

Carbon-Black Thixotropic Thermal Pastes for Improving Thermal Contacts

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This paper addresses thermal interface materials for thermal conduction of excess heat for microelectronic applications. Carbon black (30 nm) thixotropic paste based on polyol ethers is comparable to carbon black fluidic paste based on polyethylene glycol (PEG) in its effectiveness as a thermal paste, and in its dependence on pressure history. Prior pressure (up to 0.69 MPa) application is helpful. The optimum carbon black content is 2.4 vol.% for the thixotropic paste. The thermal contact conductance across copper surfaces is 30×10^4 and 11×10^4 W/m²·°C for surface roughness of 0.05 μ m and 15 μ m, respectively. The volume electrical resistivity is 3×10^3 Ω -cm. Boron nitride (BN) (5–11 μ m) and graphite (5 μ m) thixotropic pastes are less effective than carbon black thixotropic paste by up to 70% and 25%, respectively, in thermal contact conductance, due to low conformability.

Key words: Thermal paste, carbon black, boron nitride (BN), graphite, thixotropic, polyol ester, polyethylene glycol (PEG), thermal contact

INTRODUCTION

Thermal contacts are commonly encountered in heat transfer and can be described as the interface between a heat source (e.g., the microprocessor of a computer) and a heat sink. An important scenario relates to the cooling of microelectronics, as overheating is the most critical problem in the electronics industry. This problem limits the power, performance, reliability, and further miniaturization of microelectronics.

The improvement of a thermal contact is associated with a decrease of the thermal resistance imposed by the interface as the heat flows across it.^{1–3} As surfaces are never perfectly flat, the interface comprises point contacts at asperities and air pockets. The number of contact points and the size of the air pockets depends upon the degree of roughness of the surfaces involved. Since air is a thermal insulator, the removal of the air helps improve a thermal contact. An effective method of displacing the air involves the use of a thermal interface material (a material placed at the interface).^{4–12} This material should conform to the surface topo-

graphy of the mating surfaces, thereby displacing the air. Furthermore, this material should be thin in the direction perpendicular to the interface, as the thermal resistance associated with the interface material itself increases with increasing thickness of the thermal interface.^{10,11} Of course, a high thermal conductivity is preferred for the thermal interface material, and the greater the thickness of the material, the more important is its thermal conductivity. Conformability is also needed for any thickness of the interface material.

A thermal interface material that is thick—0.1 mm or more—is needed for filling the gap between the two surfaces, particularly when the surfaces are not in direct contact (as encountered when each surface is curved so that gaps exist at parts of the interface, or when the two surfaces are not exactly parallel). This category of thermal interface materials is known as gap-filling materials. They are to be distinguished from thermal interface materials that are thin—thickness of 0.01 mm or less—ideally just thick enough to fill the valleys in the topography of the mating surfaces. This paper addresses the thin type, although the materials investigated may be useable as gap-filling materials as well.

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The thin type of thermal interface material, as mentioned above, is mainly in the form of thermal pastes, which are comprised of a base medium (the vehicle) and a filler that is thermally conductive. The filler is typically in the form of particles, as it is more difficult to make a workable paste that contains fibers (discontinuous) rather than particles. The workability relates to the conformability, although the type required for a thermal paste is conformability to the surface topography in a fine scale, so that the extent of flow associated with conforming is small. In contrast, workability usually refers to the ability to flow or deform in a relatively coarse scale. Conformability in a fine scale is more challenging than that in a coarse scale. The viscosity, which is a measure of the resistance to flow, is commonly used to describe the rheology of pastes.⁸ However, similar to the workability, the viscosity relates to the ability to flow in a relatively coarse scale.

The workability and conformability of a thermal paste diminish with increasing filler content, although the thermal conductivity within the paste increases with increasing filler content. Prior work on the development of thermal pastes has emphasized the attainment of a workable paste that has a high filler content. The approach largely involves the use of surfactants and the treatment of the surfaces of the filler particles (e.g., BN, ZnO, Al₂O₃) for the purpose of improving the workability, and the use of particles of different sizes in the same paste for the purpose of increasing the filler content. In contrast, recent work by Leong and Chung^{10,11} took the approach of using carbon black, a filler (solid) that is itself conformable and is in the form of porous agglomerates of nanoparticles. Due to the porous agglomerate microstructure, carbon black is highly compressible and is, hence, conformable in the solid state. Due to the small size (30 nm) of the nanoparticles in carbon black, conformability is possible in a fine scale. Thus, a thermal paste with carbon black as the conductive filler exhibits exceptional effectiveness as a thermal interface material.^{10,11}

During use, it is preferred that a thermal paste not seep out of the interface, as the seepage can cause contamination and, in the case of an electrically conductive paste (such as carbon black paste), short-circuiting of the electronics around the thermal contact. Therefore, a thixotropic paste (a paste that flows only under stress) is preferred to a fluidic paste (a paste that flows even in the absence of stress). The carbon black thermal pastes developed in prior work^{10,11} are fluidic. In this work, carbon black thermal pastes that are thixotropic have been developed. This development involves the use of a vehicle that is thixotropic.

Boron nitride (BN) (hexagonal) is a thermally conductive filler that is commercially used in high grades of thermal pastes. Zinc oxide is a less expensive and less conductive filler that is com-

mercially used in lower grades. This paper includes a comparative study of carbon black pastes and BN pastes.

Graphite differs from carbon black in its higher degree of crystallinity, greater degree of preferred crystallographic orientation, larger crystallite size, larger particle size, and the absence of a porous agglomerate structure. These attributes make graphite a superior thermal conductor, at least in the in-plane direction of graphite. On the other hand, the absence of a porous agglomerate structure and the relatively large particle size (typically around 1 μm or more) make graphite much less conformable than carbon black. This paper includes a comparative study of carbon black pastes and graphite pastes.

In order to understand the relative effectiveness of the various thermal pastes, the conformability of the pastes needs to be measured. However, there is no standard method of conformability testing. As explained above, conformability cannot be indicated by viscosity measurement. The closest relevant method is the penetration test, which is used for testing ketchup and other pastes and involves measurement of the distance of penetration of an indenter (a cone) that is allowed to penetrate the paste freely (by gravity) for a controlled length of time (5 sec). This work modified the standard penetration test for testing a small volume of low viscosity paste. The modification involves the use of a lightweight rod penetrator and a cylindrical vial with a relatively large aspect ratio for containing the paste. Thus, this work provides conformability data, which are correlated with data on the thermal contact conductance.

The objectives of this paper are as follows: (1) to develop a thixotropic form of carbon black thermal paste, (2) to compare the performance of thixotropic thermal pastes that involve various conductive fillers (carbon black, BN, and graphite), and (3) to correlate the thermal contact conductance with the conformability. The work involved paste preparation, thermal contact conductance measurement, conformability testing, and thermogravimetric analysis (for assessing the ability to resist elevated temperatures).

EXPERIMENTAL METHODS

The formulation, ingredients, preparation method, and testing method of the thermal pastes of this work are identical to those in the prior work of these authors.¹¹ 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 m²/g, maximum ash content 0.2%, volatile content 1.07%, and density 1.7–1.9 g/cm³. 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 that of the metal or ceramic particles used in commercial thermal pastes.

The graphite particles used were natural crystalline flakes, grade Micro 850, 98.5% minimum carbon content, 5 μm typical size (as provided by Asbury Graphite Mills, Asbury, NJ). The BN particles were hexagonal BN, equiaxed in shape (as shown by scanning electron microscopy), 5–11 μm size, 280 W/m-K thermal conductivity, as provided by Advanced Ceramics Corporation, Cleveland, OH (Polartherm 180).

The vehicle used 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.

All of the pastes in this work, regardless of the type of filler, were prepared by manual stirring of a mixture of filler and vehicle. The uniformity of the mixing is indicated by the consistency of the testing data obtained from various specimens from the same batch of paste.

Various thermal pastes were sandwiched between the flat surfaces of two copper discs (both surfaces of each disc having been mechanically polished by using 0.05 μm alumina particles in the case of smooth mating surfaces, and 15 μm silicon carbide particles in the case of rough mating surfaces), which had a diameter of 12.6 mm and a thickness of 1.16 mm for one disc, and 1.10 mm for the other disc.

