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Effect of ball-milling time on structural characteristics and densification behavior of W-Cu composite powder produced from WO_3-CuO powder mixtures

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ABSTRACT

Understanding the microstructure of W–Cu nanocomposite powder is essential for elucidating its sintering mechanism. In this study, the effect of milling time on the structural characteristics and densification behavior of W-Cu composite powders synthesized from WO_3-CuO powder mixtures was investigated. The mixture of WO_3 and CuO powders was ball-milled in a bead mill for 1 h and 10 h followed by reduction by heat-treating the mixture at 800 °C in H_2 atmosphere with a heating rate of 2 °C/min to produce W-Cu composite powder. The microstructure analysis of the reduced powder obtained by milling for 1 h revealed the formation of W–Cu powder consisting of W nanoparticle-attached Cu microparticles. However, Cu-coated W nanocomposite powder consisting of W nanoparticles coated with a Cu layer was formed when the mixture was milled for 10 h. Cu-coated W nanopowder exhibited an excellent sinterability not only in the solid-phase sintering stage (SPS) but also in the liquid-phase sintering stage (LPS). A high relative sintered density of 96.0% was obtained at 1050 °C with a full densification occurring on sintering the sample at 1100 °C. The 1 h-milled W-Cu powder exhibited a high sinterability only in the LPS stage to achieve a nearly full densification at 1200 °C.

1. Introduction

Recently, W–Cu alloys have attracted a growing interest owing to their superior thermal management properties and high microwave absorption capacities [1–3]. For most of the applications, high-density W–Cu composites exhibiting a homogeneous microstructure are needed to achieve a high performance. Since the W–Cu system exhibits mutual insolubility or negligible solubility [4], W–Cu powder compacts exhibit poor sinterability, even when liquid phase sintering (LPS) is performed above the melting point of the Cu phase [5]. Fine size and well mixing of the components are known to improve the sinterability of the powder, especially in a LPS system, such as the W–Cu system, in which particle rearrangement is known to be the dominant sintering mechanism.

The sinterability of W–Cu powder can be increased by employing an activated sintering process [7] with the addition of a small amount of metals such as Co, Ni, or Fe. However, since such activators can have a negative influence on the thermal management properties of the W–Cu alloy [8], enhancing the sinterability by activated sintering seems to be one of the least acceptable options. Recently, many efforts towards the fabrication and sintering of W–Cu powders with nano-sized grains have been carried out to achieve excellent densities without adding activators [9,10].

Among various methods used for producing highly dispersed W–Cu nanocomposite powders [11–20], the mechano-chemical process, which involves ball-milling of metal oxide powders and subsequent hydrogen reduction, is a promising approach as W–Cu nanocomposite powders exhibiting a high sinterability can be produced with this method [9,12].

Understanding the microstructure of W–Cu composite powder is essential for elucidating its sintering mechanism [6]. W–Cu nanocomposite powder obtained by performing hydrogen reduction of milled oxide mixtures exhibited similar characteristics as those of micro-homogeneously mixed W–Cu aggregates [21]. A new W–Cu nanocomposite structure, consisting of W nanoparticles coated with a Cu layer, was synthesized previously [22]. The product formation seemed to depend on various factors, including the reduction and milling conditions of the oxide mixtures. In this study, the effect of ball-milling time of WO_3-CuO powder mixture on the microstructural characteristics and densification behavior of reduced W-Cu composite powder is investigated.

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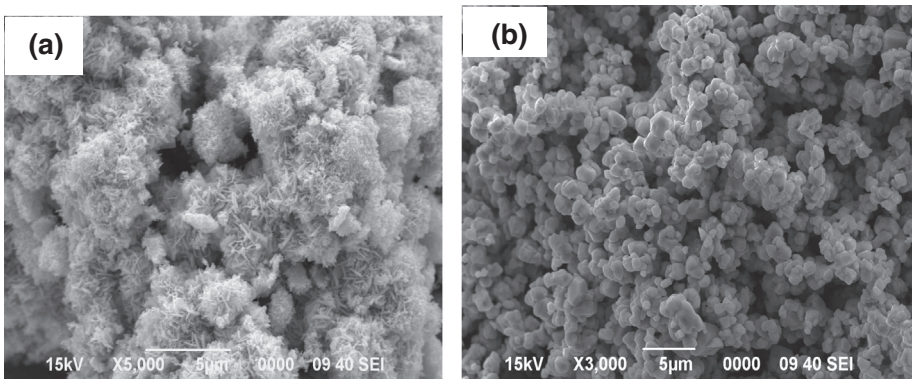

Fig. 1. FE-SEM images of the starting materials, (a) WO_3 and (b) CuO.

