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Thermomechanical properties of AlN–Cu composite materials prepared by solid state processing

Kwang-Min Lee^{a,*}, Dae-Keun Oh^a, Woong-Sub Choi^a, T. Weissgärber^b, B. Kieback^b

^a Department of Materials Science and Engineering, Chonnam National University, Gwangju 500-757, South Korea ^b Department for Powder Metallurgy and Composite Materials, Fraunhofer Institute for Applied Materials Research, Dresden 01277, Germany

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Abstract

This study investigates coefficients of thermal expansion (CTE) and thermal conductivity (TC) of pulse electric current sintered mechanical alloying (MA) AlN–Cu powder compacts for the heat sink material application. The CTEs of MA AlN–Cu powder compacts showed good consistency with Turner's model when the volume fraction of copper was less than 60%, even in presence of some pores. When it was more than 60%, the values of thermal expansion had good agreement with the rule of mixtures. The porosity compensated TCs of AlN–40, 60, and 80 vol.% Cu measured at 200 °C were 62.7, 82.2, and 95.1 W m⁻¹ K⁻¹.

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1. Introduction

Microelectronic circuits require contact with a high thermal conductivity (TC), controlled low coefficients of thermal expansion (CTE) package materials to remove heat from circuits while avoiding stresses due to thermal cycling. The new heat dissipation materials must be thermally compatible with the semiconductor and substrate materials for high performance and reliability of the chip [1]. There are few published studies employing AlN–Cu system to improve thermal properties. AlN-Al [2], AlN-Mo [3], SiC-Al [4], and W-Cu [5] have been investigated for thermal management composites. The largely different melting points and the low wettability in those constituent elements could be drawbacks to applying such heat sink materials. Mechanical alloying (MA) techniques [6] can fabricate high quality composites via solid state processing from normally incompatible components. Therefore, this study investigates the CTE and the TC of pulse electric current sintered MA AlN-Cu powder compacts for heat sink material applications.

2. Experimental procedures

The compositions used were: AlN-40 vol.% Cu (AlN-64.5 wt.% Cu), AlN-60 vol.% Cu (AlN-80.3 wt.% Cu), and AlN-80 vol.% Cu (AlN-91.6 wt.% Cu). Copper powder has an average particle size of 25 µm with dendrite morphology. Aluminum nitride powder has a size of 60 µm with purity of 99%. MA was accomplished by using a planetary ball mill (Fritsch Pulverisette 5) with 250 cm³ capacity in an argon atmosphere at room temperature with a milling velocity of 200 rpm for 16 h. The ratio of stainless steel ball to powder weight was kept at 10:1. Stearic acid of 5 wt.% as a process control agent was added to the powder mixture. The MA processed AlN-Cu composite powders were consolidated by pulse electric current sintering (PECS) (Izumi Tech, Japan). The PECS processing parameters were 50 MPa of sintering pressure, 200 °C min⁻¹ of heating rate, 900 °C of sintering temperature, 5 min of sintering time, and 3.9 Pa of vacuum. Thermal expansion was measured using a thermomechanical analyzer (TMA7, Perkin-Elmer) up to 300 °C under N2 atmosphere with a heating rate of 5 °C min⁻¹. The thermal diffusivities of MA AlN-Cu powder compacts were obtained by laser flash apparatus (Theta Co., USA) at 20 °C, $250\,^\circ\text{C},$ and $400\,^\circ\text{C}$ under argon atmosphere. The data of thermal diffusivities were used to evaluate TC of PECSed AlN-Cu powder compacts. The dimensions of specimen were 20 mm in diameter and 1 mm in thickness.

3. Results and discussion

3.1. Coefficient of thermal expansion (CTE)

Fig. 1 shows linear thermal expansion $\Delta L/L$ of the PECSed AlN–40 vol.% Cu, AlN–60 vol.% Cu, and AlN–80 vol.% Cu

^{*} Corresponding author. Tel.: +82 62 530 1697; fax: +82 62 530 1699. *E-mail address:* kmlee@chonnam.ac.kr (K.-M. Lee).

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Fig. 1. Linear thermal expansion $\Delta L/L$ of AlN–Cu powder compact after PECS at 900 °C for 5 min: (a) 40 vol.% Cu; (b) 60 vol.% Cu; and (c) 80 vol.% Cu.

powder compacts. The CTE was evaluated from slope of linear expansion as follows [7]:

$$\frac{\Delta L}{L} = \alpha \,\Delta T \tag{1}$$

where *L* is the length of specimen, α the coefficient of thermal expansion, and *T* the absolute temperature. From the slope of



Fig. 2. Comparison of the measured and the theoretical coefficient of thermal expansion (CTE) of AlN–Cu powder compacts as functions of copper content.

straight line in Fig. 1, the CTEs for AlN–40 vol.% Cu (67% of theoretical density), AlN–60 vol.% Cu (74% of theoretical density), and AlN–80 vol.% Cu (83% of theoretical density) composites were 7.9 ppm/°C, 9.6 ppm/°C, and 14.5 ppm/°C, respectively. Thus, the CTEs of AlN–Cu composite materials had a very close relationship with the copper content.

The CTE of AlN–Cu composite materials can be simply represented by "rule of mixture":

$$\alpha = \alpha_{\rm AIN} f_{\rm AIN} + \alpha_{\rm Cu} f_{\rm Cu} \tag{2}$$

where α_{AIN} and α_{Cu} are the CTEs of aluminum nitride and copper, and f_{AIN} and f_{Cu} are the volume fractions of aluminum nitride and copper. Turner [8] predicted the CTE of composite materials as follows:

$$\alpha = \frac{\alpha_{\text{AIN}} f_{\text{AIN}} K_{\text{AIN}} + \alpha_{\text{Cu}} f_{\text{Cu}} K_{\text{Cu}}}{f_{\text{AIN}} K_{\text{AIN}} + f_{\text{Cu}} K_{\text{Cu}}}$$
(3)

where K_{AIN} and K_{Cu} are the bulk moduli of aluminum nitride and copper. Fig. 2 shows a comparison of the measured and the theoretical CTEs of AIN–Cu powder compacts as functions of copper content. As shown in this figure, the values of thermal expansion obtained in the present study had a good consistency with Turner model when the volume fraction of copper was less than 60%, even in presence of pores. When it was more than 60%, the values of thermal expansion had a good agreement with the rule of mixture. The variation of CTEs in AIN–Cu composite materials with copper content has a similar tendency in W–Cu composite materials [9].

3.2. Thermal conductivity

TC of PECSed AlN–Cu power compacts was evaluated by the laser flash method. The thermal diffusivity is given by the equation

$$D = \frac{0.1388L^2}{t_{1/2}} \tag{4}$$

Table 1 Thermal diffusivity of AlN–40 vol.% Cu, AlN–60 vol.% Cu, and AlN–80 vol.% Cu powder compacts measured by laser flash method at 25 °C, 200 °C, and 450 °C

$\overline{D(\mathrm{cm}^2/\mathrm{s})}$	25 °C	200 °C	450 °C
AlN-40 vol.% Cu	0.4050	0.06773	0.07509
AlN-60 vol.% Cu	0.10269	0.11512	0.13619
AlN-80 vol.% Cu	0.16578	0.18460	0.20747

where *D* is the thermal diffusivity (cm²/s), *L* the specimen thickness, and $t_{1/2}$ the time necessary for the back face of the specimen to reach one half of the maximum temperature. The thermal diffusivities of AlN–40, 60, and 80 vol.% Cu powder compacts measured at 25 °C, 200 °C, and 450 °C are presented in Table 1. The thermal diffusivity of AlN–Cu powder compacts increased as the copper content and testing temperature increased.

The TC of PECSed AlN–Cu power compacts was calculated according to the following relation [10,11]:

$$\sigma_0 = \kappa(\rho_{\rm AIN} C_{\rm AIN} f_{\rm AIN} + \rho_{\rm Cu} C_{\rm Cu} f_{\rm Cu}) \tag{5}$$

where σ_0 is the thermal conductivity, κ thermal diffusivity, ρ_{AIN} and ρ_{Cu} the densities of aluminum nitride and copper, C_{AIN} and C_{Cu} the specific heats of aluminum nitride and copper, and f_{AIN} and f_{Cu} the volume fractions of aluminum nitride and copper. The physical constants for calculation of thermal conductivity used in the present study are as follows: ρ_{AIN} is 3.26 g/cm^3 , ρ_{Cu} is 8.96 g/cm^3 , C_{AIN} is 0.74 J/(g K), and C_{Cu} is 0.386 J/(g K) [12,13]. The TCs of AlN–40, 60, and 80 vol.% Cu powder compacts calculated at 25 °C, 200 °C, and 450 °C are presented in Table 2. The TC of AlN–Cu alloy increased with increasing copper content. However, this includes effect on TC of reduced porosity because interruption of heat conduction at the interface between the residual pores and composite matrix.

The residual pores decrease the thermal conductivity and the decrement can be estimated as follows [14]:

$$\sigma = \sigma 0 \frac{1 - \xi}{1 + 11\xi^2} \tag{6}$$

where σ_0 is the porosity compensated TC, σ the TC with pores, and ξ the porosity. Fig. 3 shows porosity compensated TC of AlN-40 vol.% Cu, AlN-60 vol.% Cu, and AlN-80 vol.% Cu powder compacts as functions of temperature. The porosity compensated TCs of AlN-40, 60, and 80 vol.% Cu measured at 200 °C were 62.7, 82.2, and 95.1 W m⁻¹ K⁻¹.

Table 2

TC of AlN–40 vol.% Cu, AlN–60 vol.% Cu, and AlN–80 vol.% Cu powder compacts evaluated at 25 $^\circ$ C, 200 $^\circ$ C, and 450 $^\circ$ C

$\sigma_0 (Wm^{-1}K^{-1})$	25 °C	200 °C	450 °C
AlN-40 vol.% Cu	11.5	19.1	21.2
AlN-60 vol.% Cu	31.2	34.9	41.3
AlN-80 vol.% Cu	53.8	59.9	67.3



Fig. 3. Porosity compensated thermal conductivity (TC) of AlN–40 vol.% Cu, AlN–60 vol.% Cu, and AlN–80 vol.% Cu powder compacts as functions of temperature.

4. Conclusions

The CTEs for AlN–40 vol.% Cu, AlN–60 vol.% Cu, and AlN–80 vol.% Cu were 7.9, 9.6, and 14.5 ppm/°C, respectively. The CTEs showed a good consistency with Turner's model when the volume fraction of copper was less than 60%, even in presence of pores. When it was more than 60%, the values of thermal expansion were in agreement with the rule of mixtures. The TC of AlN–Cu composite materials measured by a laser flash method increased with increasing copper content and testing temperature. The porosity compensated TCs of AlN–40, 60, and 80 vol.% Cu measured at 200 °C were 62.7, 82.2, and 95.1 W m⁻¹ K⁻¹.

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