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Ultra-rapid processing of high-hardness tungsten–copper nanocomposites

Walid M. Daoush [a], J. Yao [b], M. Shamma [b], K. Morsi [b,*]

[a] Department of Production Technology, Faculty of Industrial Education, Helwan University, 30 El Sawah Street, 11511-11668 Cairo, Egypt
[b] Department of Mechanical Engineering, San Diego State University, 5500 Campanile Dr., San Diego, CA 92182, USA

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ABSTRACT

This paper discusses the fabrication of tungsten (W)–copper (Cu) nanocomposites via an ultra-rapid Current-Activated Melt Infiltration (CAMI) process to produce W–Cu nanocomposites for the first time in a matter of seconds. This was accomplished through a unique composite powder layer arrangement that promotes efficient resistive heating as compared to microscopically mixed powders. The effects of current intensity on the developed macro/micro- and nano-structure are discussed. Due to the short duration of processing and limited grain growth, the nanocomposites produced the highest hardness reported. Multiphysics modeling was used to obtain an insight into the temperature distribution immediately prior to Cu infiltration.

1. Introduction

The interest in reinforcing tungsten (W) with copper (Cu) stems largely from the ability of copper to improve the thermal and electrical conductivities of tungsten [1], in addition to improving its machinability [2]. A large number of processes have been used to produce W–Cu composites, including mechanical alloying [3], liquid phase sintering [4], spark plasma sintering (SPS) [5], extrusion [6], hot pressing [7], combustion synthesis [8] and direct laser sintering [9]. Due to the high melting point of W compared to that of Cu, and favorable interfacial energies allowing Cu to wet W, it has been possible to infiltrate a porous W medium with molten Cu. A number of researchers have investigated such an approach, including work by Hamidi et al. [10], and recently Xu et al. [11] who made use of microwave energy to allow melting and infiltration of Cu at 1200 °C for 3 h in a porous W medium. Such work is valuable in providing novel approaches for the production of this important composite system. However, being able to ultra-rapidly produce such materials would not only be advantageous from the economical prospective but also have important microstructural benefits, for example minimizing W particle coarsening. The present paper discusses the *ultra-rapid* infiltration of nano-scale W powder layers by electrically melting micro-scale copper powder. This was achieved by a miniaturized spark plasma sintering setup that allows the application of ultra-high current densities. A recently published viewpoint set on SPS, can be found in [12]. It is interesting to note that Centeno et al. [13] using a multi-step process produced a carbon preform which was later infiltrated with silicon using a large scale SPS apparatus where the total infiltration cycle took 1 h. In our unique setup for W–Cu processing, the whole process takes only 10 s or less without the need for multistep processing to produce the nanocomposite. The effect of current intensity on the degree of infiltration, and developed micro/nano-structure is discussed. Moreover, finite element modeling is used to provide insight on the temperature distributions within the miniature layered preforms just prior to infiltration. In addition, the mechanical response of the material was investigated using micro-hardness.

2. Experimental procedure

Tungsten powder (~100 nm diameter, Jin Sheng International Industrial LTD) and copper powder (~45 μm, GFS Chemicals) were used in the experiments. Panels a and b in Fig. 1 are field emission scanning electron micrographs of the powders showing their sizes and morphologies. Although the individual size of W particles is approximately 100 nm, they agglomerate to a micro-scale size as seen in Fig. 1a.

Powder layers of W and Cu were stacked inside a mica die (outer and inner diameters of 10 mm and 1.5 mm respectively) in an arrangement where Cu powder was sandwiched between two W powder layers as shown in Fig. 1c. Mica was selected as the die material due to its electrical insulating properties. Fig. 1c shows a schematic of the experimental setup. A spring-loaded punch was applied on the whole arrangement, generating an initial pressure of ~5 MPa which produced porosities of ~88% in the W layers and ~57% in the Cu layer (obtained by separate tap density experiments on Cu and W powder). The experimental setup is essentially a miniaturized spark plasma sintering arrangement where a direct current (DC) up to 100 A was applied to the W–Cu–W powder layers through stainless steel punches under an argon atmosphere. A DC current of 100 A is equivalent to a nominal current density (current density = current intensity / cross-sectional area) (J_N) of

* Corresponding author.
E-mail address: kmorsi@mail.sdsu.edu (K. Morsi).

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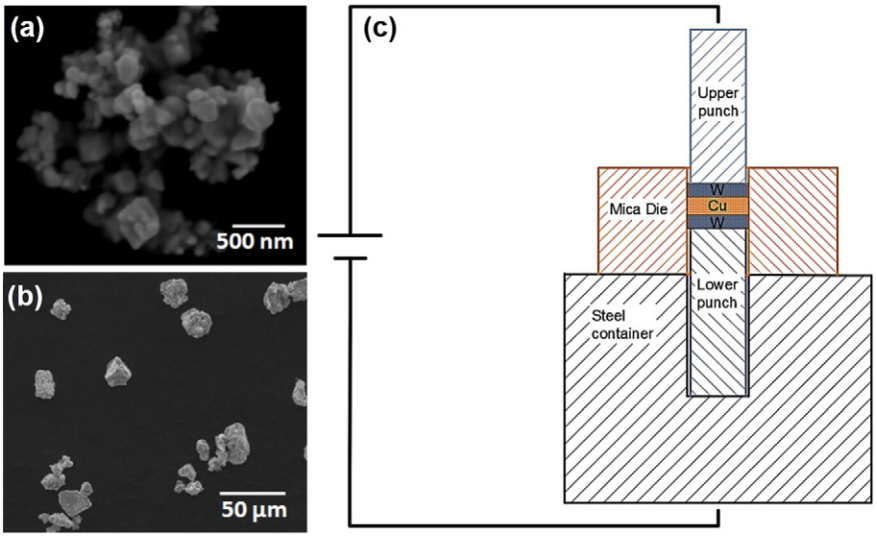

Fig. 1. Powders used and CAMI experimental setup. SEM images of the investigated (a) tungsten, and (b) copper powders. (c) Schematic diagram of the CAMI setup.

