



Materials for thermal conduction

D.D.L. Chung*

Composite Materials Research Laboratory, State University of New York at Buffalo, Buffalo, NY 14260-4400, USA

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Abstract

Materials for thermal conduction are reviewed. They include materials exhibiting high thermal conductivity (such as metals, carbons, ceramics and composites), and thermal interface materials (such as polymer-based and silicate-based pastes and solder). © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Thermal conduction; Thermal conductivity; Thermal paste; Thermal contact; Composite; Interface

1. Introduction

The transfer of heat by conduction is involved in the use of a heat sink to dissipate heat from an electronic package, the heating of an object on a hot plate, the operation of a heat exchanger, the melting of ice on an airport runway by resistance heating, the heating of a cooking pan on an electric range, and in numerous industrial processes that involve heating or cooling. Effective transfer of heat by conduction requires materials (such as a heat sink material) of high thermal conductivity. In addition, it requires a good thermal contact between the two surfaces (such as the surface of a heat sink and the surface of a printed circuit board) across which heat transfer occurs. Without good thermal contacts, the use of expensive thermal conducting materials for the components is not cost-effective. The attainment of a good thermal contact requires a thermal interface material, such as a thermal grease, which must be thin (small in thickness) between the mating surfaces, must conform to the topography of the mating surface and preferably should have a high thermal conductivity as well. This paper is a review of materials for thermal conduction, including materials of high thermal conductivity and thermal interface materials.

* Tel.: +1-716-645-2593, ext. 2243; fax: +1-716-645-3875.

E-mail address: ddlchung@acsu.buffalo.edu (D.D.L. Chung).

2. Materials of high thermal conductivity

Materials of high thermal conductivity are needed for the conduction of heat for the purpose of heating or cooling. One of the most critical needs is in the electronic industry. Due to the miniaturization and increasing power of microelectronics, heat dissipation is key to the reliability, performance and further miniaturization of microelectronics. The heat dissipation problem is so severe that even expensive thermal conductors such as diamond, metal-matrix composites and carbon-matrix composites are being used in high-end microelectronics. Due to the low coefficient of thermal expansion (CTE) of semiconductor chips and their substrates, heat sinks also need to have low CTE. Thus, the requirement for the thermal conductor material is not just high thermal conductivity, but low CTE as well. For example, copper is a good thermal conductor but its CTE is high. Therefore, copper-matrix composites containing low CTE fillers such as carbon fibers or molybdenum particles are used. For lightweight electronics, such as laptop computers and avionics, an additional requirement for the thermal conductor material is low density. As aluminum and carbon are light compared to copper, aluminum, carbon and their composites are used for this purpose. Compared to aluminum, carbon has the additional advantage of being corrosion resistant.

2.1. Metals, diamond and ceramics

Table 1 gives the thermal conductivity of various metals. Copper is most commonly used when materials of high thermal conductivity are required. However, copper suffers from a high value of the CTE. A low CTE is needed when the adjoining component has a low CTE. When the CTE of the two adjoining materials are sufficiently different and the temperature is varied, thermal stress occurs and may even cause warpage. This is the case when copper is used as heat sink for a printed wiring board, which is a continuous fiber polymer-matrix composite that has a lower CTE than copper. Molybdenum and tungsten are metals that have low CTE, but their thermal conductivity is poor compared to copper. The alloy Invar (64Fe–36Ni) is outstandingly low in CTE among metals, but it is very poor in thermal conductivity. Diamond is most attractive, as it has very high

Table 1
Thermal properties and density of various materials

Material	Thermal conductivity (W/m K)	CTE ($10^{-6}/^{\circ}\text{C}$)	Density (g/cm^3)
Aluminum	247	23	2.7
Gold	315	14	19.32
Copper	398	17	8.9
Lead	30	39	11
Molybdenum	142	4.9	10.22
Tungsten	155	4.5	19.3
Invar	10	1.6	8.05
Kovar	17	5.1	8.36
Diamond	2000	0.9	3.51
Beryllium oxide	260	6	3
Aluminum nitride	320	4.5	3.3
Silicon carbide	270	3.7	3.3

thermal conductivity and low CTE, but it is expensive. Aluminum is not as conductive as copper, but it has a low density, which is attractive for aircraft electronics and applications (e.g., laptop computers) which require low weight [2,3]. Aluminum nitride is not as conductive as copper, but it is attractive in its low CTE. Diamond and most ceramic materials are very different from metals in their electrical insulation ability. In contrast, metals are conducting both thermally and electrically. For applications which require thermal conductivity and electrical insulation, diamond and appropriate ceramic materials can be used, but metals cannot.

