International Journal of Heat and Mass Transfer 74 (2014) 358–367

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

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

Effects of multi-walled carbon nanotubes addition on thermal properties of thermal grease

Gou Yujun [a,b], Liu Zhongliang [a,*], Zhang Guangmeng [a], Li Yanxia [a]

[a] College of Environmental and Energy Engineering, Beijing University of Technology, Beijing 100022, China
[b] Tangshan College, Tangshan 063000, Hebei Province, China

ARTICLE INFO

Article history:
Received 11 November 2013
Received in revised form 26 February 2014
Accepted 5 March 2014
Available online 12 April 2014

Keywords:
Thermal interface materials
Multi-walled carbon nanotubes
Thermal conductivity
Contact pressure

ABSTRACT

Improving the thermal conduction ability of thermal interface materials (TIMs) can enhance heat transfer in electronics packages and reduce chip temperature. In the present work, multi-walled carbon nanotubes (MWCNTs) are used as additives for enhancing the thermal conductivity of the widely-used TIM silicone grease. The effects of the MWCNTs length and surface treatment on the silicone grease conductivity are studied. The results show that the MWCNT length has strong influences on the thermal performance of the grease with MWCNT additives. The equivalent thermal conductivity of the composite layer of the grease and the solid surfaces increases as the MWCNT length decreases. Only the length of MWCNTs is short enough, their addition can improve the thermal performance of the common silicone grease. Surface treatment on MWCNTs can improve the thermal performance of the grease. Through transmission electron microscope (TEM) observation it is found that after the modification of the short MWCNTs the MWCNT entanglement phenomenon in the mother silicone grease is weakened and the MWCNTs are dispersed more uniformly. The influence of the contact pressure on the thermal performance of the MWCNT TIMs is also investigated. The results show that the thermal resistance of the composite layer formed with different TIMs all decreases with the applied contact pressure, but the TIMs with the addition of the short and chemically treated MWCNTs are less sensitive to the contact pressure.

1. Introduction

With the development of metal–organic chemical vapor deposition (MOCVD) extension growth technology, multi-quantum well (MQW) structure, GaN-based materials and packaging technology, light-emitting diode (LED) applications have been gradually shifted into the lighting area from display area. However, compared with the existing lighting fixtures, the high-power lighting LED (power greater than 1 W) is facing a main technical bottleneck of poor heat dissipation that limits its stability, reliability and lifetime. The whole heat transfer process through the LED system includes several steps. The heat generated by LED chips is first transferred to the substrate, and then from the substrate to thermal interface material (TIM), after that it is transferred to the heat sink and finally from heat sink to the outside environment. Many work have been carried out to develop heat sinks to enhance the thermal performance [1–6] of the whole cooling system. However, as the heat sink thermal performance improves, the contact resistance between heat sources and heat sinks is becoming more and more important. To reduce this contact resistance, TIM is applied between heat source and heat sink. Thermal grease is one of the most widely-used thermal interface materials. Good thermal grease should be non-corrosive to the contact solid surface, durable and of course, most importantly, of large thermal conductivity.

The traditional thermal grease is mainly composed of polymer matrix. In order to enhance the thermal conductivity, some particle fillers of large thermal conductivity such as metals like Cu, and Ag, metallic oxides like Al_2O_3, CuO, and ZnO and large-thermal-conductivity nonmetals like SiC nano-particles, may be added in the grease matrix [7–12]. The measurements showed that the chip temperature increases with the LED power sharply. For the high-power lighting LED, the chip temperature is so high that the efficiency and reliability is greatly reduced with these traditional thermal greases as TIM. Development of the high performance thermal grease as TIM brooks no delay for the widespread use of high powder LED.

Carbon nanotube (CNT) has been extensively studied by many researchers owing to their potential mechanical, electrical and thermal properties since its first appearance [13,14]. In recent

* Corresponding author. Tel./fax: +86 10 6739 1917.
E-mail address: liuzl@bjut.edu.cn (L. Zhongliang).

http://dx.doi.org/10.1016/j.ijheatmasstransfer.2014.03.009
0017-9310/

Nomenclature

q_1	heat flux of the copper rod, W/m^2	R_g	grease layer thermal resistance, m^2 K/W
q_2	heat flux through grease composite layer, W/m^2	R_{c1}	thermal resistances of the composite due to the thin copper layer near surface measured from the thermocouple location, m^2 K/W
λ_1	copper conductivity, W m^{-1} K^{-1}	R_{c2}	thermal resistances of the composite due to the thin copper layer near surface measured from the thermocouple location, m^2 K/W
λ_2	equivalent thermal conductivity of the composite layer, W m^{-1} K^{-1}	δ_2	coating thickness of the grease, mm
dT/dx	temperature gradient, K/m		
ΔT_2	temperature difference of the immediate vicinity of the coating of the two copper plates, K		
R_2	composite layer thermal resistance, m^2 K/W		

years, CNTs are of great interest as a heat-conductive filler for polymer composites. Compared with the above mentioned free element nanoparticles and compound nanoparticles, CNTs are excellent candidates as dispersions for preparing grease with enhanced thermal property due to their high thermal conductivity [15–20] and very large aspect ratio. Balandin [21] reviewed the thermal properties of carbon materials focusing on recent results for graphene [22], carbon nanotubes and nanostructured carbon materials with different degrees of disorder.

