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Letter

Fabrication of 60 wt%W–40 wt%Cu composite with crystal orientation by combustion synthesis and melt-infiltration in ultrahigh-gravity field

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ABSTRACT

60 wt%W–40 wt%Cu composite has been prepared by combustion synthesis and melt-infiltration in ultrahigh-gravity field. The 60 wt%W–40 wt%Cu composite is unadulterated and homogeneous. The Cu phase has crystal orientation, because lateral heat dissipation was avoided and the vertical temperature gradient was much higher than the horizontal gradient which could almost be neglected. Accordingly, all the coefficient of thermal expansion, thermal diffusion coefficient and compression strength of the 60 wt%W–40 wt%Cu parallel to gravity direction are different from that perpendicular to gravity direction due to the crystal orientation of Cu. Especially, the CTE of the prepared W–40Cu was lower than that of commercial W–40Cu, which is beneficial for W–Cu composites to be used as micro-electronic materials.

1. Introduction

Tungsten–copper (W–Cu) composites are promising materials for micro-electronic applications, like high voltage contact materials, due to the prominent thermal and electrical properties of copper, as well as the low vapor pressure (3.7 Pa at $T_{melting}$) and high melting point (3410 °C) of tungsten [1]. Nevertheless, the distinct difference in melting point and the insolubility between W and Cu make it difficult to fabricate W–Cu composites of high quality. Many results about fabricating W–Cu composites have been reported, including laser sintering [2], infiltration method [3,4], resistance sintering [5], mechanical alloying [6], liquid-phase sintering [7,8] and plasma spraying [9]. However, each method above has some disadvantages. For example, W–Cu composite fabricated by resistance sintering shows low density and low hardness, and the cost of hot pressed sintering is very high but the efficiency is low. Conventional infiltration method is a two-step process where firstly W skeleton is prepared by sintering process and then Cu melt is infiltrated into the W skeleton. It is a time-consuming and complex process, requiring high energy.

In this work, we developed a novel method of combustion synthesis and melt-infiltration in ultrahigh-gravity field (CSMI-UHG) to fabricate 60 wt%W–40 wt%Cu composite (W–40Cu). This method integrates combustion synthesis, centrifugal casting, and infiltration technique. Firstly designed combustion synthesis system ($2Al + 3CuO = 3Cu + Al_2O_3$) could produce a great amount of heat and Cu melt, and then the Cu melt is infiltrated into W powder bed in ultrahigh-gravity field produced by centrifugal force. By combustion synthesis in high-gravity field, Zhao et al. have prepared ceramic-lined pipes, TiC–TiB_2 and Al_2O_3/ZrO_2 composite ceramics with high density (~98%) and high bending strength (1060 MPa) [10,11]. Conventional infiltration method have been widely used, such as Al liquid infiltrating into SiC preforms, graphite particle preforms or titanium carbide preforms [12–14]. However, compared with conventional infiltration method, the CSMI-UHG method has merits of low cost, easy operation and high energy efficiency, for it needs no external heat sources and the energy comes from aluminothermic reaction. Furthermore, the whole CSMI-UHG process would be finished in five minutes due to the acceleration of the ultrahigh-gravity. In this paper, the microstructures, crystal orientation of Cu and relevant properties of the prepared W–40Cu were investigated.

2. Experimental procedure

Commercial powders of W and thermite (mainly composed of CuO and Al) were used as raw materials. W powders had a purity of >99.5% and an average particle size of ~3 μm. The particle size of CuO was 150–250 μm, the particle size of Al was 45–75 μm and the ratio of Al to CuO was 2/3. A batch of ~120 g W powders was cold-pressed into a 40 mm compact with a porosity of ~50% under a pressure of 10 MPa and loaded into a graphite crucible. Another 200 g thermite was

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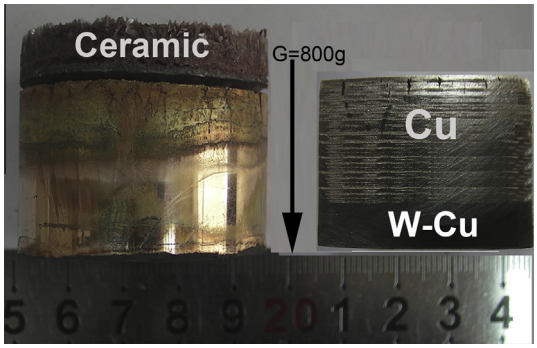

Fig. 1. Photographs of the products and the cross section of the W–Cu composite.

cold-pressed into a compact with a porosity of nearly 50% and placed above the W powders. Finally, a pressure head was placed on the top of the crucible. The crucible surrounded by SiO_2 as thermal insulator was placed in a chamber, which was evacuated to a vacuum of <300 Pa. An ultrahigh-gravity field was induced by centrifugation with an acceleration of 800g (G), where g means the gravitational acceleration. Then by passing an electric current through a tungsten coil closely above the thermite, combustion reaction ($2Al + 3CuO = 3Cu + Al_2O_3$, $\Delta H_{298} = -1209$ kJ/mol Al_2O_3 [15–17]) was triggered, and a large amount of heat as well as the products of ceramic (Al_2O_3) and Cu melt were produced. With the promotion of the ultrahigh-gravity, the ceramic separated with Cu melt due to density difference. Simultaneously, W powders sintered partially in such a high-temperature environment and was filled by the Cu melt. During the whole process, no external heating source was used. In five minutes, the process finished and bulk W–Cu composite of 40 mm diameter and 10.32 mm thickness (Fig. 1, direction of the arrow means high-gravity direction) was obtained.

The compositional content was identified by portable X-ray fluorescence analyzer (XRF; X-MET5100). The crystallographic phase was analyzed by X-ray diffraction (XRD; D8 Focus, Bruker, Germany). X-ray pole figure analysis was also performed using XRD. The microstructures were observed by scanning electron microscopy (SEM; S-3400, Hitachi, Japan) and transmission electron microscope (TEM; JEOL JEM-2100, Japan). Energy dispersive spectroscopy (EDS; INCA, Oxford Instrument, UK) was employed to characterize the purity of different sections of the prepared W–Cu composite. The coefficient of thermal expansion of the W–Cu composite was measured from 50 to 450 °C with a heating rate of 3 °C/min by using a DIL 402E thermal expansion instrument. The thermal diffusion coefficient was tested by laser thermal conductivity analyzer (LFA 457, Nestal, Germany).

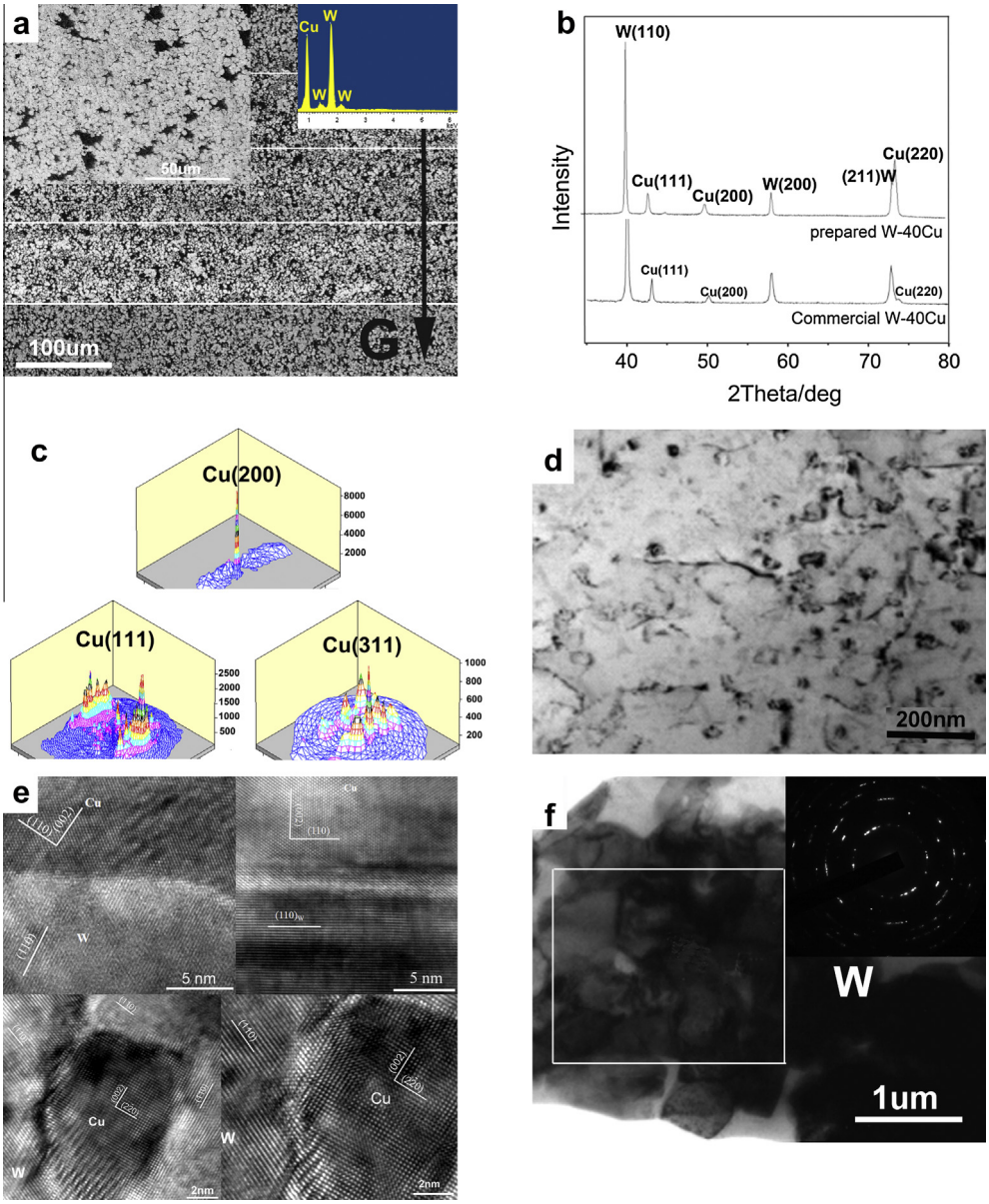

Fig. 2. (a) SEM photos coupled with EDS analysis result of the prepared W–40Cu along high-gravity field (b) XRD patterns of the prepared W–40Cu and commercial W–40Cu prepared by conventional infiltration (c) XRD pole figureof the Cu phase (d) TEM photos of dislocations in the Cu phase. (e) TEM high resolution photos of the interface between W and Cu. (f) Selected electron diffraction result of the W phase.