Advanced Materials Research Vol. 59 (2009) pp 169-172
online at http://www.scientific.net

Fabrication and Properties of Copper/Carbon Composites for Thermal Management Applications

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Keywords: Diamond composites, Powder Metallurgy, Thermal conductivity

Abstract. The ideal thermal management material working as heat sink and heat spreader should have a high thermal conductivity combined with a reduced and tailorable thermal expansion. To meet these market demands copper composites reinforced with diamond particles were fabricated by a powder metallurgical method (powder mixing with subsequent pressure assisted consolidation).

In order to design the interfacial behaviour between copper and the reinforcement different alloying elements, chromium or boron, were added to the copper matrix. The produced composites exhibit a thermal conductivity up to 700 W/mK combined with a coefficient of thermal expansion (CTE) of 7-8 x 10^{-6}/K. The copper composites with good interfacial bonding show only small decrease in thermal conductivity and a relatively stable CTE after the thermal cycling test.

Introduction

During the past decades, the rapid evolution of integration technology has resulted in a significant increase in electronic device density and speed, and such a rate of increase is expected to continue for the near future. With such progress problems arise with heat generation on the semiconducor chips. Actually, more than 50% of electronics failures are caused by an insufficient cooling.

To achieve long life and reliable performance of these components, it is necessary to keep the operating temperature of an electronic device within specified limits. Therefore, the effective thermal management is a key issue for packaging of high performance semiconductors.

One of the most promising materials is copper reinforced with diamonds. Using the simple rule of mixture the thermal conductivity of the composite with 60 vol% diamonds should be in the range of 1000 W/mK. When diamond particles are embedded in a copper matrix, the interface plays a crucial role in fixing the thermal conductivity, the CTE and also the mechanical properties of the composite. An ideal interface should provide good adhesion and minimum thermal barrier resistance.

In order to solve the interface problem between copper and diamond the use of different carbide formers added as alloying elements to the copper matrix has been investigated. It is assumed that a very thin interface layer of metallic ceramics can aid the necessary electron-phonon coupling. A high thermal conductivity was achieved for diamond reinforced CuCr or CuB matrix composites [1-4]. In addition, fast field/pressure-assisted sintering techniques (e.g. SPS) with heating/cooling rates of up to 300 K/min and holding times up to some minutes result in the most promising thermal conductivity values compared to the conventional hot pressing with heating/cooling rates of about 10 K/min [1, 5].

In the present work, an optimization of the composite and the used pressing technique results in a further improvement of the thermal conductivities up to 700 W/mK. The critical issue is the control the process parameters in order to maintain a maximum thermal interface conductance. The thermo-physical properties will be displayed and discussed as a function of the reinforcements as well as the alloying element used for composite preparation.

Experimental

Both, a pre-alloyed CuCr0,8 (in mass-%) powder prepared by gas atomization and elemental boron powder (1,0 mass-%) were used to investigate the effect of adding alloying elements to the Cu matrix.

The used reinforcements are synthetic diamond grit of mesh 400/500, 325/400, 120/140 and 70/80. This corresponds to average particle diameters of 30 µm, 40 µm, 110 µm and 195 µm, respectively. The synthetic (Ib-)diamond was of the MBD4 type purchased at Luoyang High-Tech Qiming Superhard Materials Co. Ltd., Luoyang, Henan, China. An optimized particle-size distribution was selected for the diamond particles used for composite fabrication in order to realize a high volume content by increasing the packing density of diamonds in the Cu/diamond composites.

Subsequently, the copper powders were mixed with the diamond particles to prepare composites with 50-65 vol% reinforcement. These composites were fabricated by hot pressing with a direct resistance heating for very fast heating (using FCT-HP D 250/1 of FCT Systeme GmbH, Germany or DSP 510-745 of Dr. Fritsch Sondermaschinen GmbH, Germany). The samples were heated by a pulsed or non-pulsed electric current which flows through the punch-die-sample-assembly using a high current and low voltage.

The thermal conductivity of the composites was measured by Xenon pyrometry with the Nanoflash of Netzsch, Germany. The measurement of the CTE was performed in a Netzsch dilatometer.

The interface area on the diamond composite fracture surfaces was studied using high resolution scanning-transmission electron microscope HR-STEM S5500 from Hitachi. TEM observations of the interface structure were also performed on the thin foils prepared by Focused Ion Beam (FIB) milling.

Results and Discussion

The results presented in Table 1 show that the alloying of the copper matrix with the carbide-forming element chromium and boron has a positive effect on the thermal properties of the Cu/diamond composites and can result in a high thermal conductivity up to about 700 W/mK. The necessary high diamond contents were achieved by using bi- or trimodal distributions of the diamond particles. The corresponding CTE was reduced to about (7-9) x 10^{-6}/K depending on the filler content by the improved interfacial bonding between the copper matrix and the diamond particles.

