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Carbon black pastes as coatings for improving thermal gap-filling materials

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Abstract

Carbon black pastes were found to be effective as coatings for improving the performance of thermal gap-filling materials, including flexible graphite, aluminum and copper. The thermal contact conductance across copper mating surfaces was increased by up to 180%. A fluidic form of carbon black paste (based on polyethylene glycol) was more effective than a thixotropic form (based on polyol esters). The carbon black pastes were much more effective as coatings than a commercial silver paste. With a carbon black paste coating, aluminum foil (7 μm thick) was a superior gap-filling material compared to similarly coated flexible graphite (130 μm thick). However, without a coating, flexible graphite was superior to aluminum. Commercial silicone-based gap-filling materials were inferior to flexible graphite or aluminum (whether coated or not).

Keywords: Carbon black; Exfoliated graphite; Graphite; Carbon composites; Thermal conductivity

1. Introduction

Heat dissipation is the most critical problem that limits the performance, power, reliability and further miniaturization of microelectronics. The alleviation of this problem is mostly attained by the use of heat sinks, which are materials of high thermal conductivity (such as copper) for allowing the heat to flow by conduction from the heat source (e.g., the microprocessor of a computer). However, the effectiveness of this approach is limited by the thermal resistance associated with the interface between the heat sink and the heat source [1–5]. In order to diminish the thermal resistance, a thermal interface material is placed between the heat sink and the heat source. This material can be fluids, pastes and solders (solders being applied in the molten state) [6–15].

The heat sink may be in direct contact with the heat source at the asperities of either of the mating surfaces, due to the roughness associated with each surface. There may even be a gap between the two surfaces, due to the geometry of the surfaces, the degree of alignment of the surfaces, and/or the configuration of the assembly. The presence of a gap is actually quite common. Due to the thermally insulating nature of air, the use of a thermal interface material that fills the gap is needed.

The design of a thermal interface material for filling gaps differs from that of one for filling the valleys in the surface topography of the mating surfaces that are in contact. These valleys contain air which should be displaced by a thermal interface material that is highly conformable and that is very thin (ideally just thick enough to fill the valleys, as the thermal resistance increases with thickness) [12,13]. Thus, a thermal interface material for filling the valleys is typically a paste of low viscosity. (The higher the viscosity, the less is the conformability [10].) However, for gap filling, the thermal interface material must be thick enough to fill the gap and be able to maintain its geometry. Thus, a gap-filling material typically involves a thixotropic paste (such as silicone filled with thermally conductive particles [6–9]), which is in contrast to the fluidic pastes (such as polyethylene glycol filled with thermally conductive particles

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[12,13]) that are effective for filling the valleys in surfaces that are in contact. Another type of gap-filling material is a solid sheet such as a metal foil. Ideally the sheet exhibits resiliency and hence conformability. Metals are ductile but have low values of the elastic limit. Therefore, metals are limited in resiliency or conformability. In contrast, "flexible graphite" [16–19] is a graphite sheet that is flexible and is resilient in the direction perpendicular to the sheet. The resiliency is made possible by the microstructure, which involves the mechanical interlocking of exfoliated graphite in the absence of a binder [20]. In general, a gap-filling material in the form of a solid sheet may be made more effective by coating both sides of the sheet with a thermal paste. Thixotropic pastes have been used for this purpose. Due to the relatively large thickness of a gap-filling material, high thermal conductivity is a more important attribute for a gap-filling material than for a material for filling valleys in surfaces that are in contact.

This paper is aimed at the improvement of thermal gap-filling materials by the use of carbon black paste coatings [12–14] in both fluidic and thixotropic forms. The fluidic form uses polyethylene glycol as the vehicle, whereas the thixotropic form uses polyol esters as the vehicle. Both pastes have been shown to be highly effective for filling the valleys in surfaces (especially surfaces that are quite smooth) that are in contact [12–14]. The high effectiveness of carbon black pastes stems from the high compressibility (hence conformability) of carbon black, which is in the form of porous agglomerates consisting of particles of size 30 nm [12,13].

