Proceedings of the 12th International Conference on
Aluminium Alloys, September 5-9, 2010, Yokohama, Japan

pp. 873-877

Effect of Microstructure on Thermal Properties for VGCF/ Aluminum Composites fabricated by Spark Sintering Process

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10 vol. % vapor grown carbon fiber (VGCF) dispersed pure aluminum matrix composites was fabricated by spark plasma sintering (SPS) method. By using some aluminum powders with different diameter (1-30μm) as starting material, the effects of microstructure on Vickers hardness, tensile strength, thermal conductivity and thermal expansion were investigated. As increasing a diameter of aluminum powder as starting material, VGCFs became to coagulate during aluminum power particle. Vickers hardness of monolithic aluminum block was enhanced by addition of VGCFs. As decreasing the size of aluminum powder, the hardness of the composites became to improve dramatically. Whereas, the tensile strength of composites were lower than that of monolithic aluminum block due to a week bonding at VGCF/Al matrix interface and VGCF/VGCF interface. The thermal conductivities of composites are 150-175W/m·K, and the obvious dependence of powder particle diameter was not observed. On the other hand, the thermal expansion of composites decreased as decreasing a diameter of aluminum powder due to the sliding between VGCFs.