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Microstructure and thermal properties of Al/W-coated diamond composites prepared by powder metallurgy

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ABSTRACT

Al/(35–60) vol.% diamond composites are prepared by hot pressing, and the microstructures, thermal conductivity, and coefficient of thermal expansion (CTE) are characterized considering the tungsten coating and the volume fraction of diamond. The results show that diamond particles distribute quite uniform in Al matrix without formation of Al_4C_3 phase at the interface. Although the tungsten coating effectively improves the interface bonding between the diamond and matrix, relative densities decrease with increasing diamond fraction. The thermal conductivity of W-coated specimens containing 35–50 vol.% of diamond are about 90% of the theoretical values, but it decreases with further increasing diamond fraction due to the existence of higher porosity. However, most of the thermal conductivity of uncoated specimens are less than 90% of the calculated ones. CTE of the composites is found to decrease as the diamond fraction increases, and the W-coated specimens are more effectively to suppress the thermal expansion of Al matrix.

1. Introduction

In the recent years, the continuous progress of microelectronic systems with high calculating speed and more compact size leads to the rapid increase of functional density [1]. Therefore, efficient thermal management is demanded in microelectronics to prevent overheating induced performance degradation or even device failures [2,3]. Thermal management materials with high thermal conductivity have been of significance in the electronic industry. Additionally, compatible coefficient of thermal expansion (CTE) matching with those of semiconductor materials or ceramic substrates is also demanded in terms of reliability of the electronic components [4–6]. Al/diamond composites have received increasing interest in the application of electronic packaging for thermal management owing to the combination of high thermal conductivity, low CTE, and low density [7]. However, poor wettability between the diamond and metal matrix and complex fabrication process limit the wide application of metal/diamond composites.

In order to improve the interfacial wettability, both diamond particle surface modification using W, Ti, Cr, Mo et al. [8–10] coating and matrix alloying with Ti, Cr et al. [11,12] are employed. Among these modification and alloying elements, easy carbide formable elements are selected to form carbides at the interface between the diamond and matrix, and then improves the thermal properties of composites. It is thought to be more effectively by surface modification due to the integrity of metal coating and free from selective interfacial bonding and deterioration of matrix property [13,14]. However, too much carbides formed at the interface will reduce such effect because the carbides are poor thermal conductor and also show poor wettability with metal matrix. Accordingly, tungsten has been widely used as surface modification element due to its high melting temperature and relatively high thermal conductivity [10,15].

Al/diamond composites have been prepared by various techniques, such as powder metallurgy (PM) [16], pressure or pressureless infiltration [17,18], squeeze casting [7], and high-temperature high-pressure method [19] et al. Among these techniques, the most effective and relative simple route may be the PM technique which usually involves cold pressing of mixed powder following pressure or pressureless sintering. Moreover, the PM technique is valuable to add reinforcement with a wide range of volume fraction. In this respect, hot pressing method provides some opportunities because it involves comparatively high applied pressure, low sintering temperature (500–650 °C, depending on the matrix alloy composition), near full density, and high productivity [20]. Furthermore, near net sharp products can also be obtained through proper die design.

In this work, Al/diamond composites containing uncoated and W-coated diamond particles with volume fraction of 35–60% were fabricated by hot pressing. This work aims at providing a contribution to clarify the effect of surface modification and fraction on the microstructures and thermal properties of Al/diamond composites. Therefore, the

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effect of diamond fraction on the microstructure characteristics, thermal conductivity, and CTE of the composites were evaluated. Moreover, comparisons were made between the specimens with uncoated and W-coated diamond particles.

2. Experimental procedure

2.1. Composite preparation

A commercial gas-atomized aluminum powder (99.95%) with an average size of 74 μm was used as the composite matrix. As the filling material, high-quality MBD6-type synthetic diamond particles provided by Henan Huanghe Whirlwind Co., Ltd. of China with plane-faced cubic-octahedral monocrystalline were used. The nominal particle size was 400 μm. Tungsten single layer coating was applied to the diamond particles by the diffusion method which was described in detail in the previous work [21]. The thickness of tungsten coating layer was about 420 nm which was measured gravimetrically by determining the increase in the diamond powder mass after applying the coating.

The aluminum powder with different volume fractions (35–60 vol. %) of diamond particles were mixed mechanically at room temperature. For comparison, both the uncoated and W-coated diamond particles were used. The powder mixtures were subjected to double action axial compaction in a steel die under 300 MPa for 2 min. The relative densities of the green compacts are about 82%. Then the compacts were sintered by hot pressing in a graphite die. A sheet of graphitic paper was placed between the punch and the compacts as well as between the die and the compacts for easy removal. The furnace was heated to 400 °C and held for 30 min at a pressure of 20 MPa in order to degas the powder compacts. The maximum uniaxial pressure was 45 MPa. The selected sintering temperature and holding time were 640 °C and 60 min, respectively. The heating rate was 15 °C/min. The temperature during hot pressing was monitored by a thermocouple inserted to the die. The indicated temperature fluctuated by ±2 °C around the set value. The pressure on the specimens was not released until cooled down to 200 °C. After sintering, the hot pressed specimens were cooled in furnace to the room temperature.

