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Thermal conductivity of tungsten–copper composites

Sang Hyun Lee [a], Su Yong Kwon [a,*], Hye Jeong Ham [b]

[a] Division of Physical Metrology, Korea Research Institute of Standards and Science, 1 Doryong-dong, Yuseong-gu, Daejeon 305-340, Republic of Korea
[b] Kyungpook National University, Republic of Korea

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ABSTRACT

As the speed and degree of integration of semiconductor devices increases, more heat is generated, and the performance and lifetime of semiconductor devices depend on the dissipation of the generated heat. Tungsten–copper alloys have high electrical and thermal conductivities, low contact resistances, and low coefficients of thermal expansion, thus allowing them to be used as a shielding material for microwave packages, and heat sinks for high power integrated circuits (ICs). In this study, the thermal conductivity and thermal expansion of several types of tungsten–copper (W–Cu) composites are investigated, using compositions of 5–30 wt.% copper balanced with tungsten. The tungsten–copper powders were produced using the spray conversion method, and the W–Cu alloys were fabricated via the metal injection molding. The tungsten–copper composite particles were nanosized, and the thermal conductivity of the W–Cu alloys gradually decreases with temperature increases. The thermal conductivity of the W–30 wt.% Cu composite was 238 W/(m K) at room temperature.

1. Introduction

W–Cu composites have high electrical and thermal conductivities, low contact resistances, and low coefficients of thermal expansion. Due to these properties, W–Cu alloys are used in electrical contact materials, electrodes for spark erosion, shielding materials for microwave packages, and heat sinks for high power integrated circuits (ICs) [1–3]. W–Cu alloys are normally produced through powder metallurgy and are used widely. Kim [4] mechanically mixed tungsten and copper the powder using high-energy ball milling and sintered the powder at 1200 °C to create a composite material. When the thermal conductivity was measured at temperatures between room temperature and 1000 °C, it was seen that the thermal conductivity increased with temperature up to 500 °C, and decreased when temperature increased above this point. Hong and Kim [5] used nanopowders to produce a W–20 wt.% Cu composite and attained thermal conductivity of 239 W/(m K) at room temperature. Kang [6] used a powder-in-tube method to produce a W–Cu composite plate, which showed decreases in its thermal conductivity with increases in the tungsten content. There is an ongoing effort to create W–Cu composite materials that can use the high thermal conductivity of copper and low thermal expansion of tungsten. Furthermore, Jigui [7] used a powder injection molding method with commercially available tungsten and copper powder to produce W–20 wt.% Cu.

In this research, the spray conversion method was used with tungsten as the main material and copper added in nanopowder form with 5–30 wt.% copper through liquid-phase sintering [5]. The W–Cu nanocomposite powders are designed to have higher levels of hardness, water resistance, strength, and thermal conductivity. The powders are composed of spray drying of liquid sources, subsequent hydrogen reduction and carburization, which enhance the W–Cu particle refinement and homogeneous distribution of the tungsten matrix. The thermal diffusivity was measured using a laser flash method, and the specific heat capacity was measured using a differential scanning calorimeter (DSC). The thermal conductivity was calculated from the measured density, thermal diffusivity, and specific heat capacity. The thermal expansion was measured using a dilatometer, and a composition analysis was undertaken using an energy dispersive spectroscopy (EDS). Measurement of thermophysical properties was performed within the range from room temperature, which is the temperature at which W–Cu composites are commonly used, and 400 °C.

2. Experimental

2.1. Sample

Using the spray conversion method, tungsten salt $[(NH_4)_6(6H_2W_{12}O_{40})\cdot 4H_2O]$ and copper salt $[Cu(NO_3)_2\cdot 4H_2O]$ were dissolved in water, then spray dried at 250 °C and 11,000 rpm to produce the powder starting material. After producing the W–Cu oxidized powder, the powder was mechanically composited via high-energy ball milling. The powder was molded at

* Corresponding author.
E-mail address: kweonsy@kriss.re.kr (S.Y. Kwon).

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300 MPa and sintered at 1200–1400 °C. Tungsten was used as the primary material, and 5–30 wt.% copper was added. Two different forms were used for measuring the thermal diffusivity (diameter: 10 mm, thickness: 2 mm) and specific heat capacity (diameter: 5 mm, thickness: 2 mm), respectively.

2.2. Thermal diffusivity measurements

For the thermal diffusivity measurements, the laser flash method device (LF457, Netzsch, Germany) was used for measuring temperatures between room temperature and 400 °C. The thermal diffusivity was analyzed using the Cape and Lehman method [8], and it was calculated from the temperature at the back of the sample using mercury–cadmium–tellurium infrared sensor. The temperature was controlled using a tungsten mesh heater in a vacuum; in order to obtain consistent heating in the sample, the front and back of the sample was sprayed with graphite. The sample for the laser flash measurement was a disk of approximately 2 mm in thickness and 10 mm in diameter. The thermal diffusivity data were corrected using Azumi and Takahashi's method [9] to decrease the finite pulse effect. The mean value of the five measurements of thermal diffusivity was used to calculate thermal diffusivity. Furthermore, the uncertainty of the thermal diffusivity was estimated to be approximately ±5% [10].

The thermal conductivity (λ) was calculated using the measured thermal diffusivity (α), specific heat capacity at constant pressure (C_p), and the measured density (ρ) of the sample obtained through Eq. (1).