~5.7 kA/cm^2. Moreover, if the initial porous nature of the powder layers is considered, the *effective* current density (J_E) could be defined as [14]:

$$\frac{J_E}{J_N} = \left(\frac{1}{1-\theta^{2/3}}\right) \quad (1)$$

where J_N is the nominal current density = (current intensity / cross-sectional area) and θ is the fractional porosity. Using an initial pore content of 0.88 and 0.57 for W and Cu powder layers, gives initial effective current densities of 12.2 and 3.2 times the nominal current density respectively. This means the W layer experiences an outstanding ~70,000 A/cm^2 as effective current density while the Cu layer is subjected to ~18,000 A/cm^2. Such extraordinary high current densities enabled by our miniature SPS setup has led to an ultra-rapid heating, and subsequent melting and infiltration process, that is complete in a matter of seconds.

Several W–Cu nanocomposite samples were obtained by changing the current intensity and application time. Microstructural analysis of the processed nanocomposites was obtained using a field emission scanning electron microscope (Hitachi TM3000). Microhardness measurements were conducted using a Wilson Instruments Vickers microhardness tester, Model 402 MVD, at different locations on the processed W–Cu composite samples, using a 50 g load. Temperature measurements of the samples during processing were attempted but found to be unreliable due to the miniaturized scale size of the present setup.

COMSOL multiphysics modeling and simulation software was used to model the initial heating stage in the CAMI process for W–Cu nanocomposite. As a simplifying assumption, each powder layer was treated as a continuum. Consequently, the results generated from COMSOL in this present paper are intended to provide a preliminary insight into the temperature distribution within the 3-layered structure at time-scales prior to Cu melting and infiltration, however further work is still needed to refine the model and account for the particle nature of the powder layers in addition to other factors (the subject of a separate study). Table 1 shows the material properties of W, Cu and stainless steel used in COMSOL simulation (the upper and lower punches shown in Fig. 1c schematic are made from stainless steel).

Transient heat transfer analysis was used in the model to observe the temporal temperature distributions at times prior to Cu melting. In the model, a current of 100 A was assumed to pass through the W/Cu/W powder layers. Due to the limited development in the powder properties of materials especially nanopowders, assumptions are made for the electrical resistivity input into COMSOL modeling software. Owing to the high surface area of powders, it is assumed that the resistivity of micro-scale Cu and nano-scale W powders used here are 20 times and 3.6 times that of their bulk forms (Table 1, COMSOL assumptions), this was based on a recent study that examined the effect of pore content on the electrical conductivity of powder-based materials [15].

3. Results and discussion

At 10 A applied current intensity, the W–Cu–W powder layers arrangement remained powdery, with no evidence of copper melting and consequently no infiltration. As the amperage was raised to 30 A, the transformation of copper powder to a molten state was evident resulting in one large copper coagulated particle, which was also the case at 55 A current intensity. Fig. 2a shows that even at 55 A, and despite copper melting, very limited infiltration into the W layer was observed (no more than ~10 μm). At 75 A for 10 s, infiltration was evident beyond the 10 μm layer (Fig. 2b), while at 100 A larger-scale infiltration was observed (Fig. 2c and d). The 100 A infiltrated W–Cu nanocomposite compact cross-section (Fig. 2d) shows that the middle copper layer was not completely consumed, leaving behind copper with a thickness in the order of tens of micrometers in the middle region of the sample.

Regions in the compact that have been well infiltrated, show a high degree of densification with limited pore content, while in other parts of the compact, more exaggerated porosity have been observed. The pores in the well infiltrated regions appear at two size scales, 0.8–1.5 μm and less than 200–400 nm, as seen in Fig. 3. Some regions show localized W–W particle sintering while others show this to a lesser extent, giving some slight variation in W particle size. However, in general, limited W particle growth was evident, due to the short processing duration of 5 s, resulting in an average W particle size of 300 +/− 66 nm.

From our experimental observations, it is clear that the rapid infiltration occurs within a few seconds. Hence during this short time, the melting of Cu, is followed by rapid infiltration into the W layers,

Table 1
Electrical resistivity, melting point and specific heat capacity of W, Cu and stainless steel (SS) used in COMSOL.

Properties	W	Cu	SS
Density (kg/m^3)	19,250	8960	7930
Melting point (K)	3695	1358	1700
Thermal conductivity (W/mK)	173	368.7	16.3
Specific heat capacity (J/kg K)	132	385	500
Temperature coefficient of resistance (1/K) ($\times 10^{-3}$)	4.5	3.86	0.94
Electrical resistivity (Ωm) ($\times 10^{-3}$) (for solids)	117.6 (5.65)	6.12 (1.70)	72 (72)