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Enhanced thermal conductivity in Diamond/Aluminum composites with tungsten coatings on diamond particles prepared by magnetron sputtering method

Wenshu Yang [a, *], Guoqin Chen [a, **], Pingping Wang [a], Jing Qiao [a], Fengjiao Hu [b], Shufeng Liu [c], Qiang Zhang [a], Murid Hussain [d], Ronghua Dong [e], Gaohui Wu [a]

[a] *Department of Material Science and Engineering, Harbin Institute of Technology, Harbin 150001, China*
[b] *Xi'an Division of China Academy of Space Technology, Xi'an 710000, China*
[c] *Beijing Institute of Electronic System Engineering, Beijing 100854, China*
[d] *Department of Chemical Engineering, COMSATS Institute of Information Technology, M.A. Jinnah Building, Defence Road, Off Raiwind Road, Lahore 54000, Pakistan*
[e] *Beijing Institute of Aerospace Control Devices, Beijing 100039, China*

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ABSTRACT

In the present work, tungsten (W) coatings with thickness range of 35–130 nm on the diamond particles were prepared by magnetron sputtering method, and then the Diamond/Al composites were prepared by the vacuum infiltration method. The prepared W coatings were smooth and dense on all the facets of the diamond particles. Moreover, the presence of W-coatings inhibited the interfacial debonding phenomenon and improved the interfacial bonding between diamond particles and Al matrix. The Diamond/Al composite with the 45 nm W-coating achieved the maximum thermal conductivity (622 W/(m·K)). To the best of our knowledge, it is the highest thermal conductivity obtained in the Al matrix composites reinforced with 100 μm diamond particles with coatings. Based on the Hasselman and Johnson (H-J) model, the thermal conductivity behavior of the Diamond/Al composites has been discussed. It indicates that the magnetron sputtering is a feasible and successful method to prepare thin and reliable tungsten coatings for the Diamond/Al composites.

1. Introduction

Diamond reinforced Al matrix (Diamond/Al) composites have been considered as the next generation thermal management materials due to their high thermal conductivity (TC) and tailorable coefficient of thermal expansion (CTE) [1]. The priority issue for the Diamond/Al composites is to optimize the interfacial bonding to minimize the interfacial thermal resistance (ITR) and enhance the ways of heat transferring [2]. Extensive and in-depth researches have been explored to optimize the preparation methods and parameters to improve the interfacial bonding. Several methods, such as pressureless metal infiltration [3,4], powder metallurgy (PM) [5,6], spark plasma sintering [7–9], vacuum hot pressing [10], gas pressure infiltration [11–13] and pressure infiltration method (squeeze casting) [14–16], have been adopted to prepare the Diamond/Al composites, and significant improvement in the TC has been achieved due to these afore-stated technologies. The main design of the above researches is to form limited Al_4C_3 at the diamond-Al interface [11–15]. However, the degradation of Al_4C_3 would be detrimental for the service performance of the Diamond/Al composites [17]. Meanwhile, Al_4C_3 is not the desired interfacial phase for the TC performance due to its relative low TC value [18].

Recently, other interfacial phases have been explored in the Diamond/Al composites by the addition of alloying elements and diamond coating treatments. It has been found that the addition of Si element into the Al matrix could inhibit the formation of Al_4C_3 and improve the TC of the composites [19–21]. Wu et al. [22] found

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* Corresponding author. P. O. 3023, Science park, No. 2 Yikuang street, School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150080, China.

** Corresponding author. P. O. 3023, Science park, No. 2 Yikuang street, School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150080, China.

E-mail addresses: yws001003@163.com (W. Yang), chenguoqin@hit.edu.cn (G. Chen).

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that addition of Cu element into Al matrix led to the formation of Al_2Cu phase at the interface, which promoted the diamond-Al interfacial bonding and then increased the TC from 210 to 330 W/(m·K). Xue et al. [23] found that the interfacial bonding and the TC of the composites were increased with the Ti content in Al matrix. However, the effect of alloying elements could not be easily controlled [24], and appeared to be less effective than the effect of coating treatment [23]. The main advantage of coating treatment is the effectiveness in the controlling of the coating materials and the corresponding thickness [25]. The Ti [23,26–29], TiC [30], WC [31] and W [6,32–35] coatings have been used for the diamond particles. Ti coating could significantly improve the diamond-Al interfacial bonding, and the highest TC values of 491 [23,28] and 650 W/(m·K) [29] have been obtained for the Ti-coated Diamond/Al composites reinforced with about 100 μm and 150 μm diamond particles, respectively. Liu et al. [30] found that the TiC coating could improve the bending strength and TC value of the Diamond/Al composites, and the highest TC value of 382 W/(m·K) was obtained. Liang et al. [36] and Tan et al. [37] investigated the effect of type and thickness of coating materials on the TC value of the Diamond/Al composites, and found that the Ti, TiC as well as $TiAl_3$ layers would deteriorate the TC performance of the Diamond/Al composites [37]. Moreover, Tan et al. [37] also concluded that among all the metals used as interfacial layers, the W, Mo and WC, which has relative low solubility in Al matrix, are the most promising candidates to improve the thermal property of the Diamond/Al composites. Recently, Tan et al. [33] and Ji et al. [34] used sol–gel process to prepare W-coatings on the diamond particles, and the maximum TC value of 599 W/(m·K) has been obtained. Zhang et al. [6,32] prepared W-coating by diffusion method and obtained maximum TC value of 474 W/(m·K) [6]. However, the homogeneity of the W-coatings should be further improved to fully separate the contact between the diamond and Al matrix. Moreover, the thickness of the coatings should be thinner since the composites with thicker coatings demonstrate lower TC value [36,37]. Yang et al. [31] prepared 300 nm W-coating by the magnetron sputtering method, which was then heat treated to form WC to improve the interfacial bonding, and the maximum TC value was 588 W/(m·K) [31]. However, Yang et al. [31] did not measure the TC value of the composites with W-coatings. Moreover, the thickness of the W coating was thicker than the expectation.

