and experimental results. The experimental thermal conductive performance of epoxy composites was characterized by laser flash method. According to the calculated and experimental results, TiB_2 showed optimistic potential in improving thermal conductive performance of epoxy composites. With the addition of Al_2O_3 particles, the thermal conductive performance of epoxy composites filled by $TiB_2-Al_2O_3$ composite fillers was improved remarkably at high volume fraction. After surface modification of ceramic fillers by silane coupling agents, the efficiency of the heat diffusion of epoxy composites was increased further. In comparison of the finite element analysis (FEA), the self-consistent model and experimental results, the thermal conductive values of FEA were in reasonable agreement with experimental values, and the calculated values from self-consistent models were lower than the experimental values slightly.

1. Introduction

Due to the outstanding mechanical performances and chemical resistance, epoxy resin has been widely applied in the fields of engineering materials and encapsulation of electronic devices [1]. It is inevitable that the heat is generated as the friction occurs in the engineering materials and electronic devices work. This heat causes the temperature rise and shortens the service life of epoxy resin. In order to increase the heat dissipating rate, the thermal conductive performance of epoxy resin has been discussed.

Many combinations of epoxy composites filled by high thermal conductive particles have been investigated. W. Peng et al. [2] incorporated aluminum nitride (AlN) nanoparticles into epoxy resin and indicated that the thermal conductivity of epoxy nanocomposites increased with increasing AlN content. K. Kim et al. [3] showed that the addition of the surface-modified boron nitride (BN) particles into the epoxy matrix had a remarkable effect on the thermal conductivity. The thermal conductivity of the composites containing 70 wt% surface-treated BN particles was 20.5 times higher than that of the pure epoxy matrix. J. P. Hong et al. [4] introduced high thermal conductive AlN and BN into the epoxy matrix in order to identify the effects of the particle size and the relative composition on the thermal conductivity of composites and drew the conclusion that the effect of the relative size of filler on the thermal conductive path can be defined by particle size ratio (RD) of the AlN and BN, and also schematically expressed the bimodal distribution curves. According to Gu et al.'s work, epoxy composites can be obtained high dielectric performance by filling with TiO_2 coated TiB_2 fillers [5]. It shows the potential of the introduction of TiB_2 particles in epoxy resin for the performance

* Corresponding author. Department of Materials Science, Fudan University, No. 220, Road Handan, 200433 Shanghai, China.
E-mail address: yuzhiqiang@fudan.edu.cn (Z. Yu).

http://dx.doi.org/10.1016/j.matchemphys.2015.05.056
0254-0584/

improvement. In addition, TiB_2 shows remarkable thermal properties in ceramic matrix composites (CMCs) [6–8]. It could be helpful to improve thermal conductivity of epoxy composites by being combined with TiB_2 fillers.

The compatibility of inorganic fillers in organic matrix affects the performances of composites significantly. In order to improve the compatibility of TiB_2 fillers in epoxy matrix, the Al_2O_3 particles which can be modified with silane coupling agents are combined with TiB_2 fillers. The high energy ball mill can mix TiB_2 and Al_2O_3 particles and form ceramic composite fillers [9]. The composite fillers can be modified with silane coupling agents due to the introduction of Al_2O_3 particles. The chemical bonds can be formed between epoxy matrix and Al_2O_3 particles [10] and the semi-coherent interfaces can be formed between Al_2O_3 and TiB_2 particles. Hence, the compatibility of TiB_2 particles in epoxy matrix can be improved. According to Fredrik Larsson and I. Özdemir et al.'s works, a framework for variationally consistent homogenization, combined with a generalized macrohomogeneity condition, of non-linear transient heat conduction and the heat conduction of heterogenous solids have been fully discussed [11,12]. In their works, the numerical models were applied to investigate the heat conduction and estimated the effective thermal conductivities of multiphase composites accurately. The heat conduction of composites simultaneously associated with interfacial issues and heterogenous solids is non-linear. Finite element models (FEMs) are more suitable to non-linear systems. R. Nayak et al. [13] investigated the effective thermal conductivity of micro-sized pine wood particles filled epoxy composites by ANSYS and experimental method and showed that the values obtained using finite element analysis were found to be in reasonable agreement with the experimental values. Alok Agrawal and Alok Satapathy [14] reinforced epoxy resin with 0–15 vol% AlN and analyzed thermal conductivity by experiments and Maxwell et al.'s numerical models. Maxime Villière et al. [15] studied also thermal conductivity tensor of twill-weave carbon fabric impregnated with epoxy resin by experimental determination and numerical model. In this paper, the thermal conductivities of epoxy composites filled by TiB_2–Al_2O_3 composite ceramic with the addition of silane coupling agents are discussed by comparison of the finite element model, the self-consistent model, and experimental results. The experimental samples of epoxy composites were prepared by the in situ synthesis, and the experimental thermal conductive performance of epoxy composites was characterized by laser flash method. The interfacial effects of epoxy composites were investigated by FEM and self-consistent model.