The thermal contact conductance between two copper discs with a thermal interface material between them was measured using the transient laser flash method.^{8,9,13,14} The pressure on the sandwich was controlled at 0.46 MPa, 0.69 MPa, and 0.92 MPa, as in Refs. 10 and 11. This is because the pressure affects the thermal contact conductance, even for a material that is not resilient.⁹ The thickness of the thermal interface material was 25 μm or less. The uniform distribution of the paste in the plane of the sandwich was made possible by the flow of the paste under the applied pressure. The interface material thickness was obtained by subtracting the thicknesses of the two copper discs from the thickness of the sandwich, such that all thicknesses were measured using a micrometer. The interface material thickness for all cases was the same before and after the conductance measurement.

A Coherent General Everpulse Model 11 Nd glass laser (Santa Clara, CA) with a pulse duration of 0.4 ms, a wavelength of 1.06 μm , and a pulse energy up to 15 J was used for impulse heating. The laser power was adjusted to allow the temperature rise of the specimen to be between 0.5°C and 1.0°C. Details of the setup for thermal diffusivity testing using the laser flash method can be found in Ref. 11. Calibration using a standard NBS 8426 graphite disc (thickness = 2.62 mm) was performed before testing each specimen in order to ensure measurement accuracy.

The data acquisition rate used for each test was adjusted so that there were at least 100 temperature data points during the temperature rise.

The experimental error in transient thermal contact conductance measurement consists of random error due to experimental data scatter, and systematic error, mainly due to the lag of the thermocouple response, and partly due to the method used to calculate the conductance from the temperature data. The higher the thermal contact conductance, the greater is the error. The thermal diffusivity of a standard NBS 8426 graphite disk, which had a similar transient temperature rise time as the copper sandwich with the highest thermal contact conductance, was measured prior to testing each specimen in order to determine the systematic error, if any. The random error shown by the “ \pm ” value was determined from measurement for at least four specimens, such that each specimen was measured at least two times, with the pressure increased in two steps from 0.46 MPa to 0.92 MPa in each round of measurement.

Figure 1 illustrates the penetration testing instrument, which is a gravity-penetrator based on ASTM D 1321-97. The penetrator (a 15-5 precipitation-hardened stainless steel rod of diameter 0.51 mm, length 195 mm, and mass 0.33 g) was precisely lowered to the surface of the paste under test. Then it was allowed to sink into the paste by its own weight during a defined test duration (5 sec in this work). The penetration was indicated by the extent of vertical movement of the penetrator, as measured by calipers. The instrument in this work used a timer to control the test duration of 5 sec. A solenoid connected to the timer was used to activate a simple mechanism for stopping the fall of the penetrator exactly at the end of 5 sec. Each specimen was tested three times.

Ideally, a thermal paste is electrically insulating. Thus, this work includes measurement of the DC volume electrical resistivity of the thermal pastes. This was conducted by filling a mold cavity of size 70 mm \times 10 mm \times 0.38 mm with the paste, such

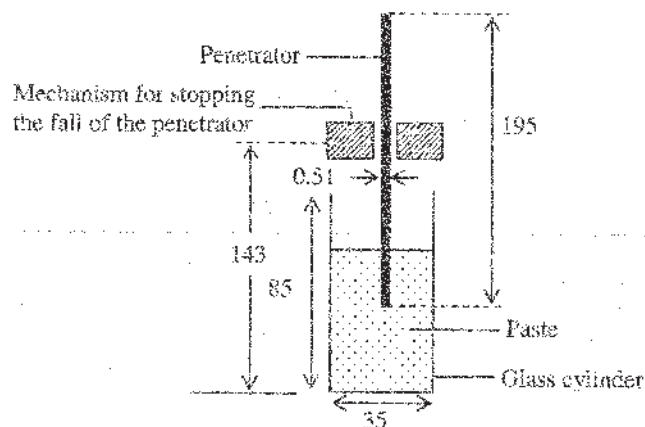


Fig. 1. Experimental setup for the penetration test for assessing the conformability of a paste. All dimensions are in millimeters.

that the top surface of the paste was flush with that of the mold. Due to the very high resistance of all the specimens, the two-probe method was used. The two electrical contacts were in the form of silver paint in conjunction with copper wires (diameter = 0.13 mm), which were parallel to one another extending across the entire 10-mm width of the mold cavity, at a distance of 10 mm from one another. Each wire drooped down from the mold wall into the shallow mold cavity. Thus, each wire was immersed in the paste, though it protruded out of the paste on both sides of the mold. Each specimen was measured three times, using a Keithley 2002 digital multimeter (Cleveland, OH).

