Journal of Alloys and Compounds 763 (2018) 305–313

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Enhanced stability of the Diamond/Al composites by W coatings prepared by the magnetron sputtering method

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A R T I C L E I N F O

Article history:
Received 12 April 2018
Received in revised form
24 May 2018
Accepted 25 May 2018
Available online 26 May 2018

Keywords:
Metal matrix composites
Al matrix composites
Diamond
W coating
Interface
Stability

A B S T R A C T

In the present work, the effect of the W coatings prepared by the magnetron sputtering method on the stability of the thermal and mechanical properties of the Diamond/Al composites has been investigated. Due to the decomposition of the interfacial Al_4C_3 phase, the thermal conductivity and bending strength of the Diamond/Al composites without W coating were decreased 23% and 33%, respectively. However, the variation of the normalized thermal conductivity (NTC) of the Diamond/Al composites has been changed from polynomial to linear relationships after the W-coating treatment, indicating the enhancement of the thermal conductivity and the bending strength stability. Diamond/Al composite with 45 nm W coating demonstrated the best thermal and mechanical stability with slight decrement of 3% after 720 h heat-moisture treatment (HMT). Due to the decomposition of the Al_4C_3 phase and the corresponding weakened interfacial bonding, more “clean {111} planes” of the diamond particles were observed in the fracture surface of Diamond/Al composites without W coating after HMT. However, no significant difference was found in the fracture surface of the Diamond/Al composites with 45 nm and thicker W coatings before and after HMT. It is suggested that the continuous, stable and well bonded interfacial structure without the formation of the Al_4C_3 phase is the key for the improvement of the stability of the Diamond/Al composites.

1. Introduction

Diamond particles reinforced Al matrix composites (Diamond/Al) have been considered as the promising thermal management materials in electronic packaging due to their high thermal conductivity [1] and tailorable coefficient of thermal expansion (CTE) [2]. Several afore-stated technologies, such as pressureless metal infiltration [3,4], powder metallurgy (PM) [2,5,6], spark plasma sintering [6–8], gas pressure infiltration [9–11] and pressure infiltration method (squeeze casting) [12–14], have been adopted to prepare the advanced composites with high thermal conductivity. Besides the properties of the Al matrix and the size of the diamond particles, it has been found that the thermal conductivity of the Diamond/Al composites is mainly affected by the interfacial microstructure and the corresponding bonding status. However, due to the large difference in the CTE of the diamond particles and the Al matrix [14], interfacial debonding is usually found in the Diamond/Al composites without interface modification [14–16]. Therefore, optimization of the interfacial bonding to minimize the interfacial thermal resistance (ITR) and to enhance the ways of heat transferring has been considered as the priority

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https://doi.org/10.1016/j.jallcom.2018.05.310
0925-8388/

issue in the Diamond/Al composites [17].

Carbides [18–21] and metallic phases [22–24] have been investigated as the interfacial phases in the diamond/Al composites. By controlling the cooling rate of the modified squeezing casting process, Wang et al. [18] promoted the formation of the Al_4C_3 phase and improved the bending strength (124%) and the thermal conductivity (89%) at the same time. However, Al_4C_3 phase is very easy to react with water in the atmosphere [25], and the corresponding decomposition would deteriorate the thermal conductivity performance of the composites significantly [26]. Yang et al. [27] prepared 300 nm W coating by the magnetron sputtering method, which was then heat treated to form WC to improve the interfacial bonding, and increased the thermal conductivity from 423 to 588 W/(m·K). Zhang et al. [28,29] prepared uniform distributed W coating by the diffusion method and improved the thermal conductivity from 400 to 474 W/(m·K) [29]. Tan et al. [30] and Ji et al. [31] used the sol–gel process to prepare W coatings on the diamond particles and obtained the maximum thermal conductivity value of 599 W/(m·K). In our previous work [32,33], uniform W coatings (35–130 nm) prepared by the magnetron sputtering method has been found to improve the thermal conductivity and mechanical properties of the Diamond/Al composites.

As mentioned in the above, most of the investigations on the Diamond/Al composites have been focused on the design and modification of the diamond/metal interface for short-term properties. However, the effect of the interface on the long-term stability of the performance, which is crucial for the application of the Diamond/Al composites, has been rarely reported. It has been unambiguously reported that the chemical decomposition of the Al_4C_3 would be occurred in a few days in composites exposed to ambient conditions [26,34,35]. Monje et al. [36] observed that the Diamond/Al composites with less Al_4C_3 (shorter contact times) were more prone to a decrease of the thermal conductivity, while the correlation between stability and carbide was up to a great extent opposite to that reported previously in Al-based composite containing other allotropic forms of carbon [35,37]. However, Yang et al. [38] found that thermal conductivity of the Diamond/Al composite was decreased 17% after immersed in water for 48 h due to the decomposition of the Al_4C_3. Meanwhile, Yang et al. [38] also coated W on the surface of the diamond particles by the magnetron sputtering method, and then annealed the W-coated diamond particles in vacuum at 1273 K for 2 h to transform the W layer to be the tungsten carbides. Yang et al. [38] concluded that the stability of the thermal conductivity of the Diamond/Al composites could be significantly improved by the tungsten carbides interfacial layer with only 0.7% decrement.