2. Experimental

2.1. Materials

The epoxy resin was synthesized from BPA (bisphenol A, >99.0%, white crystal, Sinopharm Chemical Reagent Co. Ltd) and epichlorohydrin (>99.0%, colorless transparent oily liquid, Sinopharm Chemical Reagent). The TiB_2 (>99.5%, black powder, average particle size 1 μm) and Al_2O_3 (>99.9% metals basis, α-phase, white powder, average particle size 30 nm) particles were provided by Aladdin Chemistry Co. Ltd. The γ-glycidoxypropyltrimethoxysilane (with purity of 98%, supplied by Sinopharm Chemical Reagent Co., Ltd. China) was used for silane functionalization of Al_2O_3.

2.2. High-energy ball milling

In order to combine the two types of ceramic particles, the mixed TiB_2 and Al_2O_3 powders were sealed in the zirconia jars with acetone and zirconia beads at room temperature. The two ceramic particles (TiB_2 and Al_2O_3) were weighed in a mass ratio of 40:1. The zirconia jars were fixed in the high-energy planetary ball mill (PMQ, DROIDE, China) and it was run at 600 rpm for 2 h. After the milling, the mixtures were dried at 80 °C, prepared for filling the epoxy matrix.

2.3. Surface modification of TiB_2/Al_2O_3 composite fillers

The γ-glycidoxypropyltrimethoxysilane (5 g) was added in an ethanol-aqueous solution prepared by the volume ratio of ethanol: water = 1:9 (250 mL). The solution was stirred for 5 min. Then the TiB_2/Al_2O_3 composites dried at 100 °C for 1 h (50 g) were dispersed in above solution with a mechanical stirrer for 3 h, and ultrasonic process for 10 min. The temperature of composite fillers modification was controlled at 50 °C. After the reaction, the powders were washed by acetone and dried in a vacuum oven at 80 °C for 1 h.

2.4. Filling epoxy matrix with TiB_2–Al_2O_3 composites by in situ synthesis

Bisphenol A was solved in NaOH (solution, 0.3 mol/L) at 70 °C, and TiB_2–Al_2O_3 composite particles (preprocessed by ultrasound for 30 min) were introduced into the solution. The temperature of the solution was reduced to 50 °C after TiB_2–Al_2O_3 composite particles dispersing evenly, and added epichlorohydrin by droplets. The reaction product was poured into the molds after the temperature of solution being raised to 90 °C for 2 h. The molds were heated over 150 °C until no solvent left. Molds were cooled to 70 °C, triethylene tetramine was added by 1.5 mL in each molds and then molds were heated again to 120 °C for 5 h. The ceramic composite particles were added in different percentages: 1 vol%, 3 vol% and 5 vol% with regard to the epoxy resin.

2.5. Characterization

The thermal conductive coefficients of epoxy composites can be computed from the values of density (ρ), thermal diffusion coefficient (α) and specific heat capacity (C_p) [16]. The density of samples can be measured by Archimedes principle. In order to characterize the α and C_p of the epoxy composites, the samples were analyzed by laser flash method (LFA447, NETZSCH, Germany). The experimental measurement set-up and the principle of laser flash method are illustrated in our previous study [16].

The α of samples can be obtained from output signal curves in LFA447. The output signal intensity is affected by the voltage of laser emission system and the pulse width of laser. The excessive voltage and pulse width of laser will cause the signal overflowing. In this test, the voltage of laser emission system was set at 270 V. The pulse width of laser light was set as “medium”. The testing temperature were set at 25 °C. Each sample was tested 5 times (shots) and the average results were taken from these five times test results for reducing the measurement uncertainties.

The absorbed quantity of heat of samples is known in LFA447. Moreover, the C_p of samples can be obtained by comparing the equilibrium temperature which is recorded after the measurement of standard samples and testing samples due to the mass of testing samples and standard samples, and the C_p of standard samples are known. In order to match the instrument, samples were required to shape their size to a diameter in 12.7 mm, and thickness in 2 mm of disks. Pyroceram9606 was chosen as the standard sample for getting results of C_p. In laser flash method, the laser shines on the surface of samples. The laser will penetrate the translucent neat epoxy resin samples and be detected by the heat sensor directly. It results in large errors of the measurements of neat epoxy samples. To avoid the penetration of laser, the neat epoxy resin samples were