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Comparison of W–Cu composite coatings fabricated by atmospheric and vacuum plasma spray processes

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ABSTRACT

Tungsten (W) coatings with different copper (Cu) contents (15 vol.% and 25 vol.%) were fabricated by atmospheric plasma spray (APS) and vacuum plasma spray (VPS) technologies, respectively. Phase composition and microstructure of the composite coatings were characterized. Physical properties, including porosity, oxygen content, microhardness and density of the coatings were examined. Thermal properties, including coefficient of thermal expansion (CTE) and thermal conductivity of the coatings were characterized. The results showed that the oxide content and porosity in the VPS-W/Cu coatings were apparently lower than those of the APS-W/Cu coatings. The CTE and thermal conductivity of the W–Cu composite coatings increased with the addition of Cu. The VPS-W/Cu coatings had apparently higher CTE and thermal conductivity compared with the APS-W/Cu coatings containing similar Cu content. The effects of the Cu incorporation in the composite coatings and the fabrication process on the coatings' properties were discussed.

1. Introduction

Tungsten–copper (W/Cu) composites, which combine the high thermal property of Cu and the low expansion and high melting point characters of W, have been developed and widely used over a wide range, such as in electronic packaging, electrical contacts, heat sinks and aerospace industry [1,2]. Liquid phase sintering is usually applied to fabricate W/Cu materials, which utilizes a sintered tungsten skeleton with molten Cu infiltrated [3–5]. However, grain coarsening is a common phenomenon in liquid phase sintering, which could result in shrinkage of the composites and lead to crack formation in the composite. Furthermore, the complicated processing steps of liquid phase sintering consume a lot of energy and cost.

In contrast to liquid phase sintering, plasma spray is a more convenient material fabricating technique, which combines the processes of melting, rapid solidification and consolidation into single operation [6]. During the plasma spray process, plasma flame has very high temperature (5000–10000 °C) which can melt almost all materials [7]. As a result, it possesses an obvious advantage in depositing refractory metals or alloys onto various substrates with complex shape or large surface area [8]. Kang et al. [9] and Itoh et al. [10] fabricated W/Cu composite coatings by atmospheric plasma spray (APS) technique and found that the surface oxidation of the deposited coatings occurred. It was further confirmed that the oxidation of W/Cu composite coatings could not be avoided under varied input powers [11]. Döring et al. [12] investigated the effect of powder conditions on the microstructure of the W/Cu composite coatings fabricated by vacuum plasma spray (VPS) technique and found that different powder conditions had a great effect on the porosity and therefore the thermal conductivity of the W/Cu composite coatings. Pintsuk [13] fabricated W/Cu functionally graded materials by the VPS technique, aiming to reduce the thermal stresses between the Cu substrate and top W coating. However, to the best of our knowledge, no studies have been done on the comparison between the W/Cu coatings prepared by APS and VPS technologies. And few researches have been carried out on the connection between the microstructure and chemical composition of the W/Cu composite coatings and their thermal properties.

In this work, W coatings with different Cu contents were fabricated by APS and VPS technologies, respectively. The

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microstructures and some basic properties of the composite coatings were comparatively studied. The influence of Cu content on the thermal properties, including coefficient of thermal expansion (CTE) and thermal conductivity of the W–Cu composite coatings, was investigated. The possible explanations on the differences of thermal properties between the APS-W/Cu and VPS-W/Cu coatings were discussed.

2. Experimental procedures

2.1. Coating preparation

Commercially available W powder (Zhuzhou Guangyuan Cemented Material Co., Ltd., China) with the medium size d_{50} of 37 μm and Cu powder (Zhuocheng Powder Co., Ltd., China) with the medium size d_{50} of 33 μm were used as feedstocks. W/Cu mixed powders with different contents of Cu (15 vol.%, and 25 vol.%, denoted as 85W/15Cu, and 75W/25Cu, respectively) were prepared by ball milling for 12 h in alcohol, then dried at 120 °C for 100 min. The coatings deposition was carried out by a plasma spray system (A-2000, Sulzer Metco, Switzerland) equipped with a F4-MB torch for APS and a F4-VB torch for VPS, respectively. Argon and hydrogen were used as plasma forming gases with a flow rate of 40 slpm and 10 slpm, respectively. Copper (OFHC) substrates were grit-blasted with alumina abrasive and cleaned with absolute ethyl alcohol prior to the plasma spray processes. The plasma sprayed coatings with a thickness of about 1 mm were deposited on the substrates.

2.2. Microstructure characterization

The surface and cross-sectional morphologies of coatings were investigated by scanning electron microscopy (SEM, EPMA-8705QH2, Shimadzu, Japan). The element composition of coatings was examined by energy dispersive spectrometer (EDS, INCA ENERGY, UK). Crystalline phases of coatings were analyzed by X-ray diffraction (XRD, RAX-10, Rigaku, Japan) operating with Cu Kα (λ = 1.5406 nm) radiation.

Microhardness measurements were also conducted on polished cross-sections of the coatings using a microhardness instrument (HX-100, SSOIF, China) under a load of 1.96 N and a dwell time of 15 s. The reported values of microhardness were the average of 20 point measurements randomly located along the cross-section. The oxygen content was characterized by nitrogen/oxygen analyzer (TC600, Leco, USA). The density and open porosity were measured according to the Archimedes' principle [14]. Three measurements were performed to determine the mean values of oxygen content, density and porosity for each coating.

The coefficient of thermal expansion was measured using a dilatometer (402ES-3, Netzsch, Germany) between room temperature and 300 °C. The measurements were performed with a heating rate of 2 °C/min under argon atmosphere. The coatings (1 mm × 4 mm × 12 mm) removed from substrates were used in the test. The thermal diffusivity (α) of the coatings was measured using laser-flash diffusivity method between room temperature and 200 °C under argon atmosphere with the free-standing coatings of Φ 10.2 mm × 1 mm. The specific heat capacity (Cp) was measured on the free-standing coatings of Φ 5 mm × 2 mm using a differential scanning calorimeter (DSC-2C, PE, USA) from room temperature to 200 °C. The measurements were performed with a heating rate of 10 °C/min in argon atmosphere with a flow rate of 0.05 slpm. A synthetic sapphire, NISTSRM 720, was used as the reference material.

The thermal conductivity (λ) was calculated using the measured specific heat capacity (C_p), thermal diffusivity (α) and the measured density (ρ) of the sample, as follows:

$$\lambda = \alpha \cdot C_p \cdot \rho \quad (1)$$

3. Results and discussion

3.1. Microstructure and composition characterization

Typical microstructures for the composite coatings are given in Figs. 1 and 2. Fig. 1 displays the SEI-SEM (secondary electron

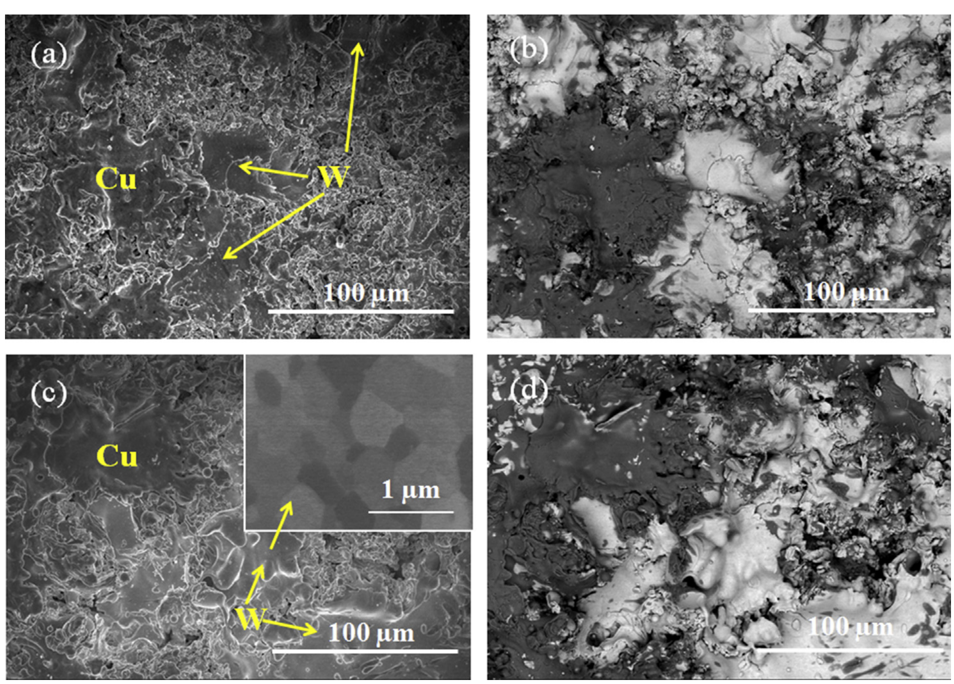

Fig. 1. SEI-SEM and BEI-SEM surface morphologies of W/Cu composite coatings: (a) and (b) APS-75W/25Cu, (c) and (d) VPS-75W/25Cu.