2.2. *Metal-matrix composites*

One way to lower the CTE of a metal is to form a metal-matrix composite [1] by using a low CTE filler. Ceramic particles such as AlN and silicon carbide (SiC) are used for this purpose, due to their combination of high thermal conductivity and low CTE. As the filler usually has lower CTE and lower thermal conductivity than the metal matrix, the higher the filler volume fraction in the composite, the lower the CTE and the lower is the thermal conductivity.

Metal-matrix composites with discontinuous fillers (commonly particles) are attractive for their processability into various shapes. However, layered composites in the form of a matrix-filler-matrix sandwich are useful for planar components. Discontinuous fillers are most commonly ceramic particles. The filler sheets are most commonly low CTE metal alloy sheets (e.g., Invar or 64Fe–36Ni, and Kovar or 54Fe–29Ni–17Co). Aluminum and copper are common metal matrices due to their high conductivity.

2.2.1. *Aluminum-matrix composites*

Aluminum is the most dominant matrix for metal-matrix composites for both structural and electronic applications. This is because of the low cost of aluminum and the low melting point of aluminum (660°C) facilitating composite fabrication by methods that involve the melting of the metal.

Liquid-phase methods for the fabrication of metal-matrix composites include liquid metal infiltration, which usually involves using pressure (from a piston or compressed gas) to push the molten metal into the pores of a porous preform comprising the filler (commonly particles that are not sintered) and a small amount of a binder [4–6]. Pressureless infiltration is less common but is possible [7,8]. The binder prevents the filler particles from moving during the infiltration, and also provides sufficient compressive strength to the preform, so that the preform will not be deformed during the infiltration. This method thus provides near-net-shape fabrication, i.e., the shape and size of the composite product are the same as those of the preform. Since machining of the composite is far more difficult than that of the preform, near-net-shape fabrication is desirable.

In addition to near-net-shape fabrication capability, liquid metal fabrication is advantageous in being able to provide composites with high filler volume fractions (up to 70%). A high filler volume fraction is necessary in order to attain a low enough CTE ($<10 \times 10^{-6} \text{ (}^\circ\text{C}^{-1}\text{)}$) in the composite, even if the filler is a low CTE ceramic (e.g., SiC), since the aluminum matrix has a relatively high CTE [9,10]. However, to attain a high volume fraction using liquid metal infiltration, the binder used must be in a small amount (so as not to clog the pores in the preform) and still be effective. Hence, the binder technology [11–13] is critical.

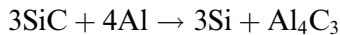
The ductility of a composite decreases as the filler volume fraction increases, so that a composite with a low enough CTE is quite brittle. Although the brittleness is not acceptable for structural applications, it is acceptable for electronic applications.

Another liquid-phase technique is stir casting [1], which involves stirring the filler in the molten metal and then casting. This method suffers from the non-uniform distribution of the filler in the composite due to the difference in density between filler and molten metal and the consequent tendency for the filler to either float or sink in the molten metal prior to solidification. Stir casting also suffers from the incapability of producing composites with a high filler volume fraction.

Yet another liquid-phase technique is plasma spraying [14], which involves spraying a mixture of molten metal and filler onto a substrate. This method suffers from the relatively high porosity of the resulting composite and the consequent need for densification by hot isostatic pressing or other methods, which tend to be expensive.

A solid-phase technique is powder metallurgy, which involves mixing the matrix metal powder and filler and subsequent sintering under heat and pressure [14]. This method is relatively difficult for the aluminum matrix because aluminum has a protective oxide on it and the oxide layer on the surface of each aluminum particle hinders sintering. Furthermore, this method is usually limited to low volume fractions of the filler.

