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Effect of metalloid silicon addition on densification, microstructure and thermal–physical properties of Al/diamond composites consolidated by spark plasma sintering

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ABSTRACT

Aluminum matrix composites reinforced with diamond particles were consolidated by spark plasma sintering. Metalloid silicon was added (Al–Si/diamond composites) to investigate the effect. Silicon addition promotes the formation of molten metal during the sintering to facilitate the densification and enhance the interfacial bonding. Meanwhile, the alloying metal matrix precipitates the eutectic-Si on the diamond surfaces acting as the transitional part to protect the improved interface during the cooling stage. The improved interface and precipitating eutectic-Si phase are mutually responsible for the optimized properties of the composites. In this study, for the Al–Si/diamond composite with 55 vol.% diamonds of 75 μm diameter, the thermal conductivity increased from 200 to 412 $\mathrm{Wm^{-1}\,K^{-1}}$, and the coefficient of thermal expansion (CTE) decreased from 8.9 to $7.3 \times 10^{-6}\ \mathrm{K^{-1}}$, compared to the Al/diamond composites. Accordingly, the residual plastic strain was 0.10×10^{-3} during the first cycle and rapidly became negligible during the second. Additionally, the measured CTE of the Al–Si/diamond composites was more conform to the Schapery's model.

1. Introduction

Advances in microelectronic technology have increasingly required developments in thermal management. Ideal thermal management materials should exhibit superior thermal conductivity and suitable coefficient of thermal expansion (CTE) to maximize heat dissipation and minimize thermal stress [1–3]. Because diamond has the highest thermal conductivity (1200–2000 $\mathrm{Wm^{-1}}$ -$\mathrm{K^{-1}}$) in nature and a low CTE ($1.0 \times 10^{-6}\ \mathrm{K^{-1}}$) [4,5], it is a desirable reinforcement for thermal transport. Meanwhile, aluminum possesses a relatively high thermal conductivity, low density and ease of processing. These characteristics make aluminum an ideal matrix material. Therefore, aluminum matrix composites reinforced with diamond particles (Al/diamond composites) have obtained increasing interest as the promising candidates for thermal management applications [2,4–18].

Currently, liquid methods, such as pressure infiltration [4–7,9,10,16–18], are often used to fabricate Al/diamond composites. But, because of the high temperature and long hours for fabrication, the degradation of diamond and severely interfacial reaction are inevitable. Spark plasma sintering (SPS) is an advanced sintering method that can be employed for rapid sintering at low temperatures. Moreover, spark plasma can purify and activate the particle surfaces to facilitate the interfacial optimization [19–21]. Therefore, SPS should be an effective method for fabricating Al/diamond with excellent properties. In previous works, uniform and dense Al/diamond composites have been fabricated by SPS. However, due to the weak interface, the obtained thermal conductivity was only 325 $\mathrm{Wm^{-1}\,K^{-1}}$ when using 50 vol.% diamond particles 70 μm in diameter [15]. Therefore, the interface must be optimized.

Alloying of metal matrix is a convenient method for improving the interface between the Al matrix and diamond. For instance, Wu [10] alloyed the matrix with copper (Cu) as the high thermally conductive element and reported the thermal conductivity was 330 $\mathrm{Wm^{-1}\,K^{-1}}$ with 65 vol.% diamond 150 μm in diameter, while the CTE decreased to $6.0 \times 10^{-6}\ \mathrm{K^{-1}}$. These improvements were ascribed to interfacial strengthening due to the precipitation of the Al_2Cu phase. Xue and Yu [9] introduced titanium (Ti) as carbide-forming element into Al/diamond composites. Because a layer of the TiC phase formed at the interface, the Al–Ti/diamond with 60 vol.% diamond 98 μm in diameter exhibited a thermal conductivity of 418 $\mathrm{Wm^{-1}\,K^{-1}}$. However, the unsatisfactory properties of

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the Al_2Cu and TiC phase limit further optimizations in the properties of the composites.

Metalloid silicon (Si) has the features standing between those of metal and ceramic, such as the mechanism of heat transfer and structure of valence electron. Simultaneously, its intrinsic thermal conductivity and mechanical properties are better than those of the above compounds. Therefore, Si can be a more effective element for acting on the interface [2,18,22,23]. It has been reported that Si addition can help improve interface for Al–Si/diamond composites prepared via SPS [13]; an enhanced thermal conductivity and reduced CTE were obtained. However, the effect of Si addition on the microstructure of the SPS sintered composites has not been investigated fully, and the mechanism responsible for the optimized thermo-physical properties remains unclear.

In the present work, Al/diamond composites with and without Si addition were fabricated using SPS. The effect of Si addition on the densification and microstructure of the composite was researched. We studied the thermal conductivity, CTE and thermal response curve for the composites. The correlation between the improvements in these thermo-physical properties and the microstructure of the metal/diamond interface was discussed.

2. Experimental procedures

2.1. Production of composites

Gas atomized Al powders (particle size 75 µm) with a 99.9 wt.% purity and Si powders (particle size 25 µm) with a 99.5 wt.% purity were used (Sinopharm Chemical Reagent Co., China). The reinforcement was synthetic monocrystalline diamond with the mean particle size of 75 µm (Henan Jinbei Special Type Diamond Co., China).

The matrix powders containing various diamond volume fractions (25%, 35%, 45%, and 55%) were dry mixed with a planetary mill (QM-QX4, China) at 300 rpm for 30 min. For the Al–Si/diamond composites, the content of Si addition was 2 wt.% in terms of the previous result [18]. Moreover, the matrix powders were mixed in two steps for better uniformity. First, the pure Al and Si powers were mixed in a 9:1 mass ratio in alcohol with the same mill at 500 rpm for 180 min. Afterward, these mixed powders and pure Al powders were blended to form the matrix powders.

The composites were fabricated using an SPS system (SPS-1050, Sumitomo Coal Mining Co., Japan) from 803 to 873 K under 40 MPa for 15 min. After cooling to room temperature, the composites were obtained and subsequently machined by the laser beam cutting. The upper graph of Fig. 1 displays the representative sintering curves. The longitudinal displacement of the punches was recorded over time to study the densification behavior of the composites.

2.2. Characterization

The microstructure of the composites was observed using field emission scanning electron microscopy (SEM, ZEISS SUPRA 55, Germany). Electron probe microanalysis (EPMA, JEOL JXA-8100, Japan) was used to characterize the elemental distribution along the metal/diamond interface. The sample density (ρ) was measured via Archimedes method using alcohol as the immersion medium, and the theoretical density was derived using the data listed in Table 1 [2,4,5,24–33] to obtain the relative density. The thermal diffusivity (a) was measured using a laser flash method (NETZSCH LFA427, Germany), and the specific heat (c) was determined using differential scanning calorimetry (DSC, NETZSCH STA 499 C, Germany). The thermal conductivity (K) can be calculated from $K = \alpha\rho c$. Dilatometer (NETZSCH DIL402C, Germany) was used for the thermal cycling test between room temperature and 623 K, acquiring the CTE and the response curve of thermal strain. To derive the thermal conductivity of the matrix, the static electrical resistance was measured using a four probe method (QUANTUM DESIGN PPMS-9, USA).

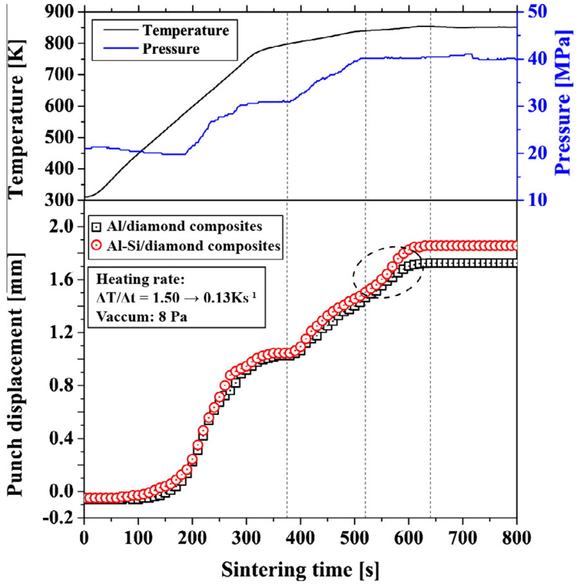

Fig. 1. Representative sintering curves and longitudinal displacement of the punches for the Al/diamond and Al–Si/diamond composites as a function of the sintering time.

3. Results and discussion

3.1. Densification behavior

Fig. 2 shows the relative densities of the composites with 55 vol.% diamonds relative to the sintering temperature. When the temperature is below 823 K, the Al/diamond and Al–Si/diamond composites have similar relative densities. However, when the temperature exceeds 823 K, the relative densities change differently. No dramatic variation in the relative density of Al/diamond composites is apparent; this value remains approximately 96.0%. In contrast, the relative density of Al–Si/diamond composites is continuously improved, reaching 99.1%.

At low sintering temperature, the densification of the composites primarily relies on the plastic deformation of the matrix powders [34]. Along with the reduction of yield strength, the densification is enhanced when the temperature increases. Consequently, these two types of composites with similar mechanical properties of the matrix achieve the same relative density. Nevertheless, because the space between the hard diamonds is filled by means of the deformation of matrix powders difficultly, the above mechanism cannot be wholly responsible for the densification.

Fortunately, another mechanism is initiated during the SPS sintering process when the temperature increases further. The plasma generated by the pulse current can help melt the powders when the temperature is below the melting point [15]. In this way, the molten matrix can fill the space between diamonds more easily, further enhancing the densification process. For the Al/diamond composites, to avoid failures in fabrication due to excessive meltdown, only the teeny surfaces of the Al powders allow to be melted. A small amount of molten metal can be produced with insufficiently enhancing the densification. Therefore, the relative density of the Al/diamond composite was not dramatically increased with further increase in the temperature, as shown in