



Microstructure and thermal properties of copper–diamond composites with tungsten carbide coating on diamond particles

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ARTICLE INFO

Article history:

Received 29 November 2013

Received in revised form 22 July 2014

Accepted 24 July 2014

Available online 25 July 2014

Keywords:

Metal matrix composites

Tungsten carbide

Coating

Microstructure

Thermal properties

ABSTRACT

An effective method for preparing tungsten carbide coating on diamond surfaces was proposed to improve the interface bonding between diamond and copper. The WC coating was formed on the diamond surfaces with a reaction medium of WO_3 in mixed molten NaCl-KCl salts and the copper–diamond composites were obtained by vacuum pressure infiltration of WC-coated diamond particles with pure copper. The microstructure of interface bonding between diamond and copper was discussed. Thermal conductivity and thermal expansion behavior of the obtained copper–diamond composites were investigated. Results indicated that the thermal conductivity of as-fabricated composite reached $658 \text{ W m}^{-1} \text{ K}^{-1}$. Significant reduction in coefficient of thermal expansion of the composite compared with that of pure copper was obtained.

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1. Introduction

Thermal considerations in power electronic devices have become extremely important with the continuing miniaturization and integration of microelectronics, requiring ever more efficient heat dissipation. The future thermal management materials for heat sink applications should have thermal conductivities above $300 \text{ W m}^{-1} \text{ K}^{-1}$ and tailorable CTE of $(4\text{--}9) \times 10^{-6} \text{ K}^{-1}$, such as microprocessors, LED, laser diodes or high power [1,2]. Metal matrix composites (MMCs) offer the possibility to tailor the properties of a metal by adding an appropriate reinforcement in order to meet the demand in thermal management. Traditional heat sink materials such as Mo/Cu, W/Cu or SiC/Al, however, despite featuring a low CTE, have thermal conductivities lower than $250 \text{ W m}^{-1} \text{ K}^{-1}$ [3,4].

Diamond has the highest thermal conductivity at room temperature of all known natural materials, and very low CTE, which makes diamond an ideal filling material in MMCs for heat sink materials [5–7]. Copper is widely used as a heat-spreading material since it exhibits high thermal conductivity ($400 \text{ W m}^{-1} \text{ K}^{-1}$). However, it presents a large CTE ($17.0 \times 10^{-6} \text{ K}^{-1}$) which is too high for heat sink applications. It is interesting to consider the use of diamond as fillers embedded in a thermal package substrate, such as a copper matrix. Therefore, in recent years, diamond reinforced copper matrix composites have received much attention, which are considered to be the next generation of heat sink materials for thermal management applications because of the excellent thermal conductivity and tailorable CTE [8–13].

Unfortunately, copper is naturally non-wetting with diamond due to chemical incompatibility, leading to weak interface bonding and high thermal resistance. Based on the previous study, alloying copper matrix with carbide formers such as Ti [9], Zr [14], Cr and B [8] offers a way to improve the interface bonding between diamond particles and copper matrix. However, the amount of additive elements is hard to control and most of the alloying elements will remain in the copper matrix, which deteriorates the thermal conductivity and interface bonding of the copper–diamond composites. In this work, an effective method is proposed to ameliorate the interface bonding between diamond and copper. In the proposed method, the WC coating is synthesized on the diamond particle surfaces using molten salts, and the coating can act as an intermediate layer to strengthen the interface between diamond and copper [15–20]. In addition, they can also protect the diamond particles from graphitization at high temperature.

Recently, a number of methods have been developed to fabricate copper–diamond composites such as hot pressing (HP) [14], spark plasma sintering (SPS) [10], and field assisted sintering technology (FAST) [21]. Another method used in fabrication of the copper–diamond composites is the high pressure–high temperature (HPHT) [22]. In this method, the authors used copper as a binder material. The highest reported value of thermal conductivity is $900 \text{ W m}^{-1} \text{ K}$ under extremely high pressure and high temperature of 8 GPa and 2100 K with 95 vol.% diamond particles.

The present study was carried out to investigate the influence of the WC coating on microstructure, thermal conductivity and CTE of copper–diamond composite. The WC coating was formed on diamond surfaces using the molten salt method. Subsequently, the copper–diamond composites were fabricated through pure copper infiltrating into the WC-

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coated diamond particles by vacuum pressure infiltration. The copper–diamond composites with uncoated diamond particles were also prepared as reference materials.

2. Experimental

2.1. Sample preparation

The infiltrated copper with purity more than 99.99% was used as matrix. Diamond particles used in this study were synthetic MBD8 grade diamond with nitrogen content of 200 ppm, purchased from Polaris Diamond Powder Co. Ltd., China. Thermal conductivity was estimated to be about $1500 \text{ W m}^{-1} \text{ K}^{-1}$ according to the level of nitrogen content [23]. The diamond particles were faceted with hexagonal or octahedral shapes and diameters around $70 \mu\text{m}$ (Fig. 1a). WO_3 powders of $\sim 45 \mu\text{m}$ with purity more than 99.5% were used as coating materials, and the SEM images are shown in Fig. 1b. The WC coating was synthesized on diamond particle surfaces using the molten salt method. Diamond particles and WO_3 powders were mixed uniformly with molar ratio of 10:1 and then embedded in an alumina crucible with a mixture of chloride salts (mol ratio of $\text{NaCl}:\text{KCl} = 1:1$), heat-treated at $900\text{--}1050 \text{ }^\circ\text{C}$ for 60 min in a tube furnace under high purity argon atmosphere and then cooled to room temperature. After treatment in molten salts, the processed powder was boiled repeatedly in distilled water with ultrasonic wave to dissolve chloride salts and then the coated diamond particles were dried in the vacuum drying oven at $120 \text{ }^\circ\text{C}$ for 60 min.

