Diamond & Related Materials 31 (2013) 1–5

Contents lists available at SciVerse ScienceDirect

Diamond & Related Materials

journal homepage: www.elsevier.com/locate/diamond

Diamond/aluminum composites processed by vacuum hot pressing: Microstructure characteristics and thermal properties

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ARTICLE INFO

Article history:
Received 5 May 2012
Received in revised form 26 September 2012
Accepted 3 October 2012
Available online 1 November 2012

Keywords:
Diamond/Al composites
Thermal conductivity
Interfacial bonding
Vacuum hot pressing

ABSTRACT

As promising thermal management materials, diamond/Al composites with 20–50 vol.% diamond were fabricated by a simple powder metallurgy method called vacuum hot pressing (VHP). The microstructure characteristics and thermal properties of the composites were studied. The results reveal that no aluminum carbide is formed at the interface and the VHP composite with 50 vol.% diamond exhibits a thermal conductivity of 496 W/mK, over 85% of the theoretical prediction by the differential effective medium (DEM) scheme, due to good interfacial bonding and high interface conductance. As a comparison, the composites consolidated by spark plasma sintering (SPS) exhibits lower thermal conductivity due to poor interfacial bonding. Thus, VHP is proved to be a more favorable way than SPS to fabricate diamond/Al composites with high thermal properties for heat management applications.

1. Introduction

Efficient heat removal is becoming increasingly urgent for the reliability and life spans of electronic components, owing to the continuously increased power density. It is essential to develop thermal management materials with high thermal conductivity (TC) and tailored coefficient of thermal expansion (CTE), which can not only dissipate the generated heat of electronic components, but also minimize thermal stress between the electronic components and the thermal management materials to ensure their reliability and lifetime [1–3]. Metal matrix composites (MMCs) are perfect candidates used as thermal management materials for good tailorability and formability. Recently, diamond/Al composites have been extensively investigated due to the combination of high TC and tailorable CTE, as well as the lower density compared to others [4–8].

Conventionally, liquid infiltration was mostly used for preparing diamond/Al composites and systematic work has been done [7–14]. However, in this process, aluminum carbide (Al_4C_3) was unavoidably formed at diamond and molten Al interface due to the elevated temperatures far above the Al melting point, usually 750–850 °C, which is believed to be detrimental to the composites for the brittleness, unstableness and poor TC of Al_4C_3 [8–12]. Therefore, it is the key point to control the formation of Al_4C_3 strictly in the fabrication of the composites for promising TC, while it is unfeasible for liquid infiltration due to the fast reaction kinetics [15]. On the other hand, powder metallurgy (PM) methods allow the consolidation of diamond and Al powders at temperatures below the Al melting point, thus the formation of Al_4C_3 could be avoided at diamond–Al interfaces [16–19].

However, so far, most consolidations of diamond/Al composites were completed by spark plasma sintering (SPS) for its high efficiency, usually with holding time as short as several minutes at rather low temperatures (only 520–600 °C). Accordingly, good interfacial bonding can hardly be attained in the SPS diamond/Al composites, thus causing poor TC [16,18]. Though, both the alloying of Al matrix and the metallization of diamond surface could somewhat improve the interfacial bonding, the TC of SPS diamond/Al composites degrades terribly due to the matrix alloying [9,20] and the introduction of additional coatings as thermal barriers [18]. Even worse, the SPS process becomes more complicated and expensive with alloying and/or metallization treatments [18,19]. Hence, it is very essential to develop alternative PM method that enable controllable diamond/Al interface to get good interfacial bonding and interface conductance without or with only traces of Al_4C_3.

The main objective of the present work is to investigate a PM method called vacuum hot pressing (VHP), to consolidate the powder compacts of diamond and pure Al. The microstructures and thermal properties of the composites were studied. The VHP composites with 20–50 vol.% diamond exhibit thermal conductivities of 320–496 W/mK, approaching the theoretical predictions, due to good interfacial bonding and favorable interface conductance without the formation of Al_4C_3. Compared with SPS, VHP is an effective way to fabricate diamond/Al composites with high thermal properties.

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0925-9635/$ – see front matter
http://dx.doi.org/10.1016/j.diamond.2012.10.008

2. Experimental

2.1. Materials

Synthetic diamond particles (Type HWD40, Henan Huanghe Whirlwind International Co. Ltd., China) with an average size of about 200 μm were used as reinforcements, whose nitrogen concentration was estimated to be 150–170 ppm from the Fourier transform infrared spectroscopy (FTIR) [21,22]. Accordingly, the TC of the diamond was estimated to be 1700–1850 W/mK according to the relationship between TC and the nitrogen concentration [23].

Atomized pure Al powders with average particle sizes of 75–105 μm were used as matrix, whose chemical composition were tested by inductively coupled plasma mass spectrometer (ICP-MS), as tabulated in Table 1. With Fe, Si and V elements as the main impurities, the Al powders were about 99.84% in purity.

