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Optimizing thermal conductivity in gas-pressure infiltrated aluminum/diamond composites by precise processing control

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ABSTRACT

The effects of processing conditions on the thermal conductivity (TC) of aluminum/diamond composites fabricated by means of gas-pressure infiltration, have been evaluated using a device that allows a highly precise control of infiltration temperature and contact time between the liquid metal and diamond. Both variables have strong effects on the TC of final materials, through chemical and morphological modifications of the interface. Thus, maximizing heat conduction requires a proper choice of those two variables. TC's up to 670 W/mK have been obtained.

1. Introduction

The use of carbon-based composites as heat sinks in different applications that require high thermal performance, is steadily increasing in recent years [1–11]. In particular, the family of diamond–metal composites is attracting much attention. Depending on alloy and whether mono- or bimodal particle distributions are used, thermal conductivities (TC) in the range 130–970 W/mK have been reported [1–3,5–7]. Several issues related to Al/diamond composites are of major importance in developing high thermal conductivity materials, being the quality of the interface one of them. Improving the thermal conductance of the interface has been tackled using different methods depending on the system at hand.

Interesting results, comparing Al/SiC composites fabricated either by squeeze casting or gas-pressure infiltration, have been recently published [12]. The two techniques involve either a short (the former) or a long (the latter) contact time between liquid metal and SiC particles [12]. A maximum thermal conductivity was by means of squeeze casting. Actually, for long contact times, Al_4C_3 onto SiC particles and Si in the metal–matrix were observed, both phases formed through reaction between liquid Al and SiC. Si in solid solution was main responsible for low TC values. Fast infiltration has also proven to be beneficial as it reduces reaction products on SiC particles which may significantly decrease the permeability of particle preforms and thus hindering infiltration [13].

Similar studies have been performed on Al/diamond composites and some of the findings are highlighted hereafter: (i) adhesion between metal–matrix and diamond, which has been correlated with thermal conductivity, was found to depend strongly on the processing conditions [3,5]; (ii) significant bonding at the metal–diamond interface is achieved only during gas pressure infiltration, which in this aspect appears to perform better than squeeze-casting [3–5]; (iii) long contact times promotes interfacial bonding through formation of Al_4C_3 enhancing TC [3]. These findings support, explicitly or implicitly, the strong effect that contact time, which determines to a great extent the characteristics of the interface, has on thermal properties.

The present work is addressed to investigate the effects on the TC of Al/diamond composites not only of the contact time but also of the temperature of infiltration (a parameter to which less attention has been devoted). The study was carried out on compacts made out of a standard type of diamond particles gas pressure infiltrated with high purity aluminum.

2. Experimental procedures

2.1. Materials

Aluminum of 99.99 wt.% purity was supplied by Goodfellow Metals (Cambridge, England) and diamond particles were bought

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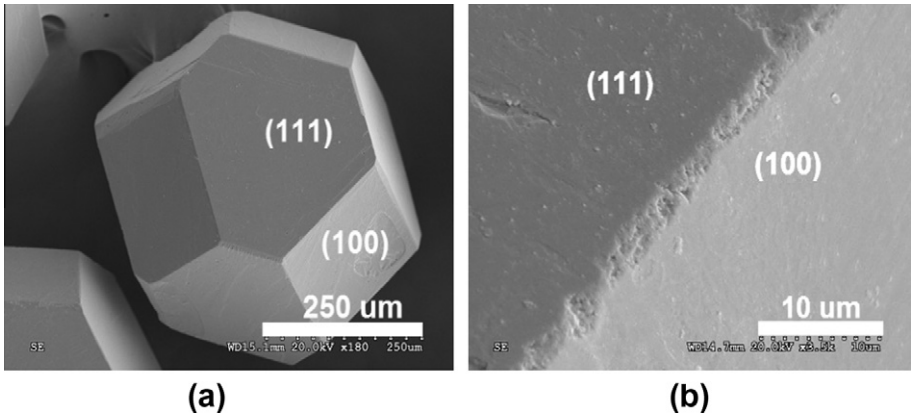

Fig. 1. SEM micrographs of a diamond particle of ISD1700 40/50 quality, (b) is a magnification of one of the sharp edges of the particle shown in (a).

