

Diamond-based Metal Matrix Composites for Thermal Management made by Liquid Metal Infiltration—Potential and Limits

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Abstract. Diamond-based metal matrix composites have been made based on pure Al and eutectic Ag-3Si alloy by gas pressure infiltration into diamond powder beds with the aim to maximize thermal conductivity and to explore the range of coefficient of thermal expansion (CTE) that can be covered. The resulting composites covered roughly the range between 60 and 75 vol-% of diamond content. For the Al-based composites a maximum thermal conductivity at room temperature of 7.6 W/cmK is found while for the Ag-3Si based composites an unprecedented value of 9.7 W/cmK was achieved. The CTE at room temperature varied as a function of the diamond volume fraction between 3.3 and 7.0 ppm/K and 3.1 and 5.7 ppm/K for the Al-based and the Ag-3Si-based composites, respectively. The CTE was further found to vary quite significantly with temperature for the Al-based composites while the variation with temperature was less pronounced for the Ag-3Si-based composites. The results are compared with prediction by analytical modeling using the differential effective medium scheme for thermal conductivity and the Schapery bounds for the CTE. For the thermal conductivity good agreement is found while for the CTE a transition of the experimental data from Schapery's upper to Schapery's lower bound is observed as volume fraction increases. While the thermophysical properties are quite satisfactory, there is a trade-off to be made in these materials between high thermal conductivity and low CTE on the one side and surface quality and machinability on the other.

Introduction

Thermal management of electronic devices has received quite some attention lately as heat dissipation density in components like CPU's, diode laser arrays, and power electronic elements increases much faster than the cooling capacity of passive substrate materials or active cooling systems. In passive substrate materials, high thermal conductivity and tailored CTE is key [1-4]. Due to its excellent thermal conductivity diamond as a reinforcement in metal matrix composites has received quite some attention lately and diamond based composites based on aluminum [5-7], magnesium [6], copper [8-11] and silver [12] and their alloys have seen the day. The achievable properties in terms of thermal conductivity and CTE of the resulting composite depend on the volume fraction of diamond and on the matrix metal for both properties, while diamond quality and size affects further the thermal conductivity [12]. The most important quality parameter of the diamond is its nitrogen content as it affects directly its intrinsic thermal conductivity [13, 14]. Standard metal bond diamond from various sources in China have been assessed with regard to their nitrogen content the lowest values being around 100-120 ppm, which corresponds to an intrinsic thermal conductivity of at most 17-18 W/cmK at RT [13, 14]. The highest thermal conductivity for Al-based diamond composites reported in the literature is 6.7 W/cmK [15], while for Ag-3Si a value of 7.7 W/cmK has been published [12].

The diamond size has its influence through the interface thermal resistance at metal/diamond interfaces due to phonon scattering at the interface. The interface thermal resistance reduces the effective conductivity of an inclusion, i.e. in our case the diamond, and does so the more, the shorter the distance between the intersection of two interfaces with the heat flux lines is. Therefore, larger diamonds offer better thermal conductivity. On the other hand, the use of large diamond particles

has also a direct impact on machinability, surface roughness and characteristic length-scale (and, thus, homogeneity) of the microstructure.

In view of these antagonist interests with regard to diamond particle size, we present in what follows (i) an assessment of what the maximum thermal conductivity of metals reinforced with commercial diamond particles would be and how close we have come to this point; (ii) what range of CTE can be covered with diamond based composites made by gas pressure infiltration and (iii) very shortly discuss some approaches how machinability and surface quality could be improved without sacrificing too much of the outstanding thermal conductivity.

Experimental procedure

Diamond based composites were made by gas pressure assisted infiltration of liquid metals into tapped and vibrated beds of synthetic industrial diamond. The metals used were pure (99.99%) aluminum and custom made eutectic Ag-3wt-pct Si alloy. The diamond powder of particularly nice cube-octahedral shape and low nitrogen content from the LD series was purchased from China Abrasives Import and Export Corp. Hainan branch at the largest available grit size, i.e. 450 μm for LD8110 and LD8120 and 350 μm for LD8160, for optimized composites with monomodal particle size distribution. The small sized (52 μm average size) MBD4 diamond for the bimodal mixtures was purchased from Qiming Superabrasive Materials, Henan, China. These diamonds have been chosen for their low nitrogen content and thus for their high thermal conductivity. The various LD qualities were taken to investigate the influence of price (a parameter including deviations from the ideal cuboctahedral shape and the level of catalyst inclusions) on thermal conductivity. The diamond qualities used, the ratio between large and small diamond particles, as well as the obtained overall diamond volume fractions are given in Tab. 1. The bimodal powders were simply mixed by putting the respective quantities of the powders in a phial and shaking it for a minute or so.

The powder was filled in a graphite mold that contained cavities shaped as the samples used for thermal conductivity and thermal expansion measurement. The graphite mould was inserted in an alumina crucible and covered with a metal ingot. The whole was placed in a pressure vessel that has a central zone that can be heated by induction using a graphite susceptor. After reaching a vacuum of better than 0.1 mbar the heating was switched on to reach the infiltration temperature (750°C for Al-based and 1070°C for Ag-3Si based composites) at a rate of roughly 200 K/hour up to 300°C and then at a rate of 500K/hour to infiltration temperature. After holding for roughly 40 min at infiltration temperature, argon gas pressure of 5-6 MPa was applied on the melt to push the metal

Table 1: Composition and physical properties of Ag- and Al-based composites

Matrix	Diamond quality	Vol. frac. Diamond [-]	Fraction of large diamonds [%]	Thermal conductivity [W/cmK]	CTE @RT [ppm/K]
Ag-3Si	-	-	-	3.7	17
Ag-3Si	LD8110	0.61	100	7.75	5.6
	LD8120	0.62	100	8.0	5.1
	LD8160	0.65	100	8.6	5.2
	LD8110+MBD4	0.73	71	9.6	3.3
	LD8120+MBD4	0.74	71	9.6	3.3
	LD8160+MBD4	0.76	71	9.7	3.1
Al	LD8110	0.64	100	6.8	6.6
	LD8120	0.62	100	6.85	7
	LD8160	0.63	100	7.6	5.7
	LD8110+MBD4	0.73	71	7.2	3.4
	LD8120+MBD4	0.72	71	7.0	3.7
	LD8160+MBD4	0.74	71	7.25	3.4

between the diamond particles. The induction heating was then switched off and temperature fell below the melting point of the metal within less than 15 min.

Thermal conductivity was measured by a steady-state comparative temperature gradient method at identical heat flux in, both, sample and reference. Pure (OFHC) copper was used as a reference that was calibrated with another piece of pure copper and pure aluminum prior to the experiment. The samples were measured in the as cast state for the aluminum based composites while the Ag-3Si based composites underwent a spheroidizing and precipitation treatment of 2h at 800°C and subsequent furnace cool down to 400°C with 24 hours of hold at 400°C prior to measurement. The CTE was measured in a TMA 402 from Netzsch in two cycles between -50 and 200°C at a rate of 5 K/min on cylindrical samples roughly 10 mm long and 4 mm diameter.

Results

The thermal conductivity of the various composites made in this work is given in Tab. 1. The thermal conductivity is highest for composites based on the highest quality diamond (LD8160) for which also the attainable volume fractions both in monomodal and bimodal composites are highest. The thermal conductivity covers the range of 6.8-7.6 W/cmK for the Al-based composites and 7.5-9.7 W/cmK for the Ag-3Si based composites, i.e. values quite significantly higher than those reported so far.

The physical CTE at RT of the composites lies in the range of 3.4 to 7.0 ppm/K and 3.1 to 5.6 ppm/K for the Al-based and Ag-based composites, respectively. The evolution of the CTE with volume fraction is shown in Fig. 1 to the left. It is worthwhile noting that other than for monolithic low expansion materials like Mo or W, the CTE of the composites depends quite strongly on temperature, cf. Fig. 1 to the right. This temperature dependence is less pronounced for Ag-based composites compared to their Al-based counterparts and generally decreases with increasing volume fraction of diamond.