A thermal paste should be able to withstand elevated temperatures (up to about 150°C when used for microelectronic cooling). Therefore, this work includes measurement of the weight loss during heating in air, using a Perkin-Elmer Corp. (Wellesley, MA). TGA7 thermogravimetric analyzer. The temperature was raised from 25°C to 100°C at a rate of 3°C/min. Then the temperature was held at 100°C for 24 h.

RESULTS AND DISCUSSION

Effects of Filler Type, Filler Content, Pressure, and Surface Roughness on the Thermal Contact Conductance

Table I gives the thermal contact conductance values obtained for various combinations of thermal paste, mating surface roughness, and contact pressure. For both smooth (0.05 μm) and rough (15 μm) mating surfaces, the conductance is highest for the paste containing 2.4 vol.% carbon black. Among the three types of filler, carbon black is the most effective and BN is the least effective, in spite of the high BN volume fraction (14–18%) compared to the low carbon black volume fraction (2.2–2.6%). Graphite is less effective than carbon black, in spite of its higher volume fraction and greater crystallinity. Neverthe-

less, graphite is much more effective than BN, in spite of its lower volume fraction (10.7–12.0%). The different ranges of filler volume fraction for the different fillers reflect the different upper limits of the filler volume fraction for forming a workable paste. The effectiveness of any of the three fillers depends on the filler volume fraction. For carbon black as the filler, the thermal contact conductance is highest at the intermediate carbon black volume fraction of 2.4%. For BN as the filler, the conductance is highest at the lowest BN volume fraction of 14.0%. For graphite as the filler, the conductance is higher at the lower graphite volume fraction of 10.7%. These trends occur at any of the pressures. The variation of the conductance with pressure is relatively small. In most cases, the conductance increases slightly with increasing pressure.

For the same paste, the conductance is much lower for the rough surface than the smooth surface, as previously reported for fluidic pastes.¹⁰ This is due to the thermal resistance within the paste, and the increase of this resistance with increasing paste thickness. Nevertheless, the relative effectiveness of the three fillers remains the same for rough and smooth surfaces.

Comparison of the carbon black thixotropic paste data in Table I with the carbon black fluidic paste data of Refs. 10 and 11 shows that the thermal performance is similar for these pastes. The highest thermal contact conductance attained is $30 \times 10^4 \text{ W/m}^2\cdot^\circ\text{C}$ for both the thixotropic pastes (this work) and the fluidic pastes.¹¹ This similarity is in spite of the relatively low fluidity of the thixotropic pastes. The observation that the fluidity of the vehicle has negligible influence on the carbon black paste performance is consistent with the previous report¹¹ that carbon black polyethylene glycol (PEG)-based paste and carbon black ether-based paste are similar in thermal effectiveness, in spite of the higher viscosity of the latter. However, the optimum carbon

Table I. Thermal Contact Conductance ($10^4 \text{ W/m}^2\cdot^\circ\text{C}$) for Various Combinations of Thermal Paste, Mating Surface Roughness, and Contact Pressure

