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Thermal conductivity of Cu/diamond composites prepared by a new pretreatment of diamond powder

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ABSTRACT

Cu/diamond composites were fabricated by spark plasma sintering (SPS) after the surface pretreatment of the diamond powders, in which the diamond particles were mixed with copper powder and tungsten powder (carbide forming element W). The effects of the pretreatment temperature and the diamond particle size on the thermal conductivity of diamond/copper composites were investigated. It was found that when 300 μm diamond particles and Cu–5 wt.% W were mixed and preheated at 1313 K, the composites has a relatively higher density and its thermal conductivity approaches 672 W $(m\,K)^{-1}$.

1. Introduction

Thermal considerations in electronic package design are becoming critically important for the electronic devices generate more heat owing to the rapid evolution of integration technology [1,2]. High performance packages and heat sink materials having relatively low coefficient of thermal expansion (CTE) in combination with efficient thermal conductivity (TC) are required [3–5]. Diamond–copper composites are considered very attractive to meet the ever-increasing demands. Copper which has a good thermal conductivity of 400 W $(m\,K)^{-1}$ and a higher coefficient of thermal expansion of $17 \times 10^{-6}\,K^{-1}$ can be used as the binder for composites, synthetic diamonds of Ib type which possesses the thermal conductivity of 1500–2000 W $(m\,K)^{-1}$ with a low CTE of about $1.0 \times 10^{-6}\,K^{-1}$ can be used as the filler for composites [6–8].

Such composites were fabricated by different fabrication methods such as spark plasma sintering (SPS) [9], metal infiltration [10,11], hot pressing [12]. However, regardless of preparation technology, the thermal conductivity of diamond/Cu composites is much lower than that of copper itself when the diamond particles compound directly with copper. The reason for this is that Cu does not wet diamond since copper is a non-carbide forming material and has little carbon affinity; as a result, their interface thermal resistance is too high, which is the main problem in obtained high thermal conductivity diamond–Cu composites. In order to reduce the interface thermal resistance, alloying Cu with minor amounts of carbide of carbide formers, e.g. B, Ti, Cr, and W are introduced into the preparation process of metal infiltration and hot pressing [8,13]; coating the diamond particle surface with carbide formers by vacuum micro-deposition technology and sputtering technology are introduced before the rapid sintering [14,15].

In this work, a new and unique diamond powder surface pretreatment technology is proposed to overcome the interface problems. In the proposed method, the diamond particle surface is coated with carbide former W.

In this paper, the pretreatment method is selected as the diamond surface coating technique for three reasons: (1) thin interface layer of a carbide phase was formed, which is contributed to the decrease of thermal resistance and enhancement of wetting properties between the diamonds and copper; and (2) the uniformity of carbide former distribution on the diamond surface was obtained when the carbide forming elements were transferred to the surface of diamond by the fluidity of the liquid copper; and (3) the pretreatment coating method is better than the other coating technology in the diamond particle surface treatment, which including the vacuum micro-deposition technology and the sputtering technology. The new pretreatment coating method not only solves the interface problem with a lower price by a simple vacuum furnace but ensures the thermal conductivity of the copper matrix without any formation of copper alloy in copper matrix.

The critical issue is to control the carbide layer to maintain a minimum interfacial thermal barrier resistance. The thermo-physical properties will be displayed and discussed as a function of the

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temperature and the diamond particle size using for the pretreatment process.

2. Experimental

Diamond/copper composites were made by SPS after the surface pretreatment process of diamond particles. The oxygen-free copper powder (99.99% in purity) with particle size in the range 5–10 μm was used as the composite matrix. The reinforcements were 100–300 μm synthetic MBD8 type diamond powder with a nitrogen content of 200 ppm, purchased from Henan Hengxiang diamond Co., Ltd. And its thermal conductivity is estimated to be about 1500 W/m K according to the level of nitrogen content. Carbide former W was added on diamond surface by mixing diamond powder with composite powder of Cu powder and W powder at high temperature. The composition was Cu–5 wt.% W (mass fraction).

The pretreatment of the diamond powders was carried out in vacuum induction furnace at 1273–1373 K in a graphite crucible for 20–80 min under hydrogen atmosphere. After the pretreated diamond powder being cooled, the redundant copper and the carbide forming element W was removed by passing through a sieve. And then the pretreated diamond powder was ultrasonically cleaned in alcohol, dried and kept in desiccators.

