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In suit synthesis and bonding of Cu to W–Cu composite by combustion synthesis and centrifugal infiltration

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ABSTRACT

A novel method for the in suit synthesis and bonding of Cu to W–Cu composite was developed by combining combustion synthesis with centrifugal infiltration. Highly exothermic combustion reaction in Al–CuO system was utilized to realize high temperature and produce molten products. The application of high gravity field of 1000*G* resulted in a rapidly phase separation of Cu melt and infiltration of W loosen powder. Composition and microstructure showed a layered variation along high gravity direction. Cu was bonded to W–Cu composite to form a Cu/W–Cu integral material. Columnar grain microstructure was observed on Cu layer due to a fast cooling rate produced along high gravity direction. Pores were found concentrated at the interface between Cu and W–Cu layer. The evolution of thermal conductivity and hardness along high gravity direction were investigated.

1. Introduction

Tungsten–copper (W–Cu) composites combining high thermal conductivity and high electrical conductivity of Cu with low thermal expansion coefficient and high erosion resistance of W have been widely used in may fields as structural, electrical and thermal management materials [1–3]. W–Cu composites with various copper contents have been fabricated by different methods, such as powder metallurgy, pressure-less infiltration, plasma spray and cold spray [4–7]. In general, W–Cu composites with copper contents less than 20 wt.% have been produced by the infiltration of a tungsten skeleton with copper melt [5]. While W–Cu composites containing higher copper contents usually have been manufactured by liquid phase sintering [4]. To reduce the mismatch in mechanical and thermal properties W–Cu functionally graded material was introduced with a gradual transition of compositions and properties from one side to the other [8,9]. However, in some extreme environmental conditions such as high voltage arc contact materials and high heat flux components for the future nuclear fusion reactor W–Cu composite is usually jointed to Cu to assure the reliability and durability of the components [10,11]. Common techniques of joining Cu to W–Cu composite include electron beam welding, diffusion welding, and vacuum electron beam welding [12,13]. As unfavorable filler metal or defects like pore and crack often encountered on the welding interface, the mechanical, thermal and electrical properties of the joints are thus limited. Therefore, it is still attractive to develop fast, inexpensive and simple synthetic methods for the synthesis and bonding of Cu to W–Cu composite with improved bonding interface and properties.

A novel method by combining combustion synthesis with centrifugal infiltration was developed for the in suit synthesis and bonding of Cu to W–Cu composite. In this technique, highly exothermic aluminothermy reaction in Al–CuO system are utilized to realize high temperature and produce molten products. At the same time a high gravity field introduced by centrifugation is applied to accelerate the separation of metallic and ceramic melts and removal of pores from the melts. Meanwhile, the separated high temperature Cu melt is rapidly infiltrated into the W loosen powder placed under it at the drive of high gravity and form a Cu/W–Cu integral material. Combustion synthesis and centrifugal infiltration can offer a fast and economical way to synthesis and bonding of Cu to W–Cu composite in one step. Effect of high gravity on the compositions and microstructures of Cu/W–Cu integral material are discussed. The evolution of thermal conductivity and hardness along high-gravity direction are investigated.

2. Experiments

Commercial powder of Al (100 μm, 99.9% purity), CuO (45 μm, 99.9% purity), Cu (45 μm, 99.9% purity) and W (2.6–5 μm, 99.9% purity) were used as raw materials. The Al and CuO powders were used to produced high temperature and Cu melt by the aluminothermy reaction as follow

$$2Al + 3CuO = Al_2O_3 + 3Cu \quad (\Delta H^\circ_r = -893.69\ \mathrm{kJ/mol},\ T_{ad} = 2846\ \mathrm{K}) \tag{1}$$

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The reactant powders were mixed and homogenized for 1 h by roller ball grinder with a speed of 60 r/min. Then a batch of 200 g mixed powders was cold-pressed into a compact with a diameter of 40 mm and porosity about 50%. The compact was placed above 100 g W powder bed in a graphite crucible with an inner diameter of 40 mm. The crucible was coated with a carbon cap and then put into a steel vessel, which was mounted into a reaction chamber to perform combustion synthesis and centrifugal infiltration. The reaction chamber was evacuated to a vacuum of ~100 Pa. A high gravity field with an acceleration of 1000*G* ($G = 9.8\ m/s^2$) was induced by centrifugation. The combustion reaction was triggered by passing an electric current about 10 A through a tungsten coil closely above the thermite. A schematic illustration of the equipment is shown in Fig. 1. During the reaction a large amount of heat was created and the molten products were separated in a high gravity field due to their density difference. The separated high temperature Cu melt was rapidly infiltrated into the W powder bed to form a Cu/W–Cu integral material. After cooling and solidification ceramic and metal ingots were obtained. The metal ingot was machined and polished for later characterization.

The phase composition was identified by X-ray diffraction (XRD; D8 Focus, Bruker, Germany). The microstructure was examined by scanning electron microscopy (SEM; S-3400, Hitach, Japan). The bulk density (ρ) was measured according to the Archimedes principle. The room temperature thermal diffusivity (α) was measured by laser flash using a transient thermal flash technique (LFA 457, Netzsch, Germany). Specific heat capacity (C_p) was measured using differential scanning calorimetry (TA-instruments, Q100, USA). The thermal conductivity (κ) was calculated from measurements of density, thermal diffusivity and specific heat capacity using the relation $\kappa = \alpha \rho C_p$ [14]. The hardness was determined by a Rockwell hardness tester using a ball indenter of diameter of 1.588 mm at a load of 100 kg applied for 5 s. The average values were obtained from five different independent measurements.