Filler	Smooth Surfaces			Rough Surfaces		
	0.46 MPa	0.69 MPa	0.92 MPa	0.46 MPa	0.69 MPa	0.92 MPa
2.6 vol.% CB ^a	19.05 \pm 0.93	22.26 \pm 1.34	23.38 \pm 0.96	—	—	—
2.4 vol.% CB ^a	28.78 \pm 1.02	30.03 \pm 2.31	30.43 \pm 1.25	8.73 \pm 0.27	10.79 \pm 0.45	11.26 \pm 0.36
2.2 vol.% CB ^a	22.52 \pm 0.35	23.81 \pm 1.00	22.81 \pm 0.56	—	—	—
18.0 vol.% BN ^b (Ref. 8)	18.9 \pm 0.8	—	—	—	—	—
18.0 vol.% BN ^a	8.44 \pm 0.35	7.14 \pm 0.36	7.21 \pm 0.22	—	—	—
16.0 vol.% BN ^a	8.86 \pm 0.25	12.73 \pm 0.33	11.11 \pm 0.22	2.89 \pm 0.07	3.83 \pm 0.11	4.79 \pm 0.19
14.0 vol.% BN ^a	8.95 \pm 0.32	11.80 \pm 0.91	12.23 \pm 0.81	—	—	—
12.0 vol.% graphite ^a	16.35 \pm 0.59	19.58 \pm 0.84	21.97 \pm 1.21	—	—	—
10.7 vol.% graphite ^a	20.19 \pm 1.64	25.84 \pm 1.84	27.66 \pm 1.19	8.28 \pm 0.51	8.94 \pm 0.27	8.46 \pm 0.33
Ceramique ^c (Ref. 10)	21.48 \pm 1.12	24.10 \pm 0.76	25.59 \pm 0.96	7.21 \pm 0.10	8.47 \pm 0.53	9.92 \pm 0.41

CB = carbon black

^a Thixotropic paste with polyol ester vehicle.

^b Fluidic paste with lithium-doped PEG vehicle.

^c Commercial paste of Arctic Silver, Inc., with synthetic oil vehicle.

Table II. Conformability (as Shown by the Depth of Penetration) and Volume Electrical Resistivity of Various Thixotropic Thermal Pastes

Filler	Penetration Depth (mm)	Electrical Resistivity ($\Omega\text{-cm}$)
2.4 vol.% CB	45.54 ± 0.53	$(3.2 \pm 0.7) \times 10^3$
16.0 vol.% BN	20.07 ± 0.56	*
10.7 vol.% graphite	29.54 ± 1.07	$(1.32 \pm 0.07) \times 10^5$

CB = carbon black

* Too high to be measured.

black volume fraction is different for these pastes. For the thixotropic pastes of this work, the optimum carbon black volume fraction is 2.4%. For the fluidic pastes that utilize a vehicle in the form of PEG with 3 vol.% dissolved ethyl cellulose, the optimum carbon black volume fraction is 1.25%.¹¹ For the fluidic pastes that utilize a vehicle in the form of butyl ether with 40 vol.% dissolved ethyl cellulose, the optimum carbon black volume fraction is 0.20%.¹¹ The optimum filler content depends on the effects of the filler content on both the conformability and the thermal conductivity within the paste. Different vehicles are associated with different degrees of conformability, thus resulting in different values of the optimum filler content.

The thermal contact conductance is much lower for the thixotropic BN pastes of this work than the fluidic BN paste based on lithium doped PEG,⁸ as shown in Table I. The BN used in Ref. 8 and in this work are identical in source, grade, and volume fraction (18%). The higher conductance for the fluidic paste is probably due to the greater fluidity and the resulting higher conformability. The importance of fluidity or conformability to the thermal paste performance is also indicated by the fact that the BN paste decreases in thermal contact conductance as the BN content increases from 14 vol.% to 18 vol.% (Table I).

The fact that the BN thixotropic paste is less effective than the BN fluidic paste, whereas the carbon black thixotropic paste is comparable in effectiveness to the carbon black fluidic paste, is attributed to the large particle size of BN (5–11 μm) compared to carbon black (30 nm). In paste formulation, vehicle fluidity is more important for large particles than for small particles.

The thermal contact conductance is much lower for the thixotropic BN pastes of this work than some of those that are commercially available. For example, the paste known as Ceramique (Arctic Silver Inc., Visalia, CA) contains BN, zinc oxide, and aluminum oxide submicron particles, but without metal particles. Its thermal contact conductance is much higher than the BN pastes of this work for both smooth and rough surfaces (Table I). The low conductance of the BN pastes of this work is attributed mainly to the large particle size of BN (5–11 μm) used in this work and the submicron particle size of BN used in Ceramique.

Effect of the Filler on the Conformability

A high conformability is indicated by a large depth of penetration. As shown in Table II, the conformability is highest for the carbon black paste, lowest for the BN paste, and intermediate for the graphite paste. Correlation of the data in Tables I and II shows that a high thermal contact conductance indeed is associated with a high conformability. The penetration test could not be performed on fluidic pastes, such as those based on PEG, due to the off scale (excessive) extent of penetration.