The most common filler used is silicon carbide (SiC) particles, due to the low cost and low CTE of SiC [15]. However, SiC suffers from its reactivity with aluminum. The reaction is



It becomes more severe as the composite is heated. The aluminum carbide is a brittle reaction product which lines the filler–matrix interface of the composite, thus weakening the interface. Silicon, the other reaction product, dissolves in the aluminum matrix, lowering the melting temperature of the matrix, and causing non-uniformity in the phase distribution and mechanical property distribution [16]. Furthermore, the reaction consumes a part of the SiC filler [17].

A way to diminish this reaction is to use an Al–Si alloy matrix, since the silicon in the alloy matrix promotes the opposite reaction and is thus against this reaction. However, the Al–Si matrix is less ductile than the Al matrix, thus causing the mechanical properties of the Al–Si matrix composite to be very poor compared to those of the corresponding Al-matrix composite. Thus, the use of an Al–Si alloy matrix is not a solution to the problem.

An effective solution is to replace SiC by aluminum nitride (AlN) particles, which do not react with aluminum, thus resulting in superior mechanical properties in the composite [18]. The fact that AlN tends to have a higher thermal conductivity than SiC helps the thermal conductivity of the composite. Since the cost of the composite fabrication process dominates the cost of producing the composites, the higher material cost of AlN compared to SiC does not matter, especially for electronic packaging. Aluminum oxide (Al₂O₃) also does not react with aluminum, but it is low in thermal conductivity and tends to suffer from particle agglomeration [18].

Other than ceramics such as SiC and AlN, another filler used in aluminum-matrix composites is carbon in the form of fibers of diameter around 10 μm [19–23] and less commonly, filaments of diameter less than 1 μm [24]. Carbon also suffers from reactivity with aluminum to form aluminum carbide. However, fibers are more effective than particles for reducing the CTE of the composite. Carbon fibers can be even continuous in length. Moreover, carbon, especially when graphitized, is much more thermally conductive than ceramics. In fact, carbon fibers that are sufficiently gra-

phitic are even more thermally conductive than the metal matrix, so that the thermal conductivity of the composite increases with increasing fiber volume fraction. However, these fibers are expensive. The mesophase-pitch-based carbon fiber K-1100 from Amoco Performance Products (Alpharetta, GA) exhibits longitudinal thermal conductivity 1000 W/m K [25,26].

Both carbon and SiC suffer from forming a galvanic couple with aluminum, which is the anode – the component in the composite that is corroded. The corrosion becomes more severe in the presence of heat and/or moisture.

The thermal conductivity of aluminum-matrix composites depends on the filler and its volume fraction, the alloy matrix heat treatment condition, as well as the filler–matrix interface [18,27].

To increase the thermal conductivity of SiC aluminum-matrix composite, a diamond film can be deposited on the composite [28]. The thermal conductivity of single crystal diamond is 2000–2500 W/m K, though a diamond film is not single crystalline.

2.2.2. *Copper-matrix composites*

Because copper is heavy anyway, the filler does not have to be lightweight. Thus, low CTE but heavy metals such as tungsten [29,30], molybdenum [31,32] and Invar [33–35] are used as fillers. These metals (except Invar) have the advantage that they are quite conductive thermally and are available in particle and sheet forms, so that they are suitable for particulate as well as layered [36,37] composites. Yet another advantage of the metal fillers is the better wettability of the molten matrix metal with metal fillers than with ceramic fillers, in case the composite is fabricated by a liquid phase method.

An advantage of copper over aluminum is its nonreactivity with carbon, so carbon is a highly suitable filler for copper. Additional advantages are that carbon is lightweight and carbon fibers are available in a continuous form. Furthermore, copper is a rather noble metal, as shown by its position in the Electromotive Series, so it does not suffer from the corrosion which plagues aluminum. Carbon used as a filler in copper is in the form of fibers of diameter around 10 μm [22,38–45]. As carbon fibers that are sufficiently graphitic are even more thermally conductive than copper, the thermal conductivity of a copper-matrix composite can exceed that of copper.

Less common fillers for copper are ceramics such as silicon carbide, titanium diboride (TiB_2) and alumina [46–48].

The melting point of copper is much higher than that of aluminum, so the fabrication of copper-matrix composites is commonly done by powder metallurgy, although liquid metal infiltration is also used [22,49,50]. In the case of liquid metal infiltration, the metal matrix is often a copper alloy (e.g., Cu–Ag) chosen for the reduced melting temperature and good castability [50].