2.2. Preparation of diamond/Al composites

The as-received diamond particles were first ultrasonicated in distilled water to eliminate impurities on the surface, and then they were dried and directly mixed with pure Al powders with a nominal volume fraction of 20–50%. The powder mixtures were first cold pressed into powder compacts, and then sintered by VHP in a graphite mould with inner and outer diameters of 10 and 75 mm, respectively. As illustrated in Fig. 1a, the graphite mould was heated by Mo coils and the temperature was monitored by a thermocouple mounted about 20 mm away from it. The indicated temperature fluctuated by ±2 °C around the set value. Fig. 1b shows a typical VHP procedure including five stages. The furnace was heated up to 400 °C by 10 °C/min (stage A), held for 30 min at 400 °C to degas the powder compacts (stage B). Afterwards, it was heated up to 650 °C and kept for 90 min (stage C and D), while a uniaxial pressure of 67.7 MPa was applied in stage D. Finally, after furnace cooling (stage E), disk-shaped samples with 3 mm in thickness and 10 mm in diameter were obtained. During the VHP process, a vacuum less than 0.005 Pa was maintained in the furnace.

As a comparison, the powder compacts were also sintered by SPS at 550 °C under a uniaxial pressure of 50 MPa for 5 min in vacuum less than 4 Pa, using the equipment of Model SPS SYNTEX DR. SINTER 2040. Details of the SPS process was described elsewhere [3,16–19]. The optimized sintering parameters in SPS and VHP process are tabulated in Table 2.

2.3. Characterizations

The thermal diffusivities of the sintered samples were measured by a laser flash technique using a Netzsch LFA447 thermal constant analyzer in the Applied Lab of Netzsch Company, Shanghai. The results represent the average value of three tests in all cases and the standard errors are smaller than ±2%. The density of the sintered samples was measured by the Archimedes method. TC of the sintered samples was calculated by the product of the density, thermal diffusivity, and specific heat capacity. The phase composition of the sintered samples was characterized by X-ray diffraction (XRD) using a D/max-2550 instrument (Cu Ka) at step scan rate of 4°/min in the ranges of 20–80°. The microstructure of the sintered samples was observed by scanning electron microscopy (SEM) in the backscattered electron (BSE) mode at 20 kV using an FEI Quanta FEG 250 electron microscope.

3. Results and discussion

3.1. Microstructure characteristics

XRD patterns of the composites with 40 vol.% diamond are shown in Fig. 2. Only Al and diamond phases are detected in the SPS and VHP composites. None of the peaks could be indexed as aluminum carbide, probably due to the fact that either no Al_4C_3 is formed at all or the content of Al_4C_3 as formed is too limited to be found. It is also verified that no Al_4C_3 is formed by the morphology of diamond particles in the fractured composites in Section 3.2. Therefore, it is effective to avoid the formation of Al_4C_3 in the PM diamond/Al composites, sintered both by SPS and by VHP, owing to rather lower processing temperatures (below Al melting point) than liquid infiltration [8–15].

The microstructures of diamond/Al composites are shown in Fig. 3. Plastic dimples of Al matrix (Fig. 3a and c) are clearly observed in both the SPS and VHP composites. The plastic dimples are considered as a sign of good sintering properties of the Al matrix, in which diamond particles are embedded. Obviously, there are more plastic dimples in the VHP composites than their SPS counterparts, which indicate better consolidation and higher deformation ability of Al matrix achieved during VHP. This is mainly ascribed to the higher sintering temperature and longer holding time in the process of VHP than SPS, as listed in Table 2, because VHP may coarsen Al grains in the sintering process while SPS always retains smaller ones [24,25].

The intact diamond particles (Fig. 3a and b) indicate poor diamond/Al interfacial bonding in the SPS composites, because the fracture often occurs at interfaces with weak interfacial bonding. As a comparison, good diamond/Al interfacial bonding is revealed in the VHP composites by the fact that, plastic fractures always occur within Al matrix rather than at diamond/Al interfaces, leaving the Al adhered diamond particles (Fig. 3c and d), due to preferred and strong adherence of Al on diamond {100} faces [9,12,15]. This indicates that in the VHP composites, the diamond/Al interfacial bonding is much stronger than the strength of Al matrix. Some cracked diamond particles (Fig. 3c), formed when the VHP composites were broken up, are also supposed to be relevant to the strong interfacial bonding, as analyzed in Ref. [26]. Therefore, VHP endows the composites with much better interfacial bonding than SPS, probably originating from sufficient interface diffusion.

In the solid-state PM process, interface diffusion occurs rather slowly, which makes it possible to tune interfacial bonding and interface conductance. However, the low sintering temperature and short holding time in SPS process always result in smaller matrix grains even with no interface diffusion [24,25]. Conducted at lower vacuum, higher sintering temperature and longer holding time, VHP enables sufficient interface diffusion and perfect interfacial bonding. Compared with VHP, liquid infiltration exhibits too fast reaction kinetics at elevated temperatures above the Al melting point, and usually results in excess interface diffusion and the formation of Al_4C_3 [15], while SPS always results in insufficient interface diffusion and poor interfacial bonding due to very slow reaction kinetics [18]. Therefore, VHP used in this work is feasible and effective to get good interfacial bonding for enhanced thermal properties.

3.2. Thermal properties

Fig. 4 shows the relative density of diamond/Al composites with 20–50 vol.% diamond. The relative densities of the VHP composites with 20–40 vol.% diamond are 2% higher than the SPS counterparts,

Table 1
Chemical composition of the pure Al powders used in this work.

Element	B	Cr	Cu	Fe	Mg	Ni	Pb	Si	Ti	V	Zn	Al
Mass (%)	0.0008	0.0006	0.0021	0.072	0.001	0.0026	0.0039	0.0362	0.0065	0.0397	0.0012	Bal.