



# Overcoming selective interfacial bonding and enhancing thermal conductivity of diamond/aluminum composite by an ion bombardment pretreatment



Wulin Yang<sup>a,b,\*</sup>, Jianquan Sang<sup>a</sup>, Lingping Zhou<sup>a,b</sup>, Kun Peng<sup>a</sup>, Jiajun Zhu<sup>a</sup>, Deyi Li<sup>a</sup>

<sup>a</sup> College of Materials Science and Engineering, Hunan University, Changsha 410082, China

<sup>b</sup> Hunan Province Key Laboratory for Spray Deposition Technology and Application, Hunan University, Changsha 410082, China

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## ABSTRACT

Low energy Ar<sup>+</sup> ion beam bombardment is adopted as a pretreatment process of diamond particles for fabricating diamond/aluminum composite. We demonstrate that the combined effects of higher defect density and concentration of oxygen functional groups on diamond {100} plane lead to a selective interfacial bonding in the raw diamond/aluminum composites. It limits the thermal transfer performance of the composite. With the help of ion implantation and cleaning effects, similar diamond surface states are obtained by a conversion of sp<sup>3</sup> C to sp<sup>2</sup> C which occurs on both the diamond {100} and {111} planes simultaneously. The similar surface states are beneficial for the aluminum matrix adhered to the bombarded diamond surface uniformly during the interfacial reaction. Furthermore, the mean interfacial thermal conductance between the bombarded diamond particles and the matrix reaches up to  $9.3 \times 10^7 \text{ W}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$ . The thermal conductivity of the diamond/aluminum composite is improved significantly to  $713 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ , indicating that low energy ion beam bombarded is a promising approach for further improving the interfacial bonding and the thermal conductivity of the diamond/aluminum composite.

## 1. Introduction

The rapidly increasing power density and heat dissipation of high-power integrated circuits cause serious issues in efficient heat removal for maintaining the performance and reliability of modern electronic devices [1]. To solve this problem, metal matrix composites with excellent thermal conductivity and adjustable thermal expansion have been considered to be the most promising thermal management material in the electronic packaging field. Diamond is well known for its superior thermal conductivity (TC) in nature, about  $2000 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ , and an appropriate coefficient of thermal expansion ( $2.3 \times 10^{-6} \text{ K}^{-1}$ ). Many previous studies have pointed out that diamond is an ideal filling material for metal matrix composites that used as heat spreader or heat sink [2–4].

In recent years, diamond reinforced metal matrix composites are extensively investigated. Due to the bad wettability of the diamond particles and high interfacial thermal resistance, the thermal conductivity of the composite is far from the calculated in theory. The moderate interfacial reaction is conducive to reducing interfacial thermal resistance of the composites, so surface coating of diamond or adding active elements with higher affinity to carbon into the metal

matrix has been adopted to overcome the problem by forming a buffer layer between the diamond particles and the matrix [5,6]. The thermal conductivity of the composite has been improved, but there is still a large gap comparing with the theoretical value.

Interfacial reaction rates of different orientation planes of diamond monocrystal are significantly different under the same reaction condition. J. Wang [7] and W. Smirnov [8] etc. found that the {100} plane was etched more rapidly in comparison to the {111} plane when synthetic diamond monocrystal was etched by metal powder. Actually, the similar researches have also found selective interfacial bonding behavior in diamond reinforced pure Al or Cu-alloy matrix composites [9,10]. The interface is a bridge of heat transmission between the diamond and the matrix, so it is reasonable to deduce that the selective interfacial bonding will lead to the anisotropy of the interfacial thermal conductance. It has demonstrated that the very different interfacial configurations are still obvious at different orientation diamond/aluminum interfaces even at the optimized preparation process [11]. That is to say, it is difficult to average the interface properties by only optimizing the preparation process. As far as we know, the carbide layer formed by coating diamond particles [12–14] or metal matrix alloying [2,5,15] has been also employed to solve the problem. Although these

\* Corresponding author at: College of Materials Science and Engineering, Hunan University, Changsha 410082, China.  
E-mail address: [hnywl@hnu.edu.cn](mailto:hnywl@hnu.edu.cn) (W. Yang).

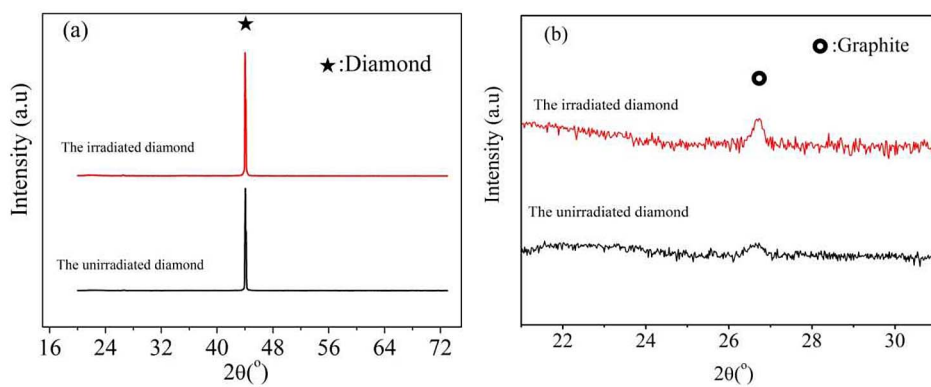


Fig. 1. (a) XRD patterns of the raw and the  $\text{Ar}^+$  ion bombarded diamond particles (b) enlarged view of the XRD patterns.

methods show a remarkable effect on improving the interfacial bonding, they would decrease the interfacial thermal conductance or the thermal conductivity of the matrix. How to avoid the selective interfacial bonding and further increase the thermal conductivity of the composite simultaneously should be still given more attentions.