Although magnetron sputtering is an effective method to prepare uniform and thin coatings, the effect of thin W-coating prepared by the magnetron sputtering method for the Diamond/Al composites has not been explored yet. Therefore, in the present work, the W coatings with the thickness range of 35–130 nm on the diamond particles were prepared by the magnetron sputtering method, and the effect of W-coatings on the microstructure and thermal conductivity properties of the Diamond/Al composites have been investigated. To the best of our knowledge, it is the highest thermal conductivity obtained in the Al matrix composites reinforced with 100 μm diamond particles with coatings. The highest TC value for the Al matrix composites reinforced with 100 μm diamond particles with coatings has been obtained (622 W/(m·K)) in the present work. This indicates that the magnetron sputtering is a feasible and successful method to prepare thin and reliable tungsten coatings for the Diamond/Al composites.

2. Materials and methods

Diamond particles with mean size of 100 μm (MBD4, supplied by Henan Famous Diamond Industries) and commercial purity aluminum (1060, 99.6 wt.% in purity, supplied by Northeast Light Alloy Co., Ltd. China) have been used as raw materials in the present study. The chemical composition (wt.%) of the 1060Al alloy was 0.124% Si, 0.031% Mg, 0.0415%Cu, 0.257%Fe, 0.033%Mn, 0.041%Zn, 0.02%Ti, and Al balance. Magnetron sputtering was performed on MSP-5100B system (Beijing Chuangshiweina Technology Co., Ltd. China). During the deposition process, a constant Ar flow was kept leading to a process pressure $<8 \times 10^{-3}$ Pa, while the diamond substrate was heated to 300 °C and holding for 30 min. The current and voltage for the magnetron sputtering process were 0.9 A and 600 V, respectively. A pure W target (99.99%) with the dimension of Φ100 mm × 50 mm was used in the present work. Continuous particle tumbling technique was adopted during the magnetron sputtering process to expose all sides of the diamond particles to the sputtered flux to prepare uniform W-coatings on the three dimensions of diamond particles. For each magnetron sputtering process, 50 g diamond particles were put into a petri dish which was vibrated 30 times/min by the ultrasonic system (30 MHz). Due to the vibration, the diamond particles were tumbled during the sputtering process. The diamond particles were magnetron sputtered 90, 180, 270 and 360 min to obtain coatings with different thickness values. The schematic images of the location and assembly of the diamond preforms and Al alloys have been shown in Fig. 1. In order to eliminate the effect of preparation parameters, diamond preform without W-coating and with various W-coatings were prepared in the same graphite mold (Fig. 1a). Due to the accumulation of diamond particles, the set volume amounts of diamond particles in the preforms were about 55 vol.%. Afterwards, the graphite mold with a porous graphite cover was put into a steel mold, and the 1060 Al alloy (about 3 Kg) and graphite indenter were put subsequently on the porous graphite cover (Fig. 1b). The steel mold with diamond preforms and Al alloy was then heated in the vacuum furnace (ZYD-200-200LC, Jinzhou Hangxing Vacuum Equipment, Co. Ltd, China) to 880 °C with heating rate of 200 °C/h and holding for 1 min under vacuum status ($<1 \times 10^{-3}$ Pa). After that, 10 MPa pressure was applied on the graphite indenter to infiltrate the molten Al into the diamond preforms. The composites were then cooled within the furnace to the room temperature. For comparison, the Diamond/Al composite without W-coating was also prepared.

Microstructure of the diamond particles and the Diamond/Al composites were observed by FEI Sirion Quanta 200 scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDS). Before the SEM observation, the Diamond/Al composite samples were by polished Ar ion beam (IB-09020CP, JEOL) with acceleration voltage of 6 kV and gun current of 160 μA. Atomic force microscopy (AFM) was performed on Dimension Icon (Bruker Corporation) to measure the thickness of the W-coatings. Before AFM observation, the W-coatings on the diamond particles were broken by ball-millings. X-ray diffraction (XRD) analysis was carried out on Rigaku D/max-rB diffractometer. The specimens were subjected to Cu-Kα radiation (0.15418 nm) with a scanning speed of 2°/min. 2θ scans were performed between 15 and 90°. Differential scanning calorimetry (DSC) was conducted on Netzsch DSC Pegasus 404F3 (NETZSCH GmbH, Selb, Germany) from room temperature to 1200 °C with the heating rate of 10 °C/min under dynamic nitrogen atmosphere (purity 99.999%, 30 ml/min). During the measurement, an alumina pan was used to load the diamond particles with W-coating (about 5 mg), and another empty alumina pan was used for reference. The baseline was calibrated at the heating rate of 10 °C/min using two empty alumina pans before measurement. The thermal diffusivities (k) of the samples with size of Φ12.7 mm × 3 mm was measured on an LFA 447 Nanoflash (NETZSCH GmbH, Selb, Germany) at room temperature. Five samples of all the composites have been measured to improve the statistical significance of the results. To be more accurate, all the thermal diffusivity tests were performed in the central area of the samples. The TC (λ) was calculated by the following equation: