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W–15 wt%Cu nano-composite produced by hydrogen-reduction/sintering of WO_3–CuO nano-powder

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ABSTRACT

A two-stage hydrogen-reduction/sintering procedure was used to synthesize W-15 wt%Cu nano-composite tablets. Hydrogen-reduction was carried out at 600, 650, 700 and 750 °C for 15–90 min and sintering was performed at 1100, 1150, 1200 and 1250 °C for 60 min. Morphology and grain size of the products were rigorously investigated by scanning electron microscope (SEM), X-ray diffractometer (XRD) and nano-particle Zeta-sizer. Maximum consolidation of the nano-composite product was achieved at 1200 °C. Hydrogen-reduction at 700 °C for 90 min showed an average particle size of ~72.9 nm. Total reduction was achieved at higher temperatures and longer times. The mixture had a homogeneous structure with 16.1 ± 0.1 g/cm^3 density when sintered at 1200 °C for 60 min.

1. Introduction

W–Cu composites can be used in ultrahigh-voltage electric contactors, microelectronic devices, microwave blocking packages and high-density integrated-circuit heat-sinks [1–7]. Conductivity of the heat-sink is a significant parameter affected by homogeneity and compactness of the W–Cu composite article. Literature survey shows that homogenous W–Cu nano-composite powder can be produced by hydrogen-reduction of ball-milled WO_3–CuO powder at above 600 °C [7–18]. Dorfman et al. have produced homogenous W–Cu composite articles via sintering of hydrogen-reduced $CuWO_4$ without activators [19,20]. A desirable way of property improvement without addition of activating agent is to increase sintering capability by dramatic decreasing of the particle sizes of W–Cu composite articles.

The reduction paths are schematically shown in Fig. 1 [15]. According to the figure, hydrogen-reduction can occur via different routes. One route consists for example of: (a) WO_3 conversion into $WO_{2.9}$ and (b) reduction of $WO_{2.9}$ first into β-W and then into α-W. The temperature required for reduction of CuO is about 400 °C. WO_3 converts to $WO_{2.9}$ at 380 °C; while $WO_{2.9}$ reduces to W above 775 °C [16,17]. Previous authors have indicated that the characteristics of the hydrogen-reduced W–Cu powder depend on reduction temperature, holding-time, hydrogen-pressure and the powder bed-height [15–18]. Lowering of the temperature results in drastic lowering of the reduction rate which is governed by the following two general steps: (a) solid-state oxygen transport and (b) $WO_2(OH)_2$ chemical vapor transport (CVT) via the gas phase [16].

Previous studies have been confined to the isothermal tests performed at 750–1000 °C. This study considers the effect of reduction temperature and holding-time on consolidation of the reduced nano-powder and its structure. W–Cu nano-composite powder was produced by hydrogen-reduction/sintering of WO_3–CuO nano-mixture even at lower temperatures (600–750 °C). The results indicated that when the initial oxide mixture was nano-structured, the nano-composite product densified close to the theoretical density and its grains became superior. No additives (Co, Ni, Pd, etc.) were needed to obtain the required densification in this case.

2. Experimental procedure

Yellow tungsten oxide powder (YTO:WO_3) with mean particle size of 13 microns and purity of 99.97% (Wolfram Company JSC, Russia) and copper oxide (CuO) powder with mean particle size of 1.5 microns and purity of 99.9% (Merck, Germany) were ball-milled in 8000 Spex mixer/miller under stearic acid for 3 h. Nano-structured mixtures produced by milling were then reduced isothermally and non-isothermally by a flow of about 0.2 l/min gaseous hydrogen. The boats containing the mixture were made of recrystallized alumina. The boatload weighed ~1.5 g with a height of ~2 mm. Hydrogen-reduction was carried out at different temperatures: 400, 600, 650, 700 and 750 °C with purified hydro-

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