Powder metallurgy conventionally involves mixing the metal matrix powder and the filler, and subsequent pressing and then sintering under either heat or both heat and pressure. The problem with this method is that it is limited to low volume fractions of the filler. In order to attain high volume fractions, a less conventional method of powder metallurgy is recommended. This method involves coating the matrix metal on the filler units, followed by pressing and sintering [32,46,51,52]. The mixing of matrix metal powder with the coated filler is not necessary, although it can be done to decrease the filler volume fraction in the composite. The metal coating on the filler forces the distribution of matrix metal to be uniform even when the metal volume fraction is low (i.e., when the filler volume fraction is high). On the other hand, with the conventional method, the matrix metal distribution is not uniform when the filler volume fraction is high, thus causing

porosity and the presence of filler agglomerates, in each of which the filler units directly touch one another; this microstructure results in low thermal conductivity and poor mechanical properties.

Continuous carbon fiber copper-matrix composites can be made by coating the fibers with copper and then diffusion bonding (i.e., sintering) [38,40,44,53–55]. This method is akin to the above mentioned less conventional method of powder metallurgy.

Less common fillers used in copper include diamond powder [50,56], aluminosilicate fibers [57] and Ni–Ti alloy rod [58]. The Ni–Ti alloy is attractive for its negative CTE of -21×10^{-6} ($^{\circ}\text{C}^{-1}$).

The coating of a carbon fiber copper-matrix composite with a diamond film has been done to enhance the thermal conductivity [43].

2.2.3. Beryllium-matrix composites

Beryllium oxide (BeO) has a high thermal conductivity (Table 1). Beryllium-matrix BeO-platelet composites with 20–60 vol.% BeO exhibit low density (2.30 g/cm^3 at 40 vol.% BeO, compared to 2.9 g/cm^3 for Al/SiC at 40 vol.% SiC), high thermal conductivity (232 W/m K at 40 vol.% BeO, compared to 130 W/m K for Al/SiC at 40 vol.% SiC), low CTE (7.5×10^{-6} ($^{\circ}\text{C}^{-1}$)) at 40 vol.% BeO, compared to 12.1×10^{-6} ($^{\circ}\text{C}^{-1}$) at 40 vol.% SiC), and high modulus (317 GPa at 40 vol.% BeO, compared 134 GPa for Al/SiC at 40 vol.% SiC) [59,60].

2.3. Carbon-matrix composites

Carbon is an attractive matrix for composites for thermal conduction because of its thermal conductivity (though not as high as those of metals) and low CTE (lower than those of metals). Furthermore, carbon is corrosion resistant (more corrosion resistant than metals) and lightweight (lighter than metals). Yet another advantage of the carbon matrix is its compatibility with carbon fibers, in contrast to the common reactivity between a metal matrix and its fillers. Hence, carbon fibers are the dominant filler for carbon-matrix composites. Composites with both filler and matrix being carbon are called carbon–carbon composites [61]. Their primary applications in relation to thermal conduction are heat sinks [62], thermal planes [63] and substrates [64]. There is considerable competition between carbon–carbon composites and metal-matrix composites for the same applications.

The main drawback of carbon-matrix composites is their high cost of fabrication, which typically involves making a pitch-matrix or resin-matrix composite and subsequent carbonization (by heating at $1000\text{--}1500^{\circ}\text{C}$ in an inert atmosphere) of the pitch or resin to form a carbon-matrix composite. After carbonization, the porosity is substantial in the carbon matrix, so pitch or resin is impregnated into the composite and then carbonization is carried out again. Quite a few impregnation–carbonization cycles are needed in order to reduce the porosity to an acceptable level, thus resulting in the high cost of fabrication. Graphitization (by heating at $2000\text{--}3000^{\circ}\text{C}$ in an inert atmosphere) may follow carbonization (typically in the last cycle) in order to increase the thermal conductivity, which increases with the degree of graphitization. However, graphitization is an expensive step. Some or all of the impregnation–carbonization cycles may be replaced by chemical vapor infiltration (CVI), in which a carbonaceous gas infiltrates the composite and decomposes to form carbon.