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

The density was measured using the Archimedes' principle. The sample was weighed in air (m_{air}) and then in water (m_{water}). The density was then calculated from the two weightings as follows:

$$\rho = \frac{m_{air}}{m_{air} - m_{water}} \cdot \rho_0 \quad (2)$$

where the density (ρ_0) is water at the given temperature.

2.3. Specific heat capacity measurements

The specific heat capacity (C_p) was measured using a differential scanning calorimeter (DSC 404C; Netzsch, Germany) at temperatures between room temperature and 400 °C. The measurements were performed with a heating rate of 10 K/min in a nitrogen atmosphere with a flow rate of 50 mL/min. A synthetic sapphire, NIST SRM 720, was used as the reference material. The specimens were cut into pieces with 5 mm diameters and 2 mm thicknesses. The uncertainty of the specific heat capacity was estimated to be ±2.0% [10].

2.4. Thermal expansion measurements

The thermal expansion was measured using a dilatometer (DIL 402; Netzsch, Germany) at temperatures between room temperature and 400 °C. The measurements were performed with a heating rate of 2 K/min in a nitrogen atmosphere with a flow rate of 50 mL/min; copper (NIST SRM 736L1) was used as the reference material. The specimens were cut into pieces with 6 mm diameters and 25 mm thicknesses. The uncertainty of the thermal expansion was estimated to be ±3.0%.

3. Results and discussion

The tungsten and copper content was analyzed using EDS. Table 1 shows the chemical composition of the five W–Cu composite samples. Copper with 5–30 wt.% was dispersed into the tungsten powder and sintered to fabricate the W–Cu composite. The theoretical density of the tungsten was 19.3 g/cm^3 and of copper was 8.96 g/cm^3 [11], but the actual density showed a slight deviation when the sample was sintered via powder mixing. The porosities of each sample due to sintering, calculated from the difference between the theoretical and measured density, were 2.3%, 2.3%, 7.4%, 4.6% and 6.8%. It should be noted that porosity has a large influence on the thermal conductivity.

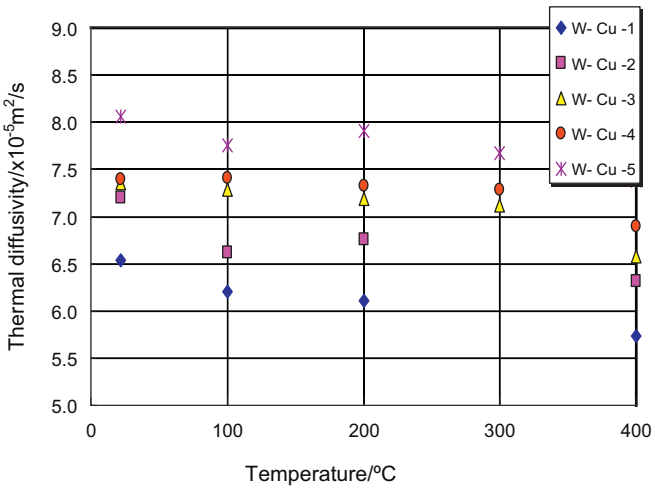

Fig. 1. Thermal diffusivity of the tungsten–copper composites.

Table 2 and Fig. 1 show the thermal diffusivity measurements. The tendency of the thermal diffusivity of WCu-1 and WCu-2 was to decrease with the temperature, but for the samples with a copper content of 16 wt.% or greater (WCu-3, WCu-4 and WCu-5), the thermal diffusivity slowly decreased with temperature increases. Furthermore, the thermal diffusivity increased when the copper content was higher.

Table 3 and Fig. 2 show the specific heat capacity measurements. The specific heat capacity of tungsten was 0.1320 J/(g K) and copper was 0.3728 J/(g K), demonstrating that the specific heat capacity increased with increases in the copper content or temperature. The specific heat capacity of WCu-1 and WCu-2, which had low copper contents, were similar at 0.1635 J/(g K) and 0.1689 J/(g K), respectively. The specific heat capacities of WCu-3, WCu-4 and WCu-5, which had high copper contents, were 0.1919 J/(g K), 0.1949 J/(g K) and 0.2134 J/(g K), respectively.

Table 4 and Fig. 3 show the thermal conductivity measurements of the W–Cu alloy with the recommended TPRC value [12].

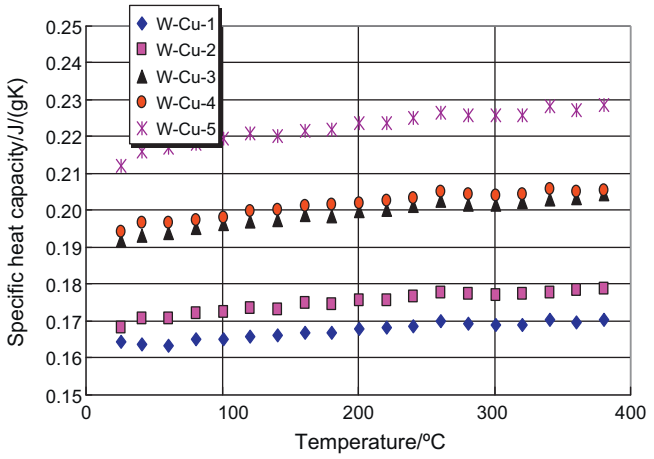

Fig. 2. Specific heat capacities of the tungsten–copper composites.