Effect of Prior Pressure Application on the Thermal Contact Conductance

Table III shows that prior application of a pressure above 0.32 MPa (specifically 0.46 MPa in history 1 and 0.69 MPa in history 2) helps increase the conductance at 0.32 MPa. The prior pressure is higher in history 2 than history 1. The higher the prior pressure is, the greater the effect of the prior pressure application. The effect of prior pressure application is similar for the thixotropic and fluidic pastes, as shown in Table II.

The effect of prior pressure application means that a high pressure does not have to be applied during the use of the thermal paste. The prior application of a high pressure advantageously reduces the need for pressure application during use.

Electrical Resistivity of Thermal Pastes

Table II shows that the carbon black thixotropic paste is considerably lower in electrical resistivity than the graphite thixotropic paste, in spite of the lower volume fraction and lower crystallinity of carbon black. This is attributed to the good dispersion

Table III. Effect of Pressure History on Thermal Contact Conductance ($10^4 \text{ W/m}^2\text{-}^\circ\text{C}$) for Smooth Surfaces

Paste	History	0.32 MPa	0.46 MPa	0.69 MPa	0.32 MPa
Thixotropic ^a	1	19.02 ± 0.57	28.78 ± 1.02	—	20.24 ± 0.71
Thixotropic ^a	2	19.02 ± 0.57	—	30.03 ± 2.31	25.04 ± 1.16
Fluidic ^b	1	21.85 ± 1.94	29.90 ± 0.79	—	23.08 ± 1.20
Fluidic ^b	2	21.85 ± 1.94	—	28.98 ± 2.11	26.76 ± 1.92

^a Polyol ester vehicle with 2.4 vol.% carbon black.^b PEG vehicle with 3 vol.% ethyl cellulose and 1.25 vol.% carbon black.

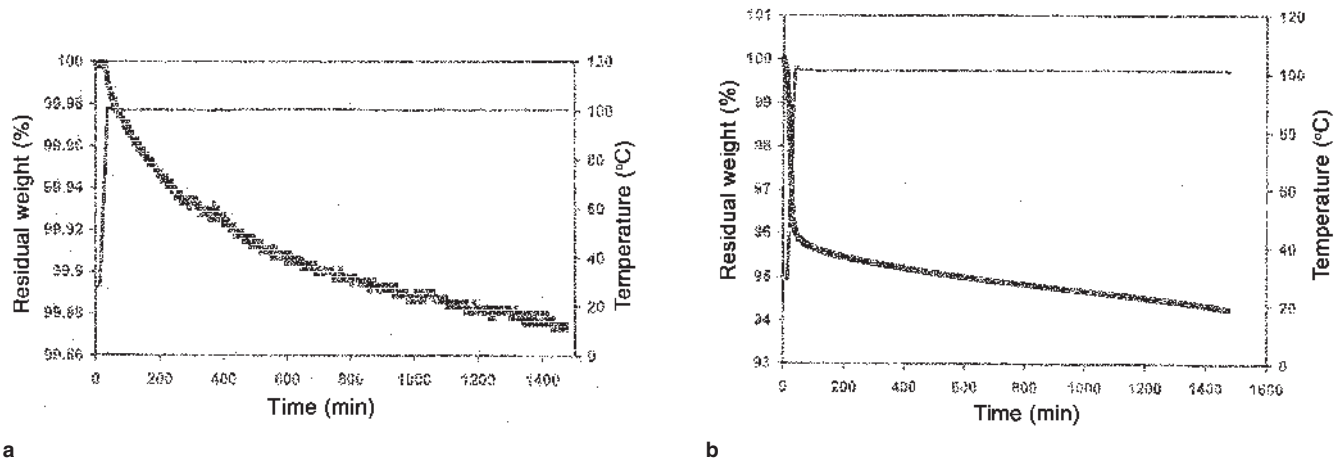


Fig. 2. Residual weight versus time during thermogravimetric analysis. (a) Thixotropic paste in the form of polyol ester vehicle with 2.4 vol.% carbon black. (b) Fluidic paste in the form of PEG vehicle with 3 vol.% dissolved ethyl cellulose and 1.25 vol.% carbon black.

of carbon black in the paste. Nevertheless, the resistivity of the carbon black paste is relatively higher, and the resistivity of the BN paste is so high that it cannot be measured.