Carbon–carbon composites have been made by using conventional carbon fibers of diameter around $10 \mu\text{m}$ [62,63,65], as well as carbon filaments grown catalytically from carbonaceous gases

and of diameter less than 1 μm [24]. By using graphitized carbon fibers, thermally conductivities exceeding that of copper can be reached.

To increase the thermal conductivity, carbon–carbon composites have been impregnated with copper [66,67] and have been coated with a diamond film [68].

2.4. Carbon and graphite

An all-carbon material (called ThermalGraph, a tradename of Amoco Performance Products, Alpharetta, GA), made by consolidating oriented precursor carbon fibers without a binder and subsequent carbonization and optional graphitization, exhibits thermal conductivity ranging from 390 to 750 W/m K in the fiber direction of the material.

Another material is pyrolytic graphite (called TPG) encased in a structural shell [69]. The graphite (highly textured with the *c*-axes of the grains preferentially perpendicular to the plane of the graphite), has an in-plane thermal conductivity of 1700 W/m K (four times that of copper), but it is mechanically weak due to the tendency to shear in the plane of the graphite. The structural shell serves to strengthen by hindering shear.

Pitch-derived carbon foams with thermal conductivity up to 150 W/m K after graphitization are attractive for their high specific thermal conductivity (thermal conductivity divided by the density) [70].

2.5. Ceramic-matrix composites

The SiC matrix is attractive due to its high CTE compared to the carbon matrix, though it is not as thermally conductive as carbon. The CTE of carbon–carbon composites is too low ($0.25 \times 10^{-6} \text{ } (^{\circ}\text{C}^{-1})$), thus resulting in reduced fatigue life in chip-on-board (COB) applications with silica chips ($\text{CTE} = 2.6 \times 10^{-6} \text{ } (^{\circ}\text{C}^{-1})$). The SiC-matrix carbon fiber composite is made from a carbon–carbon composite by converting the matrix from carbon to SiC [65]. To improve the thermal conductivity of the SiC-matrix composite, coatings in the form of chemical vapor deposited AlN or Si have been used. The SiC-matrix metal (Al or Al–Si) composite, as made by a liquid-exchange process, also exhibits relatively high thermal conductivity [71].

The borosilicate glass matrix is attractive due to its low dielectric constant (4.1 at 1 MHz for $\text{B}_2\text{O}_3\text{--SiO}_2\text{--Al}_2\text{O}_3\text{--Na}_2\text{O}$ glass), compared to 8.9 for AlN, 9.4 for alumina (90%), 42 for SiC, 6.8 for BeO, 7.1 for cubic boron nitride, 5.6 for diamond and 5.0 for glass–ceramic. A low value of the dielectric constant is desirable for electronic packaging applications. On the other hand, glass has a low thermal conductivity. Hence, fillers with relatively high thermal conductivity are used with the glass matrix. An example is continuous SiC fibers, the glass-matrix composites of which are made by tape casting, followed by sintering [72]. Another example is aluminum nitride with interconnected pores (about 28 vol.%), the composites of which are obtained by glass infiltration to a depth of about 100 μm [72–74].

3. Thermal interface materials

Thermal interface materials are needed to improve thermal contacts. By placing a thermal interface material at the interface between two components across which heat must flow, the

thermal contact between the two components is improved. A primary market for thermal interface materials is in the electronic industry, as heat dissipation is critical to the performance, reliability and further miniaturization of microelectronics. For example, a thermal interface material is used to improve the thermal contact between a heat sink and a printed circuit board, or between a heat sink and a chip carrier.

A thermal interface material can be a thermal fluid, a thermal grease (paste), a resilient thermal conductor or solder which is applied in the molten state. A thermal fluid, thermal grease or molten solder is spread on the mating surfaces. A resilient thermal conductor is sandwiched by the mating surfaces and held in place by pressure. Thermal fluids are most commonly mineral oil. Thermal greases (pastes) are most commonly conducting particle (usually metal or metal oxide) filled silicone. Resilient thermal conductors are most commonly conducting particle filled elastomers. Out of these four types of thermal interface materials, thermal greases (based on polymers, particularly silicone) and solder are by far most commonly used. Resilient thermal conductors are not as well developed as thermal fluids or greases.