In our previous work [32,33], Diamond/Al composites with uniform W coatings prepared by the magnetron sputtering method demonstrated the highest thermal conductivity in the Al matrix composites reinforced with 100 μm diamond particles with coatings (622 W/(m·K)). However, to the best knowledge of the authors, the effect of the W coatings on the stability of the thermal and mechanical properties of the Diamond/Al composites has not been explored yet. The purpose of the present work is to investigate the effect of W coatings prepared by the magnetron sputtering method on the stability of the thermal and mechanical properties of the Diamond/Al composites in a heat-moist environment.

2. Material 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 the raw materials in the present study. The volume amounts of diamond particles in the composites were about 55%. The detailed preparation process of W coatings and the Diamond/Al composites have been reported in our previous work [32]. The W-coated diamond particles were ball-milled to broken the W coatings, and then the thickness of the W coatings was measured by the atomic force microscope (AFM), and the profiles of the thickness indicated that the average thickness values of the W-coatings for 90, 180, 270 and 360 min were about 35 ± 2, 45 ± 3, 100 ± 2 and 130 ± 2 nm, respectively [32]. For comparison, Diamond/Al composite without W coating was also prepared by the same process. The heat-moisture treatments (HMT) were performed at 65 °C with relative humidity of 90% up to 720 h. The treatments were carried out in the Temperature-Humidity Alternating Chamber (THA-1000PF, Dongguan Haotian Test Equipment Co., Ltd. China), and the temperature and the humidity accuracy of the chamber was 2 °C and 2%, respectively.

The thermal diffusivities (k) of the samples with size of Φ12.7 mm × 3 mm was measured on LFA 447 Nanoflash (NETZSCH GmbH, Selb, Germany) at room temperature. Five samples of each composite have been measured to improve the statistical significance of the results. Moreover, in order to eliminate the errors that might be introduced by other factors, the thermal conductivity tests were carried out on the same five samples. Three-point bending tests were performed on Instron 5569 universal electrical tensile testing machine with a span of 30 mm and a crosshead speed of 0.5 mm/min. The dimensions of the bending test samples were 3 mm × 4 mm × 36 mm. Bending tests have been performed on at least four samples to improve statistical significance of the results. The fracture surface of the Diamond/Al composites was observed by FEI Sirion Quanta 200 SEM.

3. Results and discussion

The variation of the thermal conductivities of the Diamond/Al composites without and with W coatings (35, 45, 100, 130 nm) after HMT have been shown in Fig. 1a. It is clear the thermal conductivities of the composites were all decreased after HMT regardless of the thickness of the W coatings. The thermal conductivity of the Diamond/Al composites is mainly affected by the diameter of the diamond particles, properties of Al matrix and the characters of the interface. Usually, the diamond particles and the Al matrix demonstrate relatively good stability in the present condition (65 °C with relative humidity of 90%). Therefore, the decreased thermal conductivity after HMT was mainly due to the evolution of the interfacial microstructure. The thermal conductivity of the Diamond/Al composites without W coating was decreased significantly from 556 to 428 W/(m K). It should be noted that the interface of the Diamond/Al composites without W coating was mainly characterized by the interfacial debonding and the Al_4C_3 phase [32], which has been reported to be detrimental for the thermal stability of the Diamond/Al composites [26,38]. However, the thermal conductivities of the Diamond/Al composites with W coatings were slightly decreased after 720 h HMT compared with that of the composites without W coating, which agrees well with the results reported by Yang et al. [38]. Therefore, the interfacial layer is beneficial for the improvement of the stability of the composites exposed to moist environments.

To be more comparative, the thermal conductivities of the Diamond/Al composites were normalized to their individual initial results, as shown in Fig. 1b. It has been found that the thermal conductivity of the Diamond/Al composites without and with 45 nm W coating has been decreased about 23% and 3%, respectively, indicating that the presence of the W coatings improved the thermal conductivity stability. The variation of the normalized thermal conductivity (NTC) of the Diamond/Al composites without W coating could be fitted with adjust R-squared value of 0.9992 as: