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Effect of particle size on the microstructure and thermal conductivity of Al/diamond composites prepared by spark plasma sintering

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Abstract

Spark plasma sintering (SPS) was used to fabricate Al/diamond composites. The influence of diamond particle size on the microstructure and thermal conductivity (TC) of composites was investigated by combining experimental results with model prediction. The results show that both composites with 40 μm particles and 70 μm particles exhibit high density and good TC, and the composite with 70 μm particles indicates an excellent TC of 325 $W \cdot m^{-1} \cdot K^{-1}$. Their TCs lay between the theoretical estimated bounds. In contrast, the composite with 100 μm particles demonstrates low density as well as poor TC due to its high porosity and weak interfacial bonding. Its TC is even considerably less than the lower bound of the predicted value. Using larger diamond particles can further enhance thermal conductive performance only based on the premise that highly dense composites of strong interfacial bonding can be obtained.

Keywords: metal matrix composites; thermal properties; sintering; particle size

1. Introduction

The miniaturization of electronic devices has been leading to a significant increase in power density, and there is a need for materials with TC exceeding 250-300 $W \cdot m^{-1} \cdot K^{-1}$ [1]. Yet the main thermal management materials such as Al(Cu)/SiC_P composites [2-3] have a limit on their TC and can not keep up with the dramatic progress in heat sink materials and electronic packages. Diamond is known to have the highest TC, in the range of 1200-2000 $W \cdot m^{-1} \cdot K^{-1}$ [4]. Embedding diamond particles in a metal matrix such as Al, Cu or Ag provides a way to dissipate heat to the maximal degree [5]. Compared to Cu or Ag, Al is selected as a matrix material due to its relatively high TC and ease of processing, as well as its low density and low cost. For this reason, Al/diamond composites are considered to have a bright developing prospect.

Nevertheless, until now, only a limited number of studies on the Al/diamond composites have been reported. So far, Al/diamond composites with high diamond volume fraction can only be produced by infiltration of molten aluminum into diamond preforms or tap-packed diamond powder beds [6-7]. However, this processing route bears the risk of Al_4C_3 formation, which can act as a thermal diffusion barrier at the interface [8]. Recently, spark plasma sintering (SPS) was invented as a rapid solidification processing method. It has been found that composites of uniform, dense and good thermal conduction can be readily fabricated by SPS [9-11]. Therefore, as a supplement to the infiltration method, SPS is expected to synthesize Al/diamond composites with excellent thermal conduction property.

In this study, SPS was used to fabricate Al/diamond composites. The influence of diamond particle size on the microstructure and thermal conductivity of composites was investigated by combining experimental results with model prediction.

2. Experimental

2.1. Composites preparation

The aluminum powder used was 200 mesh in size and 99.9% in purity. The selected reinforcement diamond particles were synthetic MBD4 diamonds with mean sizes of 40, 70, and 100 μm, respectively. The aluminum powder and 50 vol.% diamond particles of different sizes were dry mixed by using a vibratory mill for 2.5 h, respectively. The vibration frequency of the ball mill was 1400 r/min. The milling balls were not used due to the ultra-high hardness of diamond particles which could wear the surface of the balls and bring impurities from the balls into the powders during the mixing process.

The SPS-1050 system was used to synthesize

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Al/diamond composites. The mixed powders were packed into a graphite die (10 mm in diameter). The powder compacts were sintered at 550°C under a fixed uniaxial pressure of 30 MPa in vacuum (less than 4 Pa). The sintering process is shown in Fig. 1. Samples 1#, 2#, and 3# represent the as-sintered composites with particle sizes of 40, 70 and 100 μm, respectively.