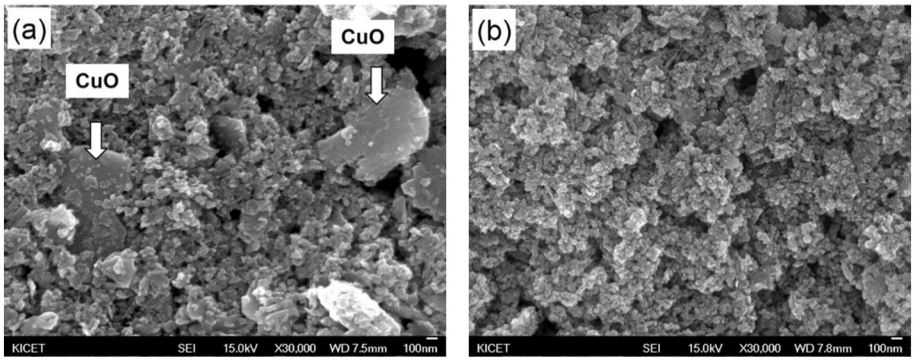

Fig. 2. FE-SEM images of the WO_3-CuO powder mixture ball-milled for (a) 1 h and (b) 10 h.

2. Experimental procedure

To produce W–20 wt.% Cu (W–20Cu) nanocomposite powder, WO_3 powder (Taegu Tec, Korea), with a mean particle size of 3 μm and a purity of 99.9%, and CuO powder (Kojundo, Japan), with a mean particle size of 12 μm and a purity of 99.9%, were used. Ball milling process was performed using a horizontal bead mill (UBM-5, Nanointek, Korea). ZrO_2 milling balls and jars were used to avoid contaminants, such as Fe, Cr, and Ni elements, which could act as sintering activators for W–Cu. More details on the milling experiment are described elsewhere [22]. The WO_3–CuO powder mixtures milled for 1 h and 10 h were used as starting materials in this study.

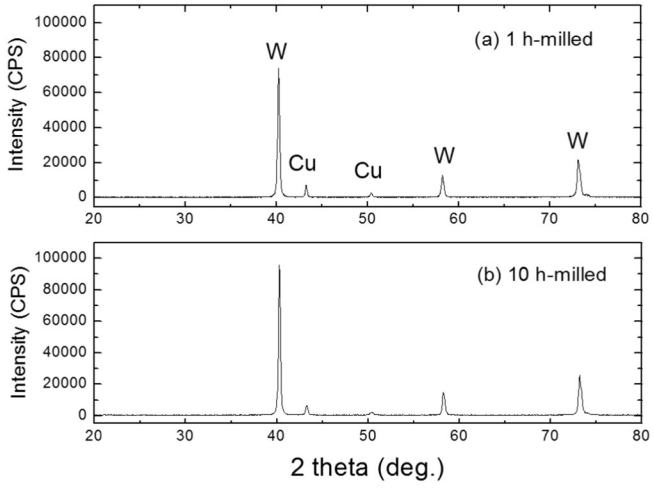

Fig. 3. XRD patterns of W-Cu powder reduced from WO_3-CuO powder mixture ball-milled for (a) 1 h and (b) 10 h in the bead mill.

The milled WO_3-CuO powder mixtures were dried at 80 °C and reduced in a 5 mm powder bed at 200 °C for 1 h. The temperature was then increased to 800 °C for 1 h in dry H_2 atmosphere (dew point of −76 °C) with a heating rate of 2 °C/min in order to form the W-20Cu composite powder. Microstructural analysis of the reduced W–Cu powder was conducted using field emission-scanning electron microscopy (FE-SEM, JSM-9701, JEOL, JAPAN) and field-emission transmission electron microscopy (FE-TEM, Tecnai G2 F30 S-Twin, FEI, Netherlands).

The reduced W–Cu composite powder was pressed at 100 MPa using the die compaction method, followed by performing cold isostatic pressing at 500 MPa in order to minimize the shrinkage anisotropy. The green density of the powder compact was found to be 45 ± 1% of the theoretically obtained value. Sintering was also conducted in the temperature range of 1050–1250 °C for 1 h, with a heating rate of 5 °C/min in H_2 atmosphere.

Density of the sintered sample was measured using the Archimedes' displacement method, and the sintered microstructure was examined using FE-SEM (JSM-9701, JEOL, JAPAN).

3. Results and discussion

Fig. 1 shows the FE-SEM images indicating the morphologies of the WO_3 and CuO powders, which were used as starting materials in this study. As shown in the figure, both the powders consist of micron-sized particles. WO_3 consists of agglomerates of needle shaped primary particle with ultra-fine size. The CuO powder consists of uniform micron-sized particles.

Fig. 2 shows the FE-SEM images of the WO_3-CuO powder mixture ball-milled for 1 h and 10 h in the bead mill. As shown in Fig. 2(a), the

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