As shown in this work, the use of carbon black paste (particularly the fluidic type) to coat a thermal gap-filling sheet results in a composite material that exhibits high conformability (due to the carbon black paste) as well as the ability to provide a substantial thickness and to maintain the geometry associated with the gap. In particular, this paper addresses the use of carbon black paste to coat flexible graphite, aluminum foil and copper foil for thermal gap filling. Flexible graphite is attractive for its resiliency in the direction perpendicular to the sheet, whereas metal foils are attractive for their high thermal conductivity.

The mating surfaces used in this study for measurement of the thermal contact conductance are copper, due to the common use of copper for heat sinks. Furthermore, this study includes comparison of copper surfaces that are of different degrees of roughness, because different degrees of roughness are encountered in practice and both conformability and thickness of the paste in valleys depend on the roughness.

The objectives of this work are (i) to evaluate the effectiveness of carbon black pastes as coatings for improving thermal gap-filling materials, (ii) to compare the effectiveness of carbon black paste coatings in fluidic and thixotropic forms, (iii) to compare the effectiveness of carbon black paste coatings on various supporting sheets, including flexible graphite, aluminum foils and copper foils of various thicknesses, (iv) to compare the effectiveness of carbon black paste coatings for mating surfaces of different degrees of roughness, and (v) to provide a comparative study that includes commercial gap-filling materials.

2. Experimental methods

The formulation, ingredients, preparation method and testing method of the carbon black thermal paste of this work are identical to those in the prior work of these authors [12–14]. The carbon black was Vulcan XC72R GP-3820 from Cabot Corp., Billerica, MA. It was a powder with particle size 30 nm, a nitrogen specific surface area 254 m^2/g, maximum ash content 0.2%, volatile content 1.07%, and density 1.7–1.9 g/cm^3. The carbon black powder was mixed with a vehicle by hand stirring to form a uniform paste. The particle size (30 nm) of the carbon black is much less than those of the metal or ceramic particles used in commercial thermal pastes.

Polyethylene glycol (PEG, or $HO(CH_2CH_2O)_nH$) was used as the organic vehicle for the fluidic type of thermal paste. It was PEG 400 from EM Science (Gibbstown, NJ). It had an average molecular weight of 400 amu; this average value corresponds to $n \sim 8.68$. It was a liquid at room temperature and contained ethyl cellulose (E8003, Sigma Chemical Co., St. Louis, MO) at 3 vol.%. The ethyl cellulose was a white powder that was dissolved in the vehicle. It served to improve the dispersion and suspension of the solids in the paste.

The vehicle used for the thixotropic type of thermal paste consists of polyol esters, which are attractive for their thixotropic behavior and ability to resist elevated temperatures. The polyol esters in the vehicle are pentaerythritol ester of linear and branched fatty acids and dipentaerythritol ester of linear and branched fatty acids. The polyol ester mixture is provided by Hatco Corp., Fords, NJ. The specific gravity is 0.97.

The fluidic thermal paste was prepared by first dissolving ethyl cellulose (3 vol.%) in the vehicle PEG. The dissolution was performed at about 60 °C (with heat provided by a hot plate). The heating was applied to hasten the dissolution of ethyl cellulose. After this, carbon black (1.25 vol.%) was added. Mixing was conducted by using a ball mill and stainless steel balls for 30 min. After mixing, the paste was placed in a vacuum chamber (which involved a mechanical vacuum pump) without heating for the purpose of air bubble removal.

The thixotropic paste contained 2.4 vol.% carbon black. It was prepared by manual stirring of a mixture of carbon black and vehicle.

Conventional thermal gap-filling materials in the form of thin sheets were cut to size 1×1 in. (25×25 mm) in the plane of the sheet. Then the thermal paste was applied manually on both 1×1 in. (25×25 mm) surfaces of a gap-filling sheet, such that the thickness of the paste on either side was 25 μm or less. The conventional thermal gap-filling materials used were flexible graphite, copper foil (thickness = either 13 or 130 μm) and aluminum foil