The selective interfacial bonding of diamond/metal composite is closely related to surface termination, surface energy and atomic arrangement of different orientation diamond planes. It has been reported that the {111} plane is the close-packed plane of diamond with a surface energy of  $5.4 \text{ J}\cdot\text{m}^{-2}$ , which is lower than that of {100} plane ( $9.4 \text{ J}\cdot\text{m}^{-2}$ ) [16]. So the {100} plane has higher reactivity than the {111} plane in the process of interfacial reaction. From the view of the surface atom arrangement, a qualitatively explained is given. For the diamond {111} plane, surface carbon atoms are bonded threefold to the bulk by three carbon-carbon bonds, while carbon atoms on the diamond {100} plane are only bonded twofold to the bulk by two carbon-carbon bonds. Hence, comparing with the diamond {111} plane, the critical energy barrier of dissolution a carbon atom from the {100} plane is lower, and the selective interfacial bonding is remarkable [9].

In this paper, ion beam bombardment technology is used to modify diamond particles for further optimizing interfacial reaction and thermal conductivity of the diamond/aluminum composites. Regulating the surface state and chemical bonding of diamond particles by ion bombardment and its cleaning effects can solve the selective interfacial bonding problem. Meanwhile, the interfacial thermal resistance of different orientation diamond planes will be aligned and optimized. Combining the two aspects, the all interfacial thermal transmission bridges in the composite are built up, resulting in enhancing the thermal conductivity of the composites.

## 2. Experimental

HWD40 type diamond with an average particle diameter about  $200 \mu\text{m}$  was purchased from Henan Huanghe Whirlwind Co., Ltd. The nitrogen concentration of the diamond was about 200 ppm, thus the intrinsic thermal conductivity of the diamond was estimated to be  $1500 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  from the linear relationship between the thermal conductivity and the nitrogen content [17]. Commercial purity aluminum (99.99% in purity) was used as the matrix material, and the thermal conductivity of the matrix was considered to be  $237 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ .

The  $\text{Ar}^+$  ion beam bombardment was performed in a homemade vacuum chamber system equipped with an 8 cm Kaufman ion source. The Base pressure of the vacuum system was better than  $5 \times 10^{-4} \text{ Pa}$ , and typical working pressure of the ion source was  $1 \times 10^{-2} \text{ Pa}$ . During the ion beam bombardment the diamond particles were continually moved by vibration and translation motions in the same plane. The grid voltages and total ion beam current of the bombardment were 0.6–1.2 kV and 50 mA, respectively. The bombardment lasted about 1 h for every 5 g diamond particles.

After the ion bombardment, the diamond particles reinforced aluminum composites were fabricated by a vacuum pressure infiltration method. Specifically, the vacuum of the furnace is less than 1 Pa during the infiltration process, and the diamond particles and bulk aluminum were simultaneously heated up to the infiltration temperature ( $750 \text{ }^\circ\text{C}$ ) with rate  $10 \text{ K}/\text{min}$ , the temperature maintained for 20 min until the infiltration process completed. After that, the specimen was furnace-cooled to room temperature. The volume fraction of the diamond in the composites was approximately 62%.

The samples for interfacial characterization were prepared by an electrochemical etching method [18]. Absolute ethyl alcohol solution of perchloric acid (10 vol%) was used as an electrolyte, etching voltage and current density were 19 V DC and  $1 \text{ A}\cdot\text{cm}^{-2}$  respectively. Morphologies of the fracture samples and the interfacial characterization were recorded by a scanning electron microscope (SEM, FEI Quanta 200) at 20 kV. X-ray diffraction (XRD) patterns were recorded by a D5000 Siemens diffractometer, with voltage 40 kV, current 30 mA, XPS surface chemical analyses of different diamond planes were performed by K-Alpha spectrometer using a monochromatic  $\text{Al K}\alpha$  X-ray Source (1486.6 eV). The binding energy scale was calibrated according to the Au 84.0 eV.

The thermal conductivity (TC) of the composite was calculated by the equation  $\lambda = \alpha\rho c_p$ , where  $\lambda$  is the thermal conductivity,  $\alpha$  is thermal diffusivity,  $\rho$  is sample density and  $c_p$  is specific heat capacity. The thermal diffusivity was measured by a laser flash technique using a Flashline™ 3000 thermophysical testing machine at room temperature. The sample density was measured by the Archimedes method. The specific heat capacity was calculated by means of the rule of mixture.

## 3. Results and discussion

The effect of the ion beam bombardment pretreatment on the phase composition of the diamond particles is presented in Fig. 1. The grid voltage of the bombardment is 1 kV. According to a trace of graphite (002) diffraction peak in the bombarded diamond sample, it can be inferred that the diamond surface partially transform into a graphite structure.

A conversion of  $\text{sp}^3$  to  $\text{sp}^2$  structure [19] on the ion bombarded diamond surface should be the main reason for the partial graphitization of the diamond surface. In addition to the conversion, defect structure also occurs with the displacement of carbon atoms or the breaking of bonds [20]. Whether the above mentioned changes evenly take place on both the diamond {100} and {111} planes in our pretreatment is the key issue to regulate the selective interfacial bonding. Thus, the  $\text{sp}^3$  to  $\text{sp}^2$  conversion induced by the  $\text{Ar}^+$  ion bombardment on both the diamond {100} and {111} planes deserve even greater attention. To quantify this conversion, X-ray photoelectron spectroscopy was employed to monitor the concentration of  $\text{sp}^3$  and  $\text{sp}^2$  carbon atoms on each plane before and after the bombardment. The C 1s XPS spectrums of different orientation diamond planes are shown in Fig. 2.

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