from Iljin Diamond (Korea). Their characteristics and morphology are reported in Fig. 1 and Table 1, respectively. The ISD1700 diamond particles were also characterized by its nitrogen content, which was indirectly determined from absorption peaks at 1130 cm^{-1} and 2120 cm^{-1} in Infrared Spectrometry [14]. For the infrared absorption measurements, a Bruker Equinox 55 equipment was used, coupled with an IR-Scope II microscope. These measurements indicated the presence of about 200 ppm of nitrogen in diamond. Diamond particles were packed in the as-received condition into graphite crucibles following a procedure based on the wet packing technique described in Ref. [15]; in the present case, it was carried out using a weight ratio of 15:1 of ethanol:diamond. Composite samples were fabricated using crucibles of 36 mm height and 12 mm in diameter. The inner surface of the crucibles was coated with BN to facilitate demoulding. Densitometry allowed measuring particle volume fractions that were in the range 0.60–0.63.

2.2. Fabrication

A major difficulty in handling diamond-based materials is that standard machining, drilling, etc., operations cannot be performed. Thus, in order to facilitate the placement of the thermocouples needed for TC measurements, two small holes in the radial direction were machined 4 mm apart from each crucible end, into which pencil leads 0.9 mm in diameter were introduced 6 mm into the crucible space.

Preforms of diamond particles packed into those crucibles, were infiltrated with liquid aluminum by means of the gas pressure infiltration technique [16–18]. A solid piece of metal was placed on top of the packed preform and, prior to melting, vacuum was applied until a pressure of 0.2 mbar was reached. Then, the temperature (controlled within (4 °C) was increased up to the chosen infiltration temperature T_{inf} and, before proceeding, the system was maintained 10 min at temperature to allow attaining thermal equilibrium. Pressure in the chamber was increased up to five bars by using Argon gas. The infiltration process was carried out using a small pressure chamber, specially designed for allowing a rapid cooling and, thus, a rather accurate measurement of the time during which diamond particles and liquid aluminum were in contact (here referred to as contact time t_c).

Table 1
Characteristics of the ISD1700 diamond particles used in this work. D is the average diameter of the particles. The span of the size distribution is defined as $[D(90) - D(10)]/D(50)$, where $D(x)$ is the diameter below which x% of the particles are found. Both D and $D(x)$ are given in μm.

Diamond (mesh)	D	$D(90)$	$D(10)$	Span
ISD 1700 (40/50)	395	472	333	0.35

2.3. Thermal conductivity measurement

The TC of the composites was measured within two days after fabrication by means of a relative steady-state (equal-flow) technique, in an experimental setup assembled at the University of Alicante [18,19]. The cylindrical-shaped composite sample was placed in contact between a cold base (through which a flux of room temperature water is circulating) and a reference block (high purity copper 99.9998 wt.%) of the same dimensions through their circular bases. The reference block was connected to a stabilized hot water bath at 70 °C. The temperature gradients in the sample and the reference block, measured by means of two thermocouples in each of their respective ends, allowed determining the TC of the sample. The estimated error in TC was ±5%.

2.4. Interfacial characterization

The Al–diamond interface was characterized immediately after TC measurement by following a recently developed preparative method of metal electro-etching that preserves the interfacial reaction products [20]. The interface was analyzed by means of: (i) direct observation with scanning electron microscopy (S-3000N Hitachi microscope); and (ii) X-ray diffraction analysis (JSO-DEBYEFLEX 2002 Seifert diffractometer) immediately after etching.

Table 2
Thermal conductivity K (W/mK) of Al/diamond composites fabricated at two infiltration temperatures (T_{inf}) for different contact times. V_p refers to the volume fraction of diamond particles. Each data reported here is an average of several measurements corresponding to three independent samples (sample-to-sample variability is <5%).

Contact time (min)	T_{inf} = 760 °C		T_{inf} = 850 °C	
	K (W/mK)	V_p	K (W/mK)	V_p
0	411	0.63	582	0.62
0.5	394	0.62	656	0.62
0.75	–	–	674	0.62
1.0	–	–	676	0.63
1.5	–	–	636	0.62
2.0	425	0.63	–	–
5.0	481	0.62	–	–
10	609	0.62	–	–
15	636	0.61	609	0.61
20	626	0.60	–	–
30	600	0.62	–	–
45	568	0.60	614	0.62