Discussion

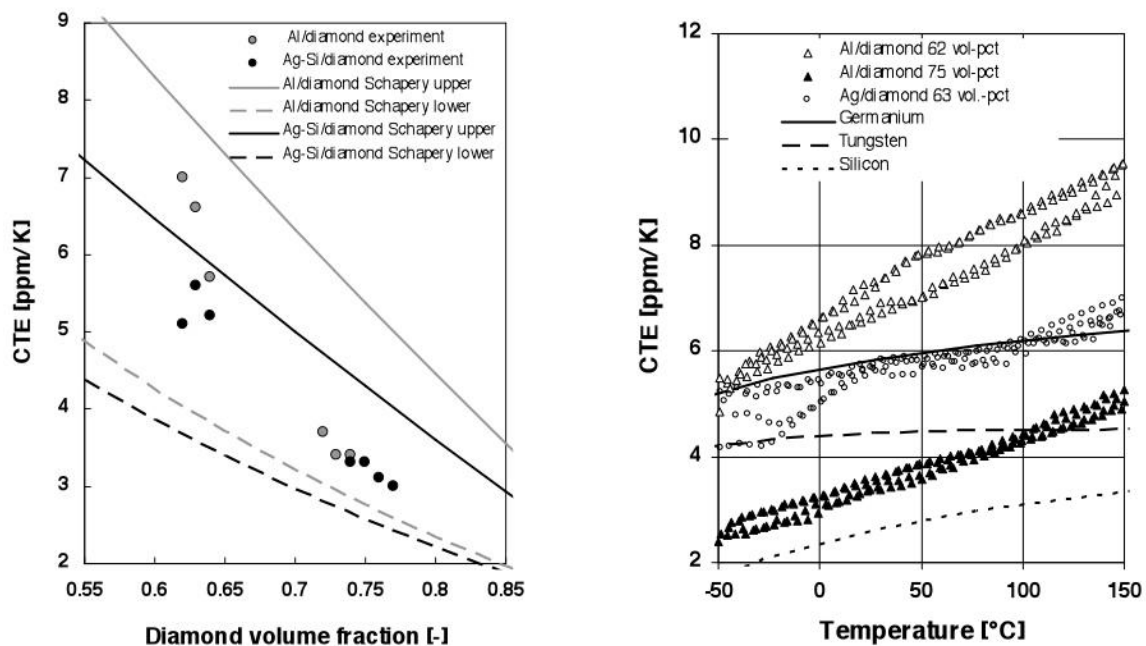


Fig. 1: Evolution of the CTE at room temperature with volume fraction of diamond (left) indicating a transition from Schapery's upper bound at lower volume fractions to Schapery's lower bound at high volume fraction. The change of CTE with temperature (right) is stronger than in comparable low CTE materials like W, Ge and Si. This effect is more pronounced in the Al-based composites than in their Ag-based counterparts and at lower diamond contents.

The data on CTE of the composites can be compared to the classical bounds by Schapery [16]. In Fig. 1 to the left a transition from Schapery's upper bound at lower volume fractions to Schapery's lower bound at higher volume fractions can be inferred. This can be rationalized that at high volume fraction diamond particles touch each other and will oppose further contraction as the matrix tries to contract. This is equivalent to the presence of solid bridges between the diamond particles, which would then act at least partially as matrix phase. For lower volume fractions the number of contact points is more limited and the diamonds are acting as inclusion phase, which would be in agreement with the topology underlying the calculations of the two Schapery bounds.

The quite important changes in CTE with temperature especially for the Al-based composites are due to the fact that both the aluminum matrix and the diamond change their CTE quite rapidly around room temperature, the aluminum matrix because of its low melting point and the diamond because of the strongly increasing number of thermally activated phonons.

The thermal conductivity is higher than all previously reported values and the question whether this is the limit that can be attained quite naturally arises. We can compare the values with predictions by the differential effective medium scheme (DEM), which has previously been shown to be appropriate for prediction of thermal conductivity in metal diamond composites [12]. The finite size of the diamond particles is taken into account by using the effective thermal conductivity concept [12] and for bimodal composites the two particle populations are treated as two different particles (because of their difference in effective conductivity), cf. Ref. [14]. The assessment of the limits is based on the maximum intrinsic conductivity of commercial synthetic diamond, i.e., 17.5 W/cmK, the maximum packing density for monomodal powders and a fixed ratio of large to small particles for the bimodal composites of 3:1. For the interface thermal conductivity a value of $6.6 \cdot 10^7$ W/m²K [12] and $5 \cdot 10^7$ W/m²K [17] for Ag-Si-based and Al-based composites, respectively, was taken. In the former, the addition of Si to the matrix is crucial, as pure silver bonds only weakly to the diamond. As shown in Fig. 2, the theoretical limits for monomodal composites with Al- and Ag-matrix have pretty much been reached. For the bimodal composites there seems to be still some potential improvement especially if the packing of the bimodal powders can be improved. To do so, the fraction of large particles should be increased compared to the fraction used in this work.