2.2. Material characterization

Specimens used for microstructural observations were chemically etched with a hydrochloric acid solution at room temperature for about 10 s to dissolve part of the diamond particles from the matrix. Three-point bending fracture surfaces were also used to investigate the microstructures of the composites using a scanning electron microscope (SEM, FEI QUANTA-200). Interface atomic composition was analyzed by energy dispersive X-ray spectrometry (EDS) attached to the SEM, and X-ray diffraction performed on a Rigaku D/Max2500VB+ diffractometer using Cu Kα radiation at a scan step of 0.08 (°)/s.

The densities of the composites were measured by the Archimedes method with a very thin film of vaseline on each specimen to avoid the permeation of water. The theoretical density of pure aluminum and diamond were used to calculate the relative densities of the specimens in terms of rule of mixture (ROM). Three-point bending strength was measured on the specimens with a dimension of 3 × 10 × 50 mm. The bending tests were carried out with an initial strain rate of $10^{-4}\,s^{-1}$ using Instron MTS 850 materials testing system. Thermal diffusivity of the composites at room temperature were measured by laser flash method on a NETZSCH LFA427/3/G thermal physical testing instrument. The specific heat was derived from the theoretical value calculated by ROM. The thermal conductivity of the composites was then calculated as a product of the density, thermal diffusivity, and specific heat. CTE measurements were carried out on a NETZSCH DIL 402C dilatometer from 50 °C to 400 °C with a heating and cooling rate of 5 °C/min. Three or more parallel tests were conducted to ensure good reproducibility of the data.

3. Results and discussion

3.1. Microstructures

The microstructures of tungsten coated diamond particles are shown in Fig. 1. According to Fig. 1, the tungsten layer is significantly homogeneous and integrated, and the magnified image indicates that the layer is made of submicron particles. The X-ray diffractogram of the diamond with single tungsten coating is presented in Fig. 2. It can be analyzed that the applied coating on synthetic diamonds consists of metal tungsten and tungsten monocarbide.

Fig. 3 shows the cross-sectional microstructures of the hot pressed Al/diamond composites with various volume fractions of diamond after light etching. It can be seen that the distribution of diamond particles in Al matrix is quite homogeneous, and most edges and corners of diamond particles remains. No significant pores exist at the interface between the aluminum and diamond particle in Fig. 3a and b or composites containing up to 45 vol.% diamond. The use of sintering temperature close to the melting point of aluminum with the assistance of applied pressure apparently densified the composites to relative high density as compares with the ones containing high diamond fractions. Increasing pores can be observed in the specimens containing higher than 50 vol.% diamond or Fig. 3c. With increasing the diamond fraction, the bonding phase or aluminum decreases and becomes harder to infuse among the large number of stacked diamond particles, resulting in much higher percentage of pores. Furthermore, the pores are more visible in the composites with uncoated diamond particles. Large pores are formed at the interfaces in Fig. 3e and f. These pores apparently will affect the density, thermal properties, and mechanical properties of the composites.

Fig. 4 shows typical interface SEM micrographs of the Al/45 vol.% diamond composites in which the diamond particles are uncoated and W-coated, respectively. As seen in Fig. 4a, there exists obvious gap at the interfacial boundary between the diamond and Al matrix for the composite containing uncoated diamond particles. Poor interfacial wettability could cause such gap. In terms of the composite with W-coated diamond particles, the adhesion of the Al matrix to the diamond is significantly improved, and the matrix is found to adhere on almost all the diamond particle surfaces, as shown in Fig. 4b. This result suggests that a tungsten coating is effective to improve the wettability between the diamond and Al matrix. Owing to the W-coating layer formed carbides, it can directly form chemical bond between diamond and matrix during the sintering process. The chemical interfacial bonding can make the interface between diamond and Al matrix keep adhesion during the sintering process, otherwise, the debonding may happen because of the thermal expansion difference between diamond and Al matrix.

Monje et al. [22] and Khalid et al. [23] reported that aluminum and diamond interface may form needle-like Al_4C_3 phase at aluminum-diamond interface via liquid phase infiltration. The Al_4C_3 phase is brittle and tends to absorb moisture which degrades both the thermal conductivity and mechanical properties of composites. In this work, Al_4C_3 phase is not detected even in the specimens containing uncoated diamond particles. It is demonstrated that surface damages of diamond particles by direct contact with molten aluminum will reduce the materials properties because of the degradation of diamond and surface roughness [13,24]. The hot pressing temperature of 640 °C is lower than the melting point of aluminum (660 °C). Thus, the formation of Al_4C_3 can be avoided. Additionally, no graphite-like carbon phase is detected, which should come from the fact that the graphite cannot be formed or the content of graphite is too limited to be found. Although diamond is a metastable allotropic modification of carbon, the diamond particles are not graphitized to an apparent extent after sintering at 877–947 °C in the Cu/diamond composite [25]. The absence of the Al_4C_3 phase and graphitization is confirmed by XRD analysis in Fig. 5.