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Research paper



Development of Graphene Reinforced Metal Matrix Composite by Spark Plasma Sintering

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Abstract

In order to obtain the high performance materials with high thermal conductivity, high electrical conductivity, low thermal expansion, good mechanical properties and low density, Graphene has higher thermal conductivity comparison with other ceramic particle. In this study, graphene dispersed aluminum (Al) composites was developed by spark plasma sintering. Volume fraction of graphene were 10, 20 and 30 vol.%. Fabrication conditions of graphene dispersed aluminum (Al) composites were temperature of 813K and applied pressure of 80 MPa. As composite properties are affected by the dispersibility and volume fraction of the graphene particles, the relationship among the dispersibility of dispersant and the thermal conductivity and mechanical properties was investigated.

Keywords: Metal matrix composite, Graphene, Sintering, Microstructure, Manufacturing process

1. Introduction

Due to the increased market of intelligent car, the semiconductor component is widely used. Under working condition, semiconductor component generates heat, which may lead to overheat and thermal stress $[\tilde{I}, 2]$. To improve the satiability and save energy semiconductor component, the requirement of heatsink material for semiconductor component is as high as thermal conductivity (TC), low coefficient of thermal expansion (CTE) and low density ^[3, 4]. And workability of the heat sink material is also required for the complex shape ^[3]. The graphene has outstanding thermal property and ease of machining. And aluminum has a low density. Therefore, the graphene dispersed aluminum (graphene/Al) composite is a promising candidate for heat sink material. In this study, mechanical alloying method was utilized to fabricate composite power. Then the composite powder was sintered by Spark plasma sintering (SPS) method to fabricate graphene/Al composite. As composite properties are affected by the dispersibility and volume fraction of the graphene particles, the relationship among the dispersibility of dispersant, the microstructures and interface of graphene/Al composite were observed. And the thermal conductivity and mechanical properties of graphene/Al composite were investigated.

2. Experimental Procedure:

We used graphene powder with diameter of 0.6 μ m, thickness of 9.5 nm and carbon purity of >95% (Graphene Platform Co.) as reinforcement. And pure aluminum powders with diameter of 3

µm, purity of 99.9% were used as matrix. The key steps of graphene/Al composite powders were: (1) sonication of aluminum powder and graphene; (2) mixing aluminum powder and graphene for mechanical alloying and (3) SPS sintering by SPS method. In particular, aluminum powders were added 10 vol.%, 20 vol.% and 30 vol.% of graphene, then were conducted with sonication (4.2 kHz) for 3.6 ks. Next, the zirconia balls to mixture ratio of 10:1 were mixed by V-type mixing with 45 RPM for 43.2 ks or by planetary ball milling with 150 RPM for 3.6 ks. The mixtures were pre-sintered with condition of pressure 15 MPa, pulse current 100 A, pulse voltage 50 V, pulse width 100 ms for 600 s. Then the mixtures were sintered at 813 K and pressure of 80 MPa by SPS method. The microstructures and interfaces of graphene/Al composite were observed by scanning electron microscope (SEM) and field emission scanning electron microscope (FE- SEM). Electron probe micro-analyzer (EPMA) was conducted for analyzing element distribution. The densities of graphene/Al composite were tested by Archimedes method. X-Ray Diffraction (XRD) measurements were conducted for phase analysis. And the thermal conductivities and mechanical properties of graphene/Al composite were investigated by steady state method [5] and Vickers hardness test, respectively.



3. Results and Discussion:

3.1. Observation of composite powders



Fig. 1: SEM images of mixed powder: (a) V-type mixing and (b) by planetary ball milling.

SEM images of composite powders mixed by V-type mixing and planetary ball milling are shown as Fig. 1. As shown in Fig. 1 (a), the powders were squashed with almost no agglomerating. While comparing with composite powders in Fig. 1. (a), the powders fabricated by planetary ball milling were agglomerated in complex shapes. The reason was planetary ball milling proved more energy, then leading repeated shear fracture of the powders. By the impaction of balls, the graphene powers were inserted into aluminum powders. Therefore, the aluminum powders and graphene powders, as composite powders, were mechanical alloyed planetary ball milling. Thus, the graphene/Al composites were fabricated by planetary ball milled composite powder.

3.2. Microstructures and Phase Analysis of Graphene/Al Composites

Fig. 2 shows the microstructures of graphene/Al composites fabricated with different volume fractions of graphene powders. The light gray parts were graphene and the dark gray parts were aluminum. As shown in Fig. 2, the graphene phases as reinforcement were easily to connect to each other with increasing volume fractions of graphene powders, but the pores as defects also increased. Moreover, the relative density of graphene/Al composite with 10 vol.%, 20 vol.% and 30 vol.% graphene was 98.2%, 95.2% and 93.2%, respectively. Fig. 3 shows the interface of aluminum and graphene. The thickness of graphene was about 220 nm for easy agglomeration of graphene powders ^[6]. Defects in 10 vol.% graphene/Al composite and its element distribution are shown as Fig. 4. As in Fig. 4 (a) and (b), the defects were formed by agglomerated graphene in composite. In Fig. 4 (b), there were no diffusion of carbon in aluminum matrix. Fig. 5 is X-ray diffraction patterns of 10 vol.% graphene/Al composite. Fig. 5 shows only the diffraction peaks of Al and graphite. No evidence of Al_4C_3 indicates that this phase was not formed in the graphene/Al composite.