While large diamonds are advantageous from an optimized thermal conductivity standpoint, machinability and achievable surface quality after machining would clearly require much smaller diamonds. Comparable composite conductivity at smaller particle sizes can only be achieved if the

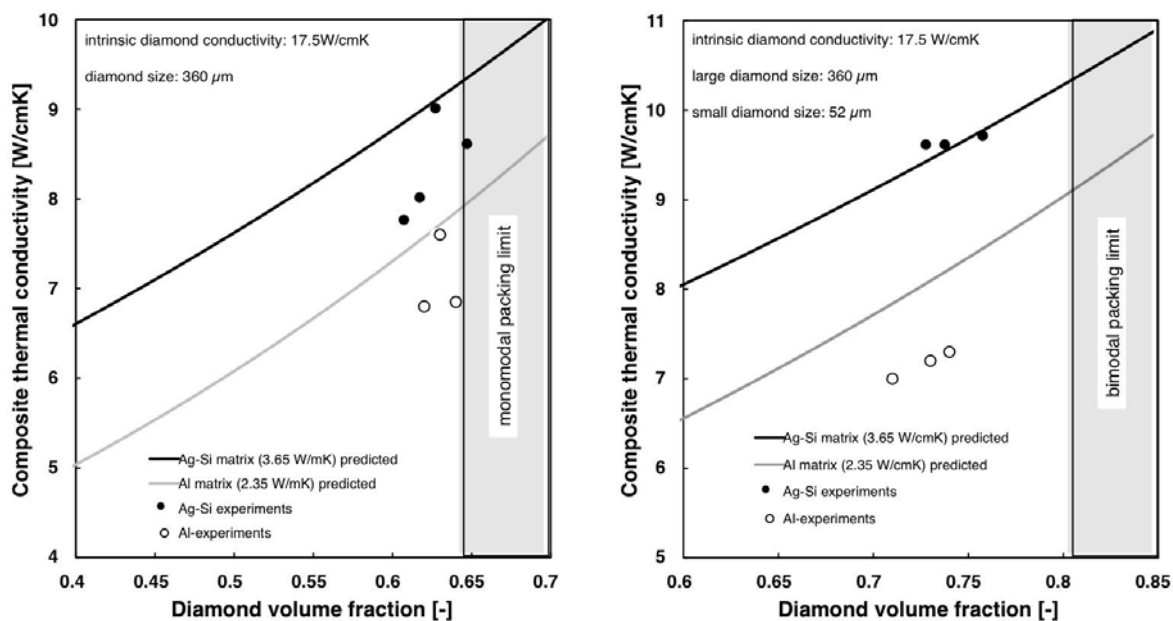


Fig. 2: Comparison of experimental thermal conductivity with predictions of the DEM scheme. For monomodal composites the limit is virtually attained (left) while for bimodal powder size distributions further optimization especially in the packing strategy is possible (right).

interface thermal conductance between matrix and particle can be improved. This may be achieved by geometrically increase the interface available for heat transfer by surface structuring of the diamond, by intelligent interlayer design, e.g. by introducing electrically conducting interlayers with high Debye-temperature that provide improved heat transfer with both the matrix and the diamond, or by introducing solid direct contacts between the diamond. The latter approach, however, may generate new problems of machinability.

Summary

We have shown that diamond composites made by gas pressure assisted liquid metal infiltration can reach exceptionally high thermal conductivity approaching 10 W/cmK, i.e. 2.5 times the thermal conductivity of pure copper. The CTE depends mainly on the volume fraction of diamond and only slightly on the matrix used, especially at high volume fractions. The CTE varies quite significantly with temperature which must be taken into account at the design level. We foresee an upper limit of thermal conductivity at room temperature of 10.5 W/cmK and 8.5 W/cmK for Ag-based and Al-based composites, respectively. Machinability and surface quality would require smaller diamonds which would lead to reduced thermal conductivity. This reduction may be limited by intelligent interface design.

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