As the materials to be interfaced are good thermal conductors (such as copper), the effectiveness of a thermal interface material is enhanced by high thermal conductivity and low thickness of the interface material and low thermal contact resistance between the interface material and each mating surface. As the mating surfaces are not perfectly smooth, the interface material must be able to flow or deform, so as to conform to the topography of the mating surfaces. If the interface material is a fluid, grease or paste, it should have a high fluidity (workability) so as to conform and to have a small thickness after mating. On the other hand, the thermal conductivity of the grease or paste increases with increasing filler content and this is accompanied by decrease in the workability. Without a filler, as in the case of an oil, the thermal conductivity is poor. A thermal interface material in the form of a resilient thermal conductor sheet (e.g., a felt consisting of conducting fibers clung together without a binder, and a resilient polymer-matrix composite containing a thermally conducting filler) usually cannot be as thin or conformable as one in the form of a fluid, grease or paste, so its effectiveness requires a very high thermal conductivity within it.

Solder is commonly used as a thermal interface material for enhancing the thermal contact between two surfaces. This is because solder can melt at rather low temperatures and the molten solder can flow and spread itself thinly on the adjoining surfaces, thus resulting in high thermal contact conductance at the interface between solder and each of the adjoining surfaces. Furthermore, solder in the metallic solid state is a good thermal conductor. In spite of the high thermal conductivity of solder, the thickness of the solder greatly affects the effectiveness of the solder as a thermal interface material; a small thickness is desirable. Moreover, the tendency for solder to react with copper to form intermetallic compounds [75] reduces the thermal contact conductance of the solder–copper interface.

Thermal pastes are predominantly based on polymers, particularly silicone [76–79], although thermal pastes based on sodium silicate have been reported to be superior in providing high thermal contact conductance [80]. The superiority of sodium–silicate-based pastes over silicone-based pastes is primarily due to the low viscosity of sodium silicate compared to silicone, and the importance of high fluidity in the paste so that the paste can conform to the topography of the surfaces which it interfaces.

A particularly attractive thermal paste is based on polyethylene glycol (PEG, a polymer) of a low molecular weight (400 amu) [81]. These pastes are superior to silicone-based pastes and are as

good as sodium-silicate-based pastes due to the low viscosity of PEG and the contribution of lithium ions (a dopant) in the paste to thermal conduction. Compared to the sodium-silicate-based pastes, the PEG-based pastes are advantageous in their long-term compliance, in contrast to the long-term rigidity of sodium silicate. Compliance is attractive for decreasing the thermal stress, which can cause thermal fatigue.

Table 2 gives the thermal contact conductance for different thermal interface materials. Included in the comparison are results obtained with the same testing method on silicone-based

Table 2

Thermal contact conductance for various thermal interface materials between copper discs at 0.46 Ma contact pressure [80,81]

Thermal interface material		Interface material thickness (μm) (± 10)	Thermal contact conductance ($10^4 \text{ W/m}^2 \text{ }^\circ\text{C}$)	Viscosity (cps) (± 0.3)
Description	Designation			
PEG	A	<25	11.0 ± 0.3	127 ^a
PEG + BN (18 vol.%)	B	25	12.3 ± 0.3	–
PEG + Li salt (1.5 wt.%)	C	<25	12.3 ± 0.3	143 ^a
PEG + Li salt (1.5 wt.%) + BN (18 vol.%)	D	25	13.4 ± 0.4	–
PEG + water + DMF	J	<25	12.5 ± 0.2	75.6 ^a
J + Li salt (0.75 wt.%)	E	<25	11.4 ± 0.3	79.7 ^a
J + Li salt (1.5 wt.%)	F	<25	16.0 ± 0.5	85.6 ^a
J + Li salt (3.0 wt.%)	G	<25	11.6 ± 0.2	99.0 ^a
J + Li salt (4.5 wt.%)	H	<25	9.52 ± 0.25	117 ^a
J + Li salt (6.0 wt.%)	I	<25	7.98 ± 0.16	120 ^a
F + BN (16.0 vol.%)	F ₁	25	18.5 ± 0.8	–
F + BN (18.0 vol.%)	F ₂	25	18.9 ± 0.8	–
F + BN (19.5 vol.%)	F ₃	25	15.3 ± 0.2	–
F + BN (21.5 vol.%)	F ₄	25	14.0 ± 0.5	–
G + BN (16.0 vol.%)	G ₁	25	17.0 ± 0.5	–
G + BN (18.0 vol.%)	G ₂	25	17.3 ± 0.6	–
G + BN (19.5 vol.%)	G ₃	25	14.9 ± 0.6	–
G + BN (21.5 vol.%)	G ₄	25	13.4 ± 0.4	–
H + BN (18.0 vol.%)	H ₁	25	13.9 ± 0.4	–
Solder	–	25	20.8 ± 0.6^b	–
Sodium silicate + BN (16.0 vol.%)	–	25	18.2 ± 0.7	–
Sodium silicate + BN (17.3 vol.%)	–	25	15.5 ± 0.4	–
Sodium silicate	–	<25	14.1 ± 0.5	206 ^c
Silicone/BN	–	25	10.9 ± 1.5	–
Silicone	–	<25	3.08 ± 0.03	8800 ^d
None	–	–	0.681 ± 0.010	–