Subsequently, the pre-treated powder was mixed with a designated amount of Cu powder to prepare diamond/Cu composites. These composites powders were heated to 1200 K within 6 min and held for 6 min in spark plasma sintering (SPS) furnace. The preparation methods not only solves the interface problem but ensures the thermal conductivity of the copper without any formation of copper alloy in copper matrix.

Based on Archimedes' law, the bulk density of the composites was measured. The microstructure was analyzed by scanning electron microscopy (SEM) using FEI Sirion-200. Thermal conductivities of the composites were measured by Xenon pyrometry with Netzsch LFA457 equipment.

3. Results and discussion

3.1. Composite characterization

SEM picture (Fig. 1) reveals a pronounced difference in morphology of the interfacial phase on pretreated diamond surface with different pretreatment temperature and diamond particle

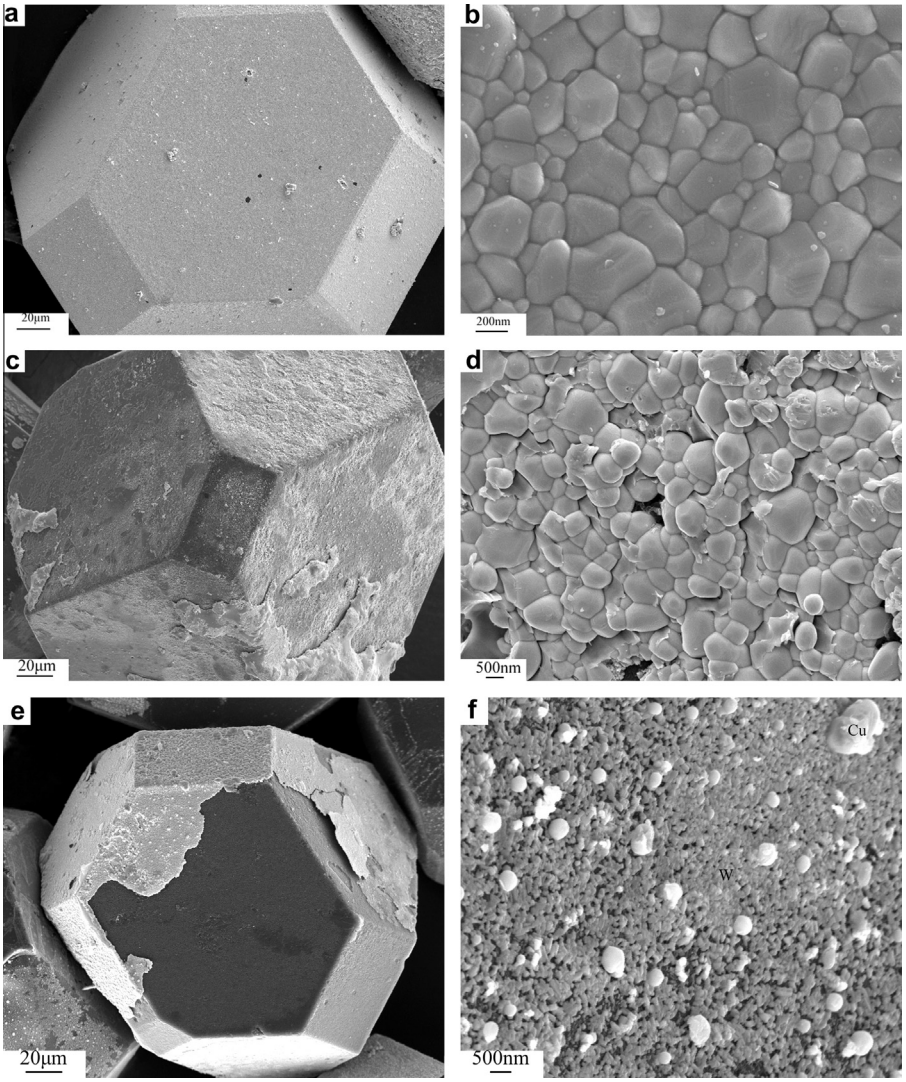

Fig. 1. SEM pictures of 200 μm diamond particle pretreated by the high temperature mixing process. (a and b) Diamond particle pretreated at 1293 K; (c and d) diamond particle pretreated at 1313 K; (e and f) diamond particle pretreated at 1373 K.