3. Results and discussion

Combustion synthesis and centrifugal infiltration is a complex process including combustion reaction, phase separation, infiltration and solidification. The XRD patterns of the prepared metal ingot analyzed along high gravity direction are presented in Fig. 2, in which only diffraction peaks of W and Cu are observed. The prepared sample shows a layered variation of composition along high gravity direction. Cu phase is on the top layer of the sample and W–Cu phases on the bottom. It can be seen form the digital photo of the obtained sample inserted in Fig. 2, the Cu layer is joined to the W–Cu layer to form a integral materials and have an obviously boundary between them. This indicated that the aluminothermy reaction in Al–CuO system is completed and Cu melt was separated from the lighter ceramic phase, then infiltrated into W loosen powder to form a integral material. It should be noticed that the Cu phase exhibits different crystallization characteristics at different sections of the sample. The Cu phase shows the strongest diffraction peak (200) at $2\theta = 50.433°$ on the transverse section of the Cu layer. However, the strongest diffraction peak change to crystal plane of (1 1 1) at $2\theta = 42.297°$ on the longitudinal section, as can be seen form Fig. 2B. This crystallization behavior is in agreement with that of continuous cast single crystal Cu rod, which has a crystallization orientation of [1 0 0] and crystallization plane of (2 0 0) [15]. Base on the above results and analysis, Cu layer produced under high gravity filed may perform preferential grain growth along (1 1 1) crystal plane, as the temperature gradient is identical with the direction of crystallization. As can be seen from Fig. 2C, the Cu phase has a relative lower diffraction intensity compared with that of W phase. This indicated a relative higher W content on the W–Cu layer.

To clarify the effect of high gravity field on the microstructure of Cu layer, the transverse and longitudinal sections of the Cu layer were etched by a mixture of 5 wt.% $FeCl_3$ solution and 2 wt.% HCl solution for 30 s. it can be seen from Fig. 3A, Columnar Cu grains about 100 μm in width and over 1 mm in length were observed on the longitudinal section, which are paralleled to the high gravity direction. As that happened in direction solidification the

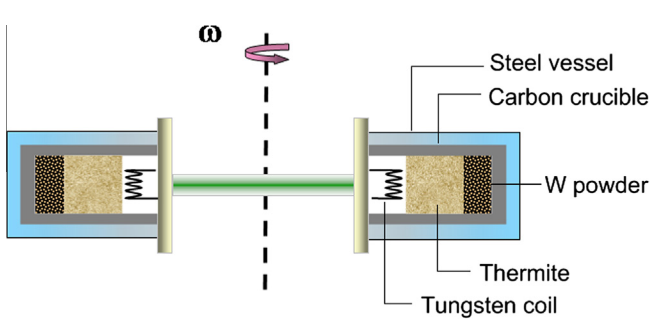

Fig. 1. A schematic illustration of the equipment for combustion synthesis and centrifugal infiltration.

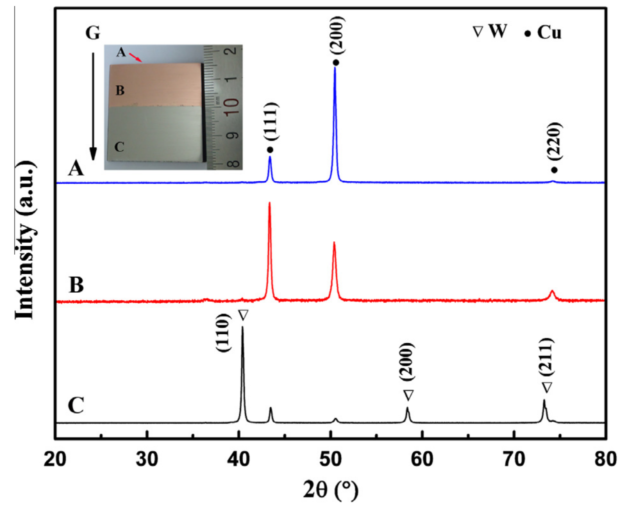

Fig. 2. XRD patterns of Cu/W–Cu integral material along high gravity direction: (A) transverse section of Cu layer, (B) longitudinal section of Cu layer and (C) longitudinal section of W–Cu layer.

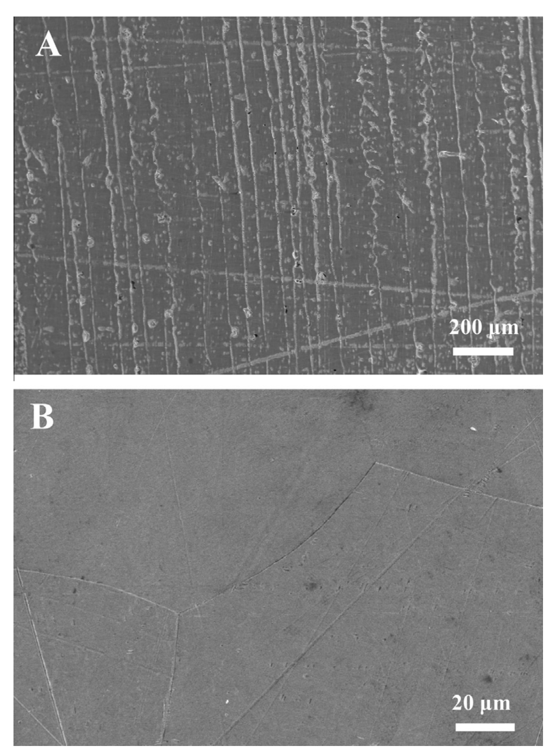

Fig. 3. SEM micrographs of etched Cu layer along high gravity direction: (A) longitudinal section and (B) transverse section.