^a Measured using the Ubbelohde method.

^b At zero contact pressure (not 0.46 MPa).

^c Measured using the Ostwald method.

^d Value provided by the manufacturer.

paste, sodium-silicate-based pastes and solder [80,81]. PEG (i.e., A) gives much higher thermal contact conductance ($11.0 \times 10^4 \text{ W/m}^2\text{°C}$) than silicone ($3.08 \times 10^4 \text{ W/m}^2\text{°C}$), due to its relatively low viscosity, but the conductance is lower than that given by sodium silicate ($14.1 \times 10^4 \text{ W/m}^2\text{°C}$), in spite of its low viscosity, due to the molecular nature of PEG. The addition of the Li salt (1.5 wt.%) to PEG (i.e., to obtain C) raises the conductance from 11.0×10^4 to $12.3 \times 10^4 \text{ W/m}^2\text{°C}$, even though the viscosity is increased. The further addition of water and DMF (i.e., F) raises the conductance to $16.0 \times 10^4 \text{ W/m}^2\text{°C}$ and decreases the viscosity. Thus, the addition of water and DMF is very influential, as water and DMF help the dissociation of the lithium salt. The still further addition of BN particles (18.0 vol.%) (i.e., F₂) raises the conductance to $18.9 \times 10^4 \text{ W/m}^2\text{°C}$. The positive effect of BN is also shown by comparing the results of C and D (which are without water or DMF) and by comparing the results of A and B (which are without Li⁺). In the absence of the lithium salt, water and DMF also help, though not greatly, as shown by comparing A and J. The viscosity increases with the lithium salt content, as shown by comparing J, E, F, G, H and I. Comparison of E, F, G, H and I shows that the optimum lithium salt content for the highest conductance is 1.5 wt.%. That an intermediate lithium salt content gives the highest conductance is probably because of the enhancement of the thermal conductivity by the Li⁺ ions and the increase of the viscosity caused by the lithium salt addition. Both high conductivity and low viscosity (good conformability) are desirable for a high contact conductance. Comparison of F₁, F₂, F₃ and F₄ shows that the optimum BN content is 18.0 vol.%, as also indicated by comparing G₁, G₂, G₃ and G₄. Among all the PEG-based pastes, the highest conductance is given by F₂, as it has the optimum lithium salt content as well as the optimum BN content. An optimum in the BN content also occurs for BN filled sodium-silicate-based pastes [80]. It is due to the increase in both the thermal conductivity and the viscosity of the paste as the BN content increases. The best PEG-based paste (i.e., F₂) is similar to the best sodium-silicate-based paste in the conductance. Both are better than BN filled silicone, but both are slightly inferior to solder. Although solder gives the highest conductance, it suffers from the need of heating during soldering. In contrast, heating is not needed in the use of PEG-based pastes, silicone-based pastes or silicate-based pastes.

4. Conclusion

Materials for thermal conduction include materials exhibiting high thermal conductivity, as well as thermal interface materials. The former includes metals, diamond, carbon, graphite, ceramics, metal-matrix composites, carbon-matrix composites and ceramic-matrix composites. The latter includes polymer-based pastes, silicate based pastes and solder. A primary area of application is in the microelectronic industry.

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