The diamond particle preforms used for the fabrication of the copper–diamond composites were made by a two-step process: that is, pressing the mixture of WC-coated diamond particles with 65% volume fraction and organic binder into a cylindrical mold of 30 mm in diameter with pressure; then degreasing the binder in high purity argon atmosphere at $400 \text{ }^\circ\text{C}$ for 2 h. The copper–diamond composites with the WC coating on diamond particles were fabricated by vacuum pressure infiltration. The diamond preform was positioned in graphite die with copper block on top, and then heated at $1150 \text{ }^\circ\text{C}$ for 10 min. After that, a pressure of 20 MPa was applied to infiltrate the molten copper into the diamond preform for 5 min. The heating was switched off and the system was cooled down to room temperature within the furnace.

2.2. Sample characterization

The phase constituents of the composites were studied using X-ray diffraction (XRD) in Siemens D5000. The microstructure and the composition of coated diamond particles were characterized by LEO1450 scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS). The bulk densities of the composites were measured using Archimedes' method and compared with the theoretical densities. Samples were machined using laser processing by a specialized processing factory for thermal property test. Thermal diffusivity of the sample (sample size: $\Phi 10 \text{ mm} \times 3 \text{ mm}$) was measured by Netzsch LFA 427 Laser Flash machine at room temperature, and the specific heat capacity

of Cu–diamond composite was calculated using the rule of mixture based on masses of each component. Thermal conductivity was specifically calculated as the product of thermal diffusivity, specific heat and density. The CTEs of composites were measured by a dilatometer (DIL 402 C NETZSCH, sample size: $3 \text{ mm} \times 4 \text{ mm} \times 25 \text{ mm}$) with a heating rate of $3 \text{ }^\circ\text{C}/\text{min}$ in the temperature range between 50 and $400 \text{ }^\circ\text{C}$, an argon atmosphere with a flowing rate of $50 \text{ ml}/\text{min}$ was used to keep the chamber temperature consistent, and the device was calibrated using an alumina sample. The uncertainty in the thermal measurements was $\pm 5\%$.

3. Results and discussion

3.1. Formation and microstructure of the WC coating

Fig. 2 displays the phase composition of the diamond particles after coating at $900\text{--}1050 \text{ }^\circ\text{C}$ for 60 min. As can be seen in Fig. 2D, only WC and diamond could be identified, and the lattice parameters of the α -WC coating (hexagonal P-6m2, $a = 2.906 \text{ \AA}$, $c = 2.837 \text{ \AA}$) were in good agreement with that of WC in the literatures [24,25]. However, as shown in Fig. 2(A,B,C), there were WO_3 , W and W_2C existing in the as-coated diamond particles, and these impurity phases could deteriorate the properties of the copper–diamond composites. Fig. 3 illustrates the microstructure of the coating on diamond particles which were coated at different temperatures. With temperature increasing, the white deposition is gradually and compactly coated on the surface of diamond particles, and the coated diamond particles maintain the original shape. In accordance with SEM combined with XRD analysis, WC coating is formed uniformly on the diamond particles at $1050 \text{ }^\circ\text{C}$ for 60 min. Compared with the TiC coating [6] and Mo_2C coating on diamond particles [19], the WC coating on diamond particles is more rough, which may be due to the complicated chemical reaction between WO_3 and diamond particles in molten salts.

The dissolution of WO_3 in molten salts is not well understood. However, the molten salt mixture is expected to facilitate the dissolution and transport of the WO_3 to the diamond surfaces to form WC coating. Prior research showed that the reaction occurred easily in molten salts [26]. Compared with the molten salt method, the conventional preparation methods for WC have been inherited from the metallurgical industry involving heat treatment of a mixture of metallic tungsten powder and carbon black in a furnace under an atmosphere of flowing hydrogen at high temperatures ranging from $1400 \text{ }^\circ\text{C}$ to $1600 \text{ }^\circ\text{C}$ [27].

3.2. Density and microstructure

The relative densities of diamond–copper composites made from two kinds of diamond particles by vacuum pressure infiltration are shown in Fig. 4. The change of relative densities of prepared composites in Fig. 4 shows that surface pretreatment of the diamond particles by WC coating raises the densities of the copper–diamond composites from 97.5% to 99.5%.

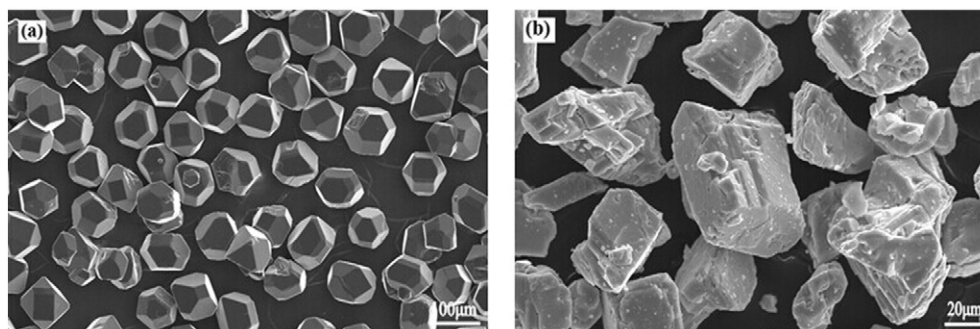


Fig. 1. SEM images of (a) the diamond particles and (b) the WO_3 powder.

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