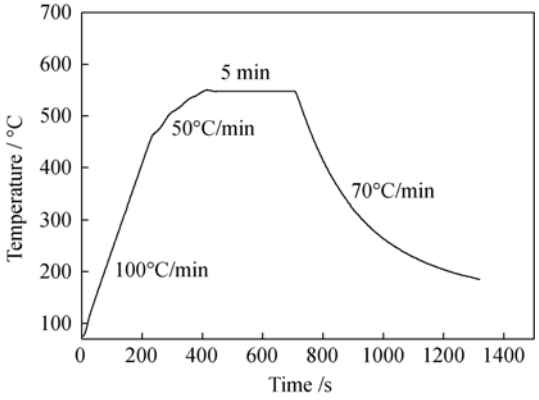

Fig. 1. **SPS process of preparing Al/diamond composites.**

2.2. Characterization

The bulk density of the composites was measured by Archimedes' principle. Fracture surface observation was used to investigate the microstructure of composites by scanning electron microscopy (SEM). Thermal diffusivity and specific heat were measured by the laser flash method and calorimetric techniques, respectively. Thermal conductivity was calculated as the product of density, thermal diffusivity and specific heat.

3. Results and discussion

3.1. Microstructure

Fig. 2 shows the microstructures of fracture surfaces of the composites with various particle sizes. In the samples 1# and 2 # (Figs. 2(a-b)), it can be found that in both, diamond particles are closely embedded into the Al matrix. Ductile dimples and the tear ridges of the Al matrix are distributed along the fracture surface. This phenomenon presents a powerful interfacial bonding between the particles and the Al matrix, which is even stronger than the toughness of the Al matrix. Few bare diamond particles can be seen under this condition. In contrast, in the case of sample 3#, cracks emerge at the Al-diamond interface and most of diamond particles are bare on the fracture surfaces (Fig. 2(c)), demonstrating weak interfacial bonding where the Al matrix is not uniformly close adhesion to the entire diamond surface. More seriously, some diamond particles could even scale off from the matrix. The significant difference in microstructures between samples 1#, 2#, and 3# can be explained as follows.

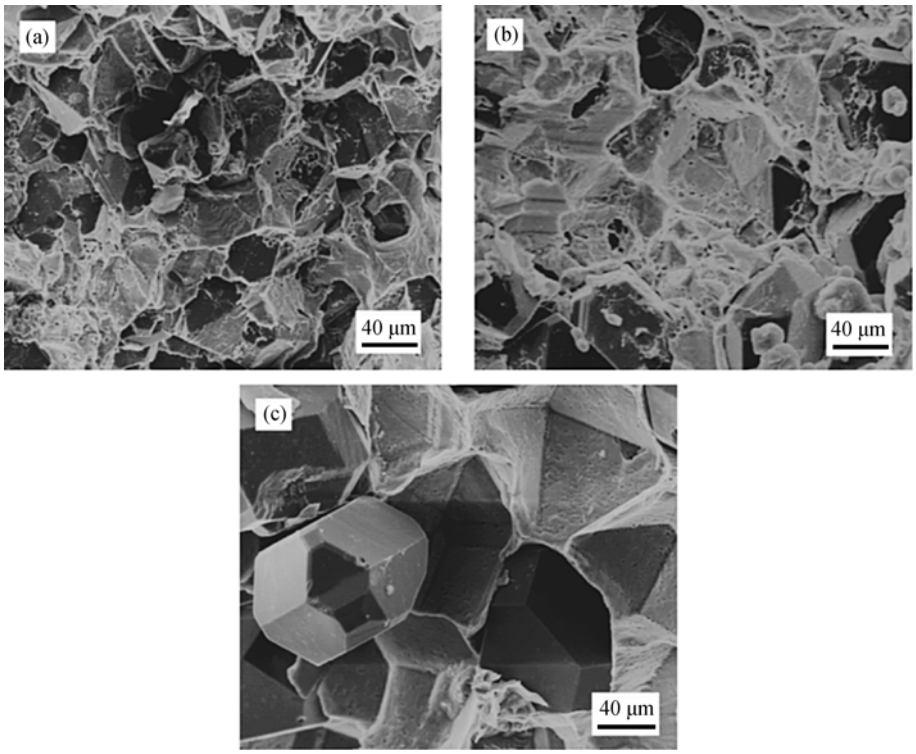

Fig. 2. Microstructures of fractures surfaces of the composites with various particle sizes: (a) 40 μm; (b) 70 μm; (c) 100 μm.

At the beginning of the SPS, due to the high fraction of diamond particles in the mixture powders, the applied uniaxial pressure can press diamond particles to be closely in contact with each other and locate Al powders in confined gaps among the diamond particles. Diamond gaps depend on particle size, and larger particles have larger gaps. Under this condition, if the gaps are very large it can be considered that the distribution of Al powders into the diamonds' gaps cannot be uniform. Pores can eventually form in some area with less Al (Fig. 3(a)). In addition, due to the considerably large difference in coefficient of thermal expansion (CTE) between diamond (2.3×10^{-6}) and Al (23.6×10^{-6}), a large thermal stress can be generated at the interface of the composites during the sintering process. Thermal stress exhibits as tensile stress on the diamond particles in the cooling process [2]. Thus, the gaps filled with large amounts of Al have a greater possibility to concentrate large residue tensile stress, which can make diamond particles debond from the Al matrix in the temperature declining process (Fig. 3(b)). While relatively small gaps can make Al evenly distribute among the diamonds' gaps, and the generated residue tensile stress is also very weak in the interspaces with less Al filling, this cannot be tough enough to result in Al departing from the diamond particles.

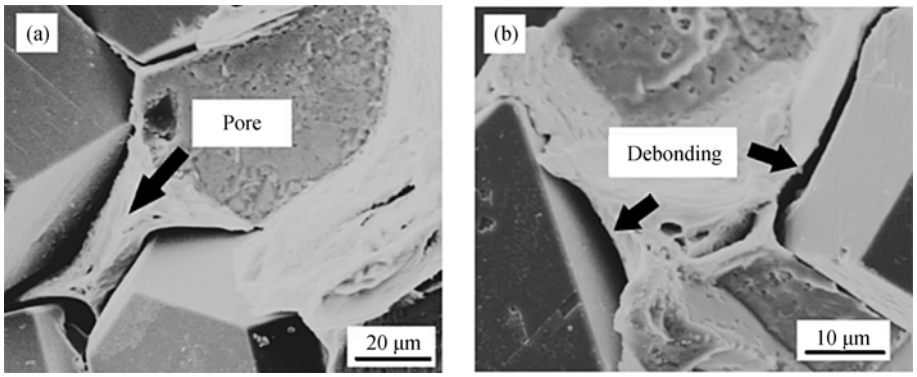

Fig. 3. Characteristics of fracture surfaces for composite with 100 μm particles: (a) uneven distribution of Al; (b) interfacial debonding.

3.2. Thermal conductivity

Fig. 4 indicates the TC and relative density (RD) of all specimens. It can be seen that the RDs of samples 1# and 2# are nearly the same, which exceed 97%, but the RD of sample 3# is only 93% due to its high porosity. Besides this, it is obviously seen that there is a sharp decrease in TC from 325 $W \cdot m^{-1} \cdot K^{-1}$ of sample 2# to 196 $W \cdot m^{-1} \cdot K^{-1}$ of sample 3#. The TC of sample 3# is even lower than that of pure Al, as a result of its high porosity and weak interfacial bonding. Pores can severely degrade the TC of the composite due to the fact that they can scatter the heat-carriers, such as electrons from Al and phonons from diamond. Weak interfacial bonding leads to a large interface thermal resistance (ITR), which can greatly deteriorate the thermal conduction performance of the composites.

In addition, the TC of sample 1# is lower than that of sample 2# within nearly the same RD. This is due to the fact that the composite with a smaller size of particles has more interfaces than that with a larger size of particles. More interfaces bring higher ITR to decrease the TC of the composite, and this result is in accordance with the spirit of the Hasselman-Johnson (H-J) model [12-14], according to the following equation:

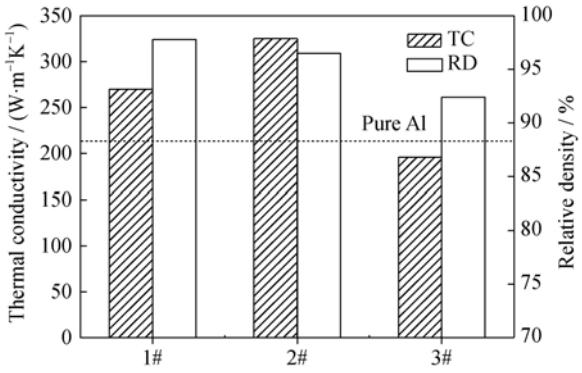

Fig. 4. Thermal conductivity (TC) and relative density (RD) of all specimens: 1#, 2# and 3# represents samples with particle sizes of 40, 70 and 100 μm, respectively; the dash line is the TC of Al matrix used in our experiments.

$$K_c = K_m \frac{2(K_p/K_m - K_p/ha - 1)V_p + K_p/K_m + 2K_p/ha + 2}{(1 - K_p/K_m + K_p/ha)V_P + K_p/K_m + 2K_p/ha + 2} \quad (1)$$

where K_m, K_p, and K_c are the TC of the matrix, reinforced particles and composite, respectively. V_p and a represent the volume fraction and radius of dispersed particles, respectively. $1/h$ is the value of the ITR.

Therefore, the H-J model can be used to estimate the TC of the metal matrix composites reinforced by particles. Nevertheless, the H-J model has an assumption of sphere particles with isotropic ITR, which is not very suitable for diamond particles with totally different ITRs in their distinct crystal surfaces. Flaquer *et al.* [15] and followed Chu *et al.* [16-17] took the shape of particles into account, and gave a more accurate model for TC prediction of composites with diamond particles based on the H-J model:

$$\begin{cases} \dfrac{1}{h} = \dfrac{1}{h_{001}S_{001} + h_{111}(1-S_{111})} & \text{(Upper bound)} \\ \dfrac{1}{h} = \dfrac{S_{001}}{h_{001}} + \dfrac{1-S_{111}}{h_{111}} & \text{(Lower bound)} \end{cases} \tag{2}$$

where h_{001} and h_{111} are the intrinsic interface thermal conduction in the diamond {001} plane and the diamond {111} plane, which are equal to 1.0×10^8 $W \cdot m^{-2} \cdot K^{-1}$ and 1.0×10^7 $W \cdot m^{-2} \cdot K^{-1}$, respectively [11]. S_{001} is the percentage of {001} planes over the entire surface of the diamonds. As the diamond particles' shape used in our work is typical cubo-octahedral, S_{001} can be taken as 1/3.

If the TC of diamond particles K_p = 1250 $W \cdot m^{-1} \cdot K^{-1}$ (provided by the manufacturer), the Al-matrix K_m = 210 $W \cdot m^{-1} \cdot K^{-1}$ (measured in our experiment), and other relative calculated parameters mentioned above are substituted into Eqs. (1) and (2); the TC range of composites investigated in our study can be evaluated by the model predictions of the upper and lower bounds, as summarized in Table 1. It can be found that the TCs of samples 1# and 2# with high RD and strong interfacial bonding lie between the theoretical bounds, whereas the TC of sample 3# with low RD and weak interfacial bonding is far below the predicted lower bound. It is clear that the model predictions are in accordance with experimental results, which confirms that the modified H-J model containing the inhomogeneous interfacial resistance is sufficiently accurate to give a prediction for the thermal conductivity of Al/diamond composites.

Table 1. TC prediction bounds of samples

Sample No.	Particle size / μm	Experimental data / ($W \cdot m^{-1} \cdot K^{-1}$)	Upper bound / ($W \cdot m^{-1} \cdot K^{-1}$)	Lower bound / ($W \cdot m^{-1} \cdot K^{-1}$)
1#	40	270	348	265
2#	70	325	396	319
3#	100	196	420	354

4. Conclusions

The influence of particle size on the microstructure and thermal conductivity (TC) of SPS consolidated Al/diamond composites was investigated and the following conclusions were obtained. Both the Al/diamond composites with 40 μm diamond particles and with 70 μm diamond particles show high density and good TC, particularly, the composites with 70 μm particles indicate excellent TC, which is as high as 325 $W \cdot m^{-1} \cdot K^{-1}$. The TCs lie between the theoretical estimation bounds. The composite with 100 μm diamond particles demonstrates low density and poor TC due to the combined effects of uneven Al distribution among the particles' gaps and weak interfacial bonding caused by large residue tensile stress during the SPS cooling process. Its TC is dramatically less than the predicted value. Therefore, only by ensuring the preparation of the composites with high density and good interfacial bonding, can the use of larger particle further improve the TC of the composites.

Acknowledgements

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