However, the CNTs are very easy to form aggregates that affect CNTs dispersing in the grease composites uniformly, so preparation of silicone grease with homogeneous CNT suspension remains a technical challenge. The functionalization of CNTs for the polymer composites has already been used for improving the dispersion of CNTs in the polymer matrix and enhancing the combination in the inter-faces between CNTs and polymer matrix.

Gulotty [23] proved that functionalized MWCNTs performed the best as filler material leading to a simultaneous improvement of the electrical and thermal properties of the composites, although the functionalization of the single-wall carbon nanotubes would reduce the thermal conductivity enhancement. The observed trends were explained by the fact that while surface functionalization increases the coupling between carbon nanotube and polymer matrix, it also leads to formation of defects, which impede the acoustic phonon transport in the single-wall carbon nanotubes. Balandin [24] have reviewed phonon properties in semiconductor nanostructures and two-dimensional graphene. It was shown that quasi-2D materials such as graphene and few-layer graphene offer many advantages for phonon engineering in comparison to conventional nanostructures.

Park et al. [25] reported functionalization of CNTs noticeably decreased the thermal conductivity and even more the electrical conductivity because of the damage to the CNT structures. Ma et al. [26] reviewed the functionalization methods of CNTs and pointed out that defects functionalization is another effective method for covalent functionalization of CNTs. This process takes advantage of chemical transformation of defect sites on CNTs. Defect sites can be created on the sidewalls as well as at the open ends of CNTs by an oxidative process with strong acids such as HNO_3, H_2SO_4 or a mixture of them. A method for the preparation of a hybrid material consisting of various size silica nanoparticles attached to multi-walled carbon nanotubes (MWCNTs) is proposed by Zhou et al. [27]. Yang et al. [28] added chemically functionalized MWCNTs into epoxy. Transmission electron microscopy showed that there was a TETA thin layer on the MWCNT surface, which contributes to the homogenous dispersion of MWCNTs in epoxy matrix and the improvement of the MWCNT–epoxy interfacial interaction. Thus the impact strength, bending strength and thermal conductivity of the composites are enhanced.

In the present work, MWCNTs are used as additives for enhancing the thermal conductivity of the widely-used TIM silicone grease. The effects of the MWCNTs length on grease conductivity are studied first. Three different-length MWCNTs of the same diameter are used to prepare the polymer composites, and their thermal performances were tested. During the samples preparation process, it is found that the MWCNTs are strongly entangled, forming aggregates owing to their strong van der Waals interactions, which will inevitably influences the thermal performance of the composites. To solve this problem, we combine the functionalization of the MWCNT surfaces with the addition of dispersants in the preparation process. The thermal performances of the composites with treated MWCNTs are compared with that of the common grease and that of the grease added plain MWCNTs. The influences of contact pressure on the contact thermal resistance are also investigated.

2. Experimental setup and materials

2.1. Experimental setup

The apparatus used in this study include a simulated LED heat source, two copper plates and a press device as shown in Fig. 1.

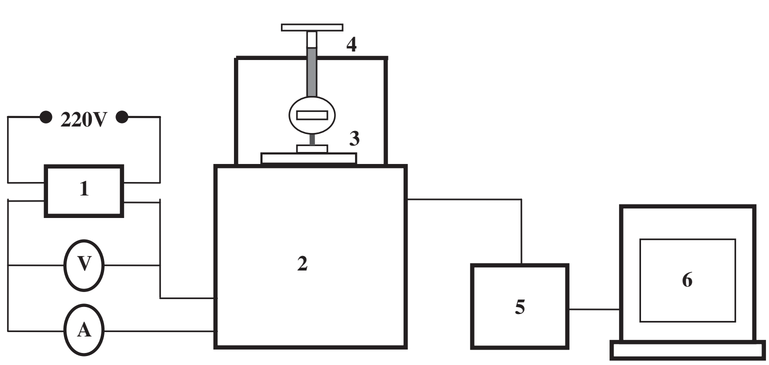

Fig. 1. Experimental system. 1, transformer; 2, heating unit; 3, copper plate; 4, press device; 5, data acquisition/switch unit; 6, PC.