

Enhanced thermal conductivity in diamond/aluminum composites with a tungsten interface nanolayer

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ABSTRACT

A tungsten (W) nanolayer was first introduced onto diamond particles by a sol–gel process, and then aluminum (Al) based composites were fabricated by vacuum hot pressing using the W coated diamond (diamond@W) particles. The microstructure of the W nanolayer and its effect on the thermal properties were explored. The results showed that the W nanolayer with a dendritic morphology and a thickness of 200 nm is the optimum combination to improve the interfacial bonding and minimize the thermal boundary resistance between diamond and Al. Such an observation was explained by the tunable formation of trace amount of W₂C. The thermal conductivity of 50 vol.% diamond@W/Al composites was 599 W/mK, 21% higher than that of the composite without the W interface nanolayer. Our results were found to be in good agreement with the theoretical predictions by the combined differential effective medium (DEM) and acoustic mismatch model (AMM) schemes.

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1. Introduction

Owing to their high thermal conductivity (TC) and tailorable coefficient of thermal expansion (CTE), diamond/Al composites have emerged as the next generation thermal management materials, and have drawn more and more attentions from both scientific and industrial communities [1–8]. These composites are usually prepared by either powder metallurgy (PM) [1–3] or liquid infiltration [5–8]. Although more than twice the value of the currently used SiC/Al or W/Cu composites, TC of the as-fabricated diamond/Al composites is still much lower than expected. This can be attributed to the additional thermal boundary resistance introduced by the poor interfacial bonding in the case of the PM composites [2,3], and by the aluminum carbide (Al₄C₃) interface layer in the case of the liquid infiltrated composites [9]. In particular, the Al₄C₃ interface layer contributes to better interfacial bonding to some extent, but it also degrades the TC of the composites, since the TC of Al₄C₃ is far below those of diamond and Al. Therefore, an interface layer with higher TC than Al₄C₃ and good interfacial bonding with both diamond and Al should be sought in the design and fabrication of diamond/Al composites.

As for diamond/Cu composites, either alloying of the Cu matrix [10,11] or surface metallization of diamond [12–16] were reported

to be helpful to improve the interfacial bonding and enhance the TC. However, alloying will cause a large TC degradation of the metal matrix, and thus it is unfavorable to develop the full potential of diamond and metals. For example, the TC of pure Cu was up to 390 W/mK, while those of Cu–0.8Zr and Cu–0.1B alloys were 335 and 355 W/mK [10,11], respectively. In comparison, surface metallization of diamond particles with carbide-forming metals, such as Ti [12], Cr [13,14], W [15] and Mo [16], has been proved to be an effective approach to improve the interfacial bonding and decrease the thermal boundary resistance in diamond/Cu composites. This is because, there is no reaction at all between Cu and diamond, while the coating metal can react with diamond to form a carbide interface layer that bonds well with the Cu matrix. In diamond/Al composites, although the Al matrix itself can easily react with diamond and form an Al₄C₃ interface layer, it has been shown that surface metallization can still help improve the TC of the diamond/Al composites. For example, a 0.5 μm-thick Ti coating on diamond resulted in a TC improvement from 325 W/mK to 491 W/mK for 50 vol.% diamond/Al composites [3]. This is, however, still much lower than the value predicted by analytical modeling, as a result of the additional thermal boundary resistance introduced, which is proportional to the thermal resistance and the thickness of the interface layer. Thus, to realize the full potential of surface metallization in diamond/Al composites, proper coating metal must be utilized, which should: (i) have high TC itself and very limited solid solubility in the Al matrix; and (ii) react with diamond but the

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kinetics is slow. The objective is to achieve a nanoscale interface layer that has good bonding with both diamond and the Al matrix. This layer should be as thin as possible, and composed of a high TC metal part on the side of Al, and a metal carbide part on the side of diamond, to reduce the thermal boundary resistance. Among the commonly used metals for surface metallization, Ti [12], Cr [13,14], W [15], and Mo [16], W has the highest TC. In addition, it also has the advantage of more controllable carbide formation, because it can only react with diamond to form carbides at elevated temperatures. However, probably due to the large density, W nanolayer has not been reported in the fabrication of diamond/Al composites yet.

This work aims to explore the potential of the W nanolayer on promoting the interfacial bonding and minimizing the thermal boundary resistance between diamond and Al. In particular, the W nanolayer with a thickness of 100–400 nm on diamond particles was first prepared by a sol–gel method, and thus the W coated diamond (diamond@W) particles were obtained, after which the diamond@W/Al composites was fabricated by vacuum hot pressing (VHP). The results revealed a ~21% TC enhancement in Al-based composites with 50 vol.% diamond by using diamond@W particles, which has been attributed to the improved interfacial bonding and the minimized thermal boundary resistance. These results were found to be in good agreement with the theoretical evaluations by the combined differential effective medium (DEM) and acoustic mismatch model (AMM) schemes.