Thermal Stability of Thermal Pastes

Figure 2a and b show the residual weight versus time during the thermogravimetric analysis for the thixotropic paste with 2.4 vol.% carbon black and the fluidic paste based on PEG and containing 3 vol.% dissolved ethyl cellulose and 1.25 vol.% carbon black, respectively. Under the same heating schedule, the fractional weight loss reached 0.13% for the former and 5.5% for the latter. Thus, the polyol ester vehicle is superior in thermal stability to the PEG vehicle.

CONCLUSIONS

Carbon black (30 nm particles) thixotropic paste (based on polyol esters) is comparable to carbon black fluidic paste (based on PEG) in its effectiveness as a thermal paste and in its dependence on pressure history, as shown by thermal contact conductance measurement. The performance is improved by prior compression (up to 0.69 MPa) of the thermal contact. With the optimum carbon black volume fraction of 2.4 vol.%, the thixotropic paste attains a thermal contact conductance of $30 \times 10^4 \text{ W/m}^2\text{-}^\circ\text{C}$ across mating copper surfaces of roughness $0.05 \mu\text{m}$ and $11 \times 10^4 \text{ W/m}^2\text{-}^\circ\text{C}$ across mating copper surfaces of roughness $15 \mu\text{m}$. However, the BN (5–11 μm particles) thixotropic thermal paste is much less effective than the boron-nitride fluidic paste at the same BN volume fraction (18%), due to the large size

of the BN particles. Both BN and graphite (5 μm) thixotropic pastes are less effective than the carbon black pastes, due to their lower conformability, as shown by penetration testing. The BN paste is up to 70% less effective and the graphite paste is up to 25% less effective. The volume electrical resistivity of the carbon black thixotropic paste is $3 \times 10^3 \Omega\text{-cm}$.

REFERENCES

1. E.G. Wolff and D.A. Schneider, *Int. J. Heat Mass Transfer* 41, 3469 (1998).
2. T. Ouellette and M. De Sorgo, *Proc. Power Electronic Design Conf.* (Cerritos, CA: Power Sources Users Conf., 1985), pp. 134–138.
3. M.R. Vogel, *Proc. Int. Intersociety of Electronic Packaging Conf.* (New York: ASME, 1995), p. 989.
4. S.W. Wilson, A.W. Norris, E.B. Scott, and M.R. Costello, *National Electronic Packaging and Production Conf.: Proc. Technical Program* (Norwalk, CT: Reed Exhibition Companies, 1996), pp. 788–796.
5. A.L. Peterson, *Proc. 40th Electronic Components and Technology Conf.* (Piscataway, NJ: IEEE, 1990), pp. 613–619.
6. X. Lu, G. Xu, P.G. Hofstra, and R.C. Bajcar, *J. Polym. Sci. Pol. Phys.* 36, 2259 (1998).
7. T. Sasaki, K. Hisano, T. Sakamoto, S. Monma, Y. Fijmori, H. Iwasaki, and M. Ishizuka, *Jpn. IEMT Symp. Proc.: IEEE/CPMT Int. Electronic Manufacturing Technology (IEMT) Symp.* (Piscataway, NJ: IEEE, 1995), p. 236.
8. Y. Xu, X. Luo, and D.D.L. Chung, *J. Electron. Packaging* 124, 188 (2002).
9. Y. Xu, X. Luo, and D.D.L. Chung, *J. Electron. Packaging* 122, 128 (2000).
10. C.-K. Leong and D.D.L. Chung, *Carbon* 42, 2323 (2004).
11. C.-K. Leong and D.D.L. Chung, *Carbon* 41, 2459 (2003).
12. Q. Ngo, B.A. Cruden, A.M. Cassell, G. Sims, M. Meyyappan, J. Li, and C.Y. Yang, *Nano Lett.* 4, 2403 (2004).
13. W.J. Parker, R.J. Jenkins, C.P. Butler, and G.L. Abbott, *J. Appl. Phys.* 32, 1679 (1961).
14. K. Inoue and E. Ohmura, *Yosetsu Gakkai Ronbunshu/Q. J. Jpn. Welding Soc.* 6, 130 (1988).