Materials and Design 120 (2017) 170–185

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Design of interfacial Cr_3C_2 carbide layer via optimization of sintering parameters used to fabricate copper/diamond composites for thermal management applications

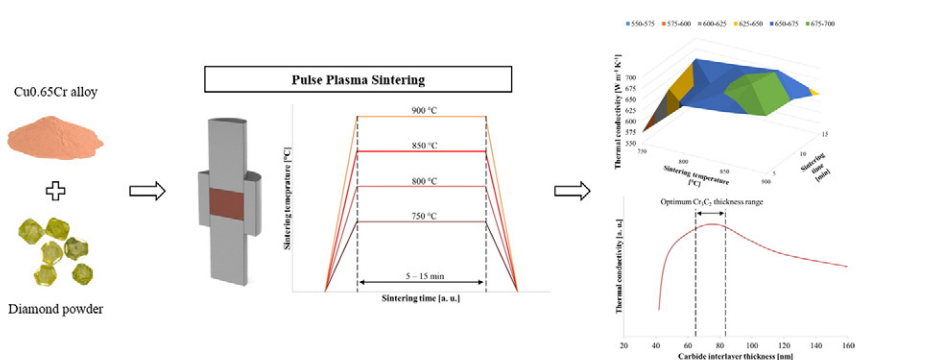
Łukasz Ciupiński [a], Mirosław J. Kruszewski [a,*], Justyna Grzonka [a,b], Marcin Chmielewski [b], Radosław Zielińsk [a], Dorota Moszczyńska [a], Andrzej Michalski [a]

[a] *Warsaw University of Technology, 141 Wołoska str., 02-507 Warsaw, Poland*
[b] *Institute of Electronic Materials Technology, 131 Wólczyńska str., 01-919 Warsaw, Poland*

HIGHLIGHTS

- Cu0.65Cr/50 vol.% diamond composites were fabricated via pulse plasma sintering.
- Varying sintering parameters were applied to optimize the fabrication process.
- Cr_3C_2 carbide formed at the matrix/diamond interface.
- The sample fabricated at 850 °C in 10 min possessed the maximum thermal conductivity of 687 W m^{-1} K^{-1}.
- The optimum thickness range of the interfacial carbide layer was estimated.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 30 November 2016
Received in revised form 3 February 2017
Accepted 5 February 2017
Available online 6 February 2017

Keywords:
Metal matrix composite
Particulate diamond filler
Cr_3C_2 coating
Carbide interface thickness
Pulse plasma sintering
Interface thermal conductance

ABSTRACT

To produce metal-diamond composite materials with high thermal conductivity, it is important for a high-quality carbide interface to exist between the metal matrix and diamond. The addition of carbide-forming elements to the matrix positively influences the interfacial thermal conductance (ITC), and is an effective method for improving the bulk thermal conductivity of composite materials. Diamond powder was mixed with Cu0.65Cr alloy powder, using a 1:1 volume ratio. The pulse plasma sintering (PPS) parameters were optimized to control the carbide interface between the diamond and matrix. The microstructures and phase compositions of the fabricated materials were examined using scanning electron microscopy (SEM) and X-ray diffraction (XRD). The interfacial layer was characterized using SEM and focused ion beam (FIB) techniques. The residual Cr content of the matrix was estimated, to determine its influence on the thermal properties of the matrix. To calculate the ITC, differential effective medium (DEM) and Hasselman-Johnson (H–J) models were used. The highest thermal conductivity of 687 W m^{-1} K^{-1} was achieved by a composite material that was fabricated at 850 °C over a period of 10 min, which had an 81-nm-thick interfacial carbide layer. An ITC_{DEM} value of $5 \cdot 10^7$ W m^{-2} K^{-1} was obtained.

* Corresponding author.
E-mail addresses: lciupins@inmat.pw.edu.pl (Ł Ciupiński), m.kruszewski@inmat.pw.edu.pl (M.J. Kruszewski), j.grzonka@inmat.pw.edu.pl (J. Grzonka), marcin.chmielewski@itme.edu.pl (M. Chmielewski), radosllawzielinski@gmail.com (R. Zielińsk), amichalski@inmat.pw.edu.pl (A. Michalski).

http://dx.doi.org/10.1016/j.matdes.2017.02.005

Table 1
The solubility of selected carbide-forming elements, and their ability to form compounds with Cu [13].

Element	Al	Ti	Si	Fe	Hf	Cr	Zr	Nb	V	B	Ta	W
Solubility in Cu [wt.%]	9.4	6	5.4	4.1	1.1	0.73	0.172	0.15	0.1	0.05	Insoluble	Insoluble
Chemical compounds with Cu	Yes	Yes	Yes	No	Yes	No	Yes	No	No	No	No	No

1. Introduction

Metal matrix composites (MMCs) are a broad group of materials that may combine unique properties that the matrix and filler cannot achieve separately. For example, these properties may include high mechanical strength with low density or high plasticity [1–4]. Considering the thermal management of high power electronics, such as insulated gate bipolar transistors (IGBT) or laser diodes, it is important to combine high thermal conductivity with an adjustable and relatively low coefficient of thermal expansion (CTE) [5]. Certain MMC-group materials can possess such properties; these include composites in which the matrix is composed of a highly thermally conductive metal (e.g., Al, Cu, Ag) with a filler that possesses high thermal conductivity and a low CTE (e.g., SiC, diamond, AlN). By careful selection of the constituent volume fractions, a composite with optimal thermal properties may be designed. Diamond is considered one of the best fillers because it possesses the highest thermal conductivity of all known bulk materials, reaching 2500 W m^{-1} K^{-1}, as well as a CTE of $1.8 \cdot 10^6$ K^{-1} [6].

When fabricating a MMC with a diamond filler, it must be noted that metals such as Cu or Ag do not form carbon compounds, and therefore, in general, will not be able to provide high quality, low interfacial thermal conductance (ITC) at the matrix/diamond interface; the exception to this rule includes composite materials fabricated with high-pressure high-temperature techniques [7,8]. Among the aforementioned examples of matrix metals (Al, Cu, Ag), only Al can bind to diamond because of its carbide-forming ability [9–11]. In the two other cases, it is necessary to introduce alloy additives (carbide-formers), whose presence within the matrix may impair the thermal conductivity of the material [12]. Table 1 shows the solubility of selected carbide-forming elements and their ability to form compounds with Cu.

To date, several methods have been developed to control the morphology and thickness of the matrix/diamond interface, which have been reported in literature. The first method (indirect) involves modification of the chemical composition of the matrix alloy via the selection of carbide-forming elements at varying contents; the other parameters are kept constant (e.g., the fabrication parameters, and the size and volume ratio of the diamond filler). An example of this type of work has been conducted in a previous study [14], in which Cu/diamond composites were fabricated via gas pressure infiltration (GPI). The infiltration parameters and volume fraction of the diamond filler were kept constant, and therefore, optimization of the CuB and matrix alloy was possible. The CuB and CuCr matrices exhibited maximum thermal conductivities of 600 and 700 W m^{-1} K^{-1}, respectively. Another study was conducted [15], in which a Cu matrix with various Zr contents was used for the fabrication of composites with an unvarying diamond content. When a Cu1.2Zr (wt.%) matrix alloy was used, the maximum thermal conductivity of the hot-pressed composites reached a value of 615 W m^{-1} K^{-1}; the thickness of the ZrC interfacial layer was approximately 320 nm. The same carbide-forming additive was used in another study [16] for a Cu/diamond MMC produced via GPI. The Cu0.5Zr (wt.%) matrix alloy had a thermal conductivity of 930 W m^{-1} K^{-1}, a record high result; the thickness of the carbide layer was approximately 400 nm. The effect of B on the thermal properties of Cu/diamond composites was examined by Fan et al. [17]; they showed that a composite with an initial matrix composition of Cu0.3B (wt.%), fabricated by GPI, had a maximum thermal conductivity of 711 W m^{-1} K^{-1}.

The second method (also indirect) of designing a matrix/diamond interface involves careful selection of the fabrication parameters of the composites. An example of this approach was presented in another study [18], in which Al/diamond composites were produced by GPI. The parameters that affected the matrix/diamond interface included the infiltration temperature, and the period of contact between the liquid Al and diamond filler. The optimal contact periods for two infiltration temperatures, 760 and 850 °C, were determined. At temperatures of 760 and 850 °C, the maximum thermal conductivities achieved were 688 and 746 W m^{-1} K^{-1}, respectively. The authors noted that each reactive system must be subjected to thorough research with regard to the fabrication parameters because these parameters control the interface between the matrix and diamond. Another study particularly focused on the fabrication parameters of MMCs [19]. Here, the authors used a hot pressing (HP) technique to fabricate Al/diamond composites. This technique, as well as other powder metallurgy techniques, introduces a number of fabrication process variables such as time, temperature, and the consolidation pressure of the composite. In this study, the optimum temperature, pressure, and heating time were determined to be 650 °C, 67 MPa, and 90 min, respectively; the maximum thermal conductivity achieved was 567 W m^{-1} K^{-1}. In the case of AlSi/diamond composites, much shorter heating times are required

Notes to Table 2
[a] wt.%.
[b] Bimodal distribution.
[c] at.%.
[d] Reaction product of matrix and diamond due to fabrication process.
[e] 1–5 at.%.
[f] x = 36–330 μm.
[g] Surface roughened.
[h] Mixture of Al + 10 vol.% 5056 alloy.
[i] Following heat treatment: 12 h at 540 °C and 40 h at 400 °C.
[j] Following infiltration process: TiC, Ti_3Al and other phases form Ti-Al-Si system.
[k] Additionally coated with 2.82 μm of Cu.
[l] $W_{0.6}Cu_{0.4}$ pseudo alloy.
[m] Different boron architectures: nanometric wires, sheets, or rods.
[n] Size of copper particles on diamond surface.
[o] 0.1–1 wt.% of B.
[p] 0.05–1.85 wt.% of B.
[q] 0.08–0.82 wt.% of Cr.
[r] Direct resistance heating.
[s] 0.2–2.5 at.% of Ti.