2. Experimental procedures

2.1. Preparation of diamond@W particles

The diamond@W particles were fabricated by a sol–gel method, as schematically illustrated in Fig. 1. The preparation of the W sol–gel has been described in detail in Refs. [17–20]. Specifically, 4 g W powders of 99.9% purity were slowly dissolved into 20 ml of 30% H_2O_2 , which were stirred continuously. Being exothermic, the reaction between the W powders and H_2O_2 was kept at 5–15 °C by a cold-water bath during the whole process. When the reaction was over, the solution became milk-like. Afterwards, a platinum sheet was dipped into the solution to reduce the excess H_2O_2 . Then after the addition of 15 ml ethanol and 4 ml glacial acetic acid, the solution was refluxed at 55 °C for 12 h until a stable sol–gel of per-

oxotungstic acid ($WO_3 \cdot nH_2O$) was finally obtained, with a nominal W concentration of 0.1 g/ml. By adding alcohol, this sol–gel could be further diluted to a lower concentration. In this study, 0.01 g/ml, 0.02 g/ml and 0.1 g/ml sol–gels were used to produce W nanolayers with different thicknesses, and were designated as W1, W2 and W3, respectively. The synthetic diamond particles (Type Ib, HWD40 from Henan Huanghe Whirlwind International Co. Ltd., China), with average particle sizes ~200 μm , were first ultrasonicated in distilled water to eliminate impurities on the surface, then were coarsened in nitric acid (65 wt.% HNO_3) to induce pits that may help the sol–gel adsorption, and finally were washed by distilled water and dried. Afterwards, the coarsened diamond particles were stirred in the $WO_3 \cdot nH_2O$ sol–gels for 10 min, and then were filtrated and dried at 60 °C for 6 h. At last, the diamond@ $WO_3 \cdot nH_2O$ particles were heat treated for 30 min at 700–950 °C in a flowing atmosphere of 20% H_2 -Ar, which reduced the WO_3 and resulted in the formation of diamond@W particles.

2.2. Preparation of the diamond@W/Al composites

The diamond@W particles were mixed with the atomized pure Al powders with a purity of 99.9%. The powder mixtures were cold pressed into powder compacts, and then sintered in a graphite mould by VHP [21,22]. The furnace was heated up to 400 °C at a rate of 10 °C/min, and was held for 30 min at 400 °C to degas the powder compact. Afterwards, it was heated up to 650 °C and kept for 90 min, during which an uniaxial pressure of 67 MPa was applied. Finally, after furnace cooling, sintered specimens of 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. For comparison, diamond/Al composites without the W interface layer (abbreviated as “diamond/Al” hereafter) were also fabricated by the same process.

2.3. Characterizations

The phase composition of the diamond@W particles was characterized by X-ray diffraction (XRD) using a D/max-2550 instrument ($Cu K\alpha$), with a step scan rate of 4°/min in the range of 20–80°. The morphology of the W nanolayer and microstructures of the sintered specimens were observed by scanning electron microscopy (SEM) using an FEI Quanta FEG 250 SEM at 20 kV.

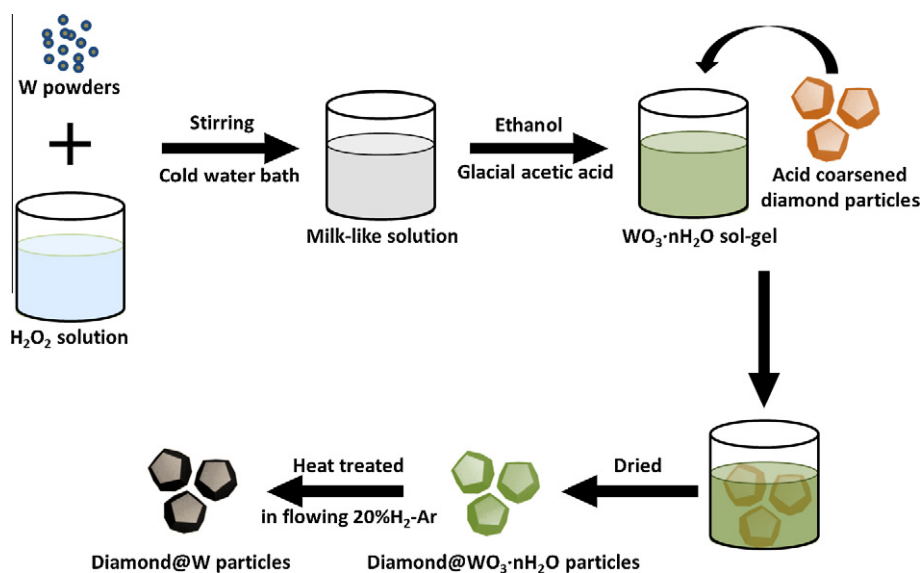


Fig. 1. Schematic illustration of fabricating diamond@W particles by a sol–gel method.

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