Fig. 2: SEM images of graphene/Al composite with different volume fractions: (a), (b) 10 vol.%; (c), (d) 20vol.% and (e), (f) 30vol.%.





Fig. 4: Defects in 10 vol.% graphene/Al composite and its element distribution: (a) SEM image, (b) EPMA images.



Fig. 5: X-ray diffraction patterns of10 vol.% graphene/Al composite.

3.3. Thermal Conductivities and Hardness of Graphene/Al Composites

The TCs of pure aluminum sintered with same conditions and graphene/Al composites are shown as Fig. 6 (a). The TCs of graphene/Al composites were lower than the pure aluminum for low relative density ^[7] and thermal resistance. But, the TC of graphene/Al composites with 20 vol.% graphene was higher than graphene/Al composites with 10 vol.% graphene. The reason was graphene formed a continuous phase in the composite, then reduced the effect of thermal resistance. The Vickers hardness of pure aluminum sintered with same conditions and graphene/Al composites are shown as Fig. 6 (b). The Vickers hardness of graphene/Al composite with 10 vol.% graphene was improved than -



pure aluminum for reinforcement of graphene^[8]. However, the Vickers hardness of graphene/Al composites were decreased with increasing volume fraction of graphene for decreasing relative density.

4. Conclusions

Graphene and Al composite powders were fabricated by mechanical alloying method and composite powders was fabricated by spark plasma sintering process. The main conclusions are as folow:

- Aluminum powders and graphene powders, as composite powders, were mechanical alloyed planetary ball milling. Graphene powders was attached on the aluminum power surface.
- (2) Defects were formed by agglomerated graphene. And the defects were increased with increasing volume fraction of graphene.
- (3) The TCs of graphene/Al composites were lower than pure aluminum for defects in composite and thermal resistance at interface. However, under condition of 20 vol.% graphene, TC was increased by continuous graphene.
- (4) The Vickers hardness of graphene/Al composites were decreased with increasing volume fraction of graphene for decreasing relative density.

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References

- Zweben C., "Advances in composite materials for thermal management in electronic packaging", *Journal of the Minerals*. Vol.50, No.6, (1998), pp. 47-51, available online: https://link.springer.com/article/10.1007/s11837-998-0128-6.
- [2] S. Mallik, N. Ekere, C. Best & R. Bhatti, "Investigation of thermal management materials for automotive electronic control units" *Applied Thermal Engineering*, Vol.31, No.2-3, (2011), pp.: 355-362, available online: https://www.sciencedirect.com/ science/ article/ pii/ S1359431110004199.
- [3] X.H. Qu, L. Zhang, M. Wu & S.B. Ren., "Review of metal matrix composites with high thermal conductivity for thermal management applications." *Progress in Natural Science: Materials International*, Vol.21, No.3, (2011), pp.: 189-197, available online: https: //www.sciencedirect.com/science/article/pii/S100200711260029X.
- [4] P.K. Schelling, L. Shi & K.E. Goodson, "Managing heat for electronics" *Materials Today*, Vol.8, No.6, (2005), pp.: 30-35, available online: https: //www.sciencedirect.com /science /article /pii /S1369702105709354.
- [5] K. Sugio, Y. B. Choi & G. Sasaki, "Effect of the Interfacial Thermal Resistance on Effective Thermal Conductivity of Al/SiC Particle-Dispersed Composites", *Materials Science Forum*, Vol. 879, (2017), pp. :1889-1894, available online: https://www.scientific.net /MSF.879.1889.
- [6] C. Zhou, G. Ji, Z. Chen, M. Wang, A. Addad, D. Schryvers & H. Wang, "Fabrication, interface characterization and modeling of oriented graphite flakes/Si/Al composites for thermal management applications" *Materials & Design*, Vol.63, (2014), pp: 719-728, available online: https: //www.sciencedirect.com /science/ article/ pii/ S0261306914005366.
- [7] S. Ren, X. He, X. Qu & Y. Li, Ren, "Effect of controlled interfacial reaction on the microstructure and properties of the SiCp/Al composites prepared by pressureless infiltration" *Journal of Alloys and Compounds*, Vol.455, No.1-2, (2008), pp: 424-431, available online:

https://www.sciencedirect.com/science/article/pii/S0925838807003 295.

[8] Fang, M., Wang, K., Lu, H., Yang, Y., & Nutt, S, "Covalent polymer functionalization of graphene nanosheets and mechanical properties of composites" *Journal of Materials Chemistry*, Vol.19, No.38, (2009), pp.: 7098-7105, available online: http://pubs.rsc.org /en/content/articlehtml/2009/jm/b908220d.