Without alloying a rather low thermal conductivity of the composite (~ 200 W/mK) was measured indicating a high thermal barrier resistance. This can be explained by no chemical affinity between copper and diamond, and, therefore, it is difficult to produce a bond of low thermal resistance and high mechanical strength between the matrix and the reinforcement. The increased thermal conductivity of the composites also demonstrates the effectiveness of the formed carbide layer to obtain a good thermal contact between the matrix and the diamond particles.

Fig. 1 shows the HR-SEM image of the fracture surface of a CuCr/diamond composite made by field activated sintering. A continuous interlayer of a contrast different from those of diamond and copper, of some 100 nm in thickness, is clearly visible in the SE mode. The electron diffraction analysis confirmed the presence of the chromium carbide (Cr_3C_2) at the Cu/diamond interface.

Table 1. Thermal properties of selected copper/diamond composites.

Material	Thermal diffusivity [mm^2/s]	Specific heat [J/gK]	Thermal conductivity [W/mK]	CTE (30-100°C) [$10^{-6}/K$]
$CuB/CD/50_p^*$	215	0,42	525	9,1
$CuCr/CD/50_p^*$	240	0,43	610	8,7
$CuCr/CD/60_p^*$	305	0,43	725	8,1
$CuCr/CD/65_p^*$	300	0,43	660	7,1

*vol% of the diamond particles

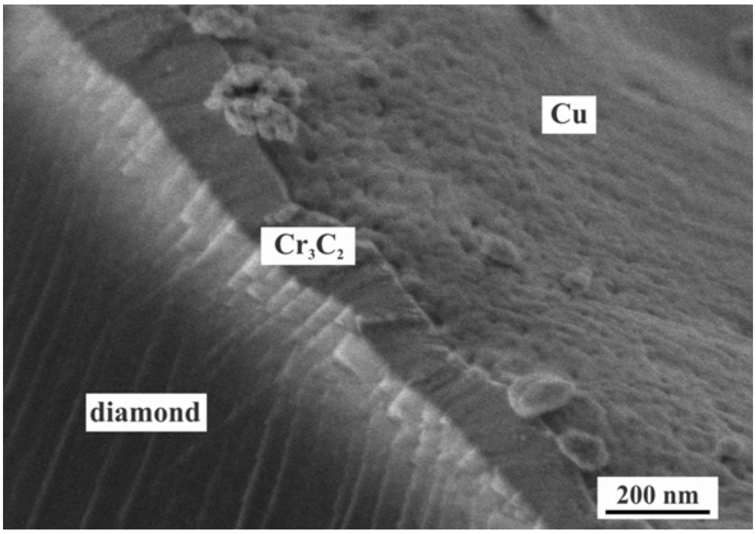

Figure 1. HR-SEM micrograph (using SE signal) of the fracture surface of $CuCr/CD/50_p$ diamond composite.

The critical issue is to control the process parameters at such a rate that the carbide layer is not growing too strongly in order to maintain a maximum thermal interface conductance or a minimum thermal boundary resistance. This thermal resistance consists of three terms, namely the resistance at the diamond/carbide interface, the resistance of the carbide itself, and the resistance of the carbide/metal interface. The intrinsic conductance of the layer may be calculated by dividing its thermal conductivity by its thickness; a Cr_3C_2 (λ = 19 W/mK [6]) layer of 100 nm thickness would thus have a thermal conductance of 19 x 10^7 W/m^2K, which is much higher than the thermal boundary conductance of about 3-4 x 10^7 W/m^2K for the CuCr/diamond composites using the model of Hasselmann and Johnson [7], which allows to estimate the thermal conductance of the interface in a composite containing nearly globular particles. This result shows that the interfacial reaction layer should have a sufficiently low thickness (<0.1µm) because thicker carbide layers tend to impair both the bonding characteristics and the heat transfer ability, if the diamond is completely covered by this carbide layer. In addition, the level of covering the diamonds with the carbide layer and the thermal resistances at both interfaces (diamond/carbide and carbide/copper) play a key role in manufacturing of highly conductive copper composites.

Overall, the resistance against thermal fatigue in the diamond composites made by powder metallurgy was satisfactory. The thermal cycling resulted in a small reduction of thermal diffusivity of the composites between 5% and 10% and relatively stable CTE after the thermal cycling.

Summary

Copper based composites containing 50-65 vol% diamond reinforcement were fabricated by hot pressing of powder mixtures. This work demonstrates that high thermal conductivities can be achieved for diamond reinforced CuCr or CuB matrix composites. The control of the interfacial reaction resulting in the formation of a nano-sized carbide layer is crucial to enable the manufacturing of Cu/diamond heat sinks with high thermal conductivities up to about 700 W/mK combined with CTE of 7-8 x 10^{-6}/K.

Acknowledgements

A part of this work has been performed within the framework of the Integrated European Project "ExtreMat" (contract NMP-CT-2004-500253) with financial support by the European Community. It only reflects the view of the authors and the European Community is not liable for any use of the information contained therein. An other part of this work was founded by the German Federal Ministry of Education and Research. The authors would like to thank Dr. Ł. Ciupiński from the Warsaw University of Technology, Faculty of Materials Science and Engineering for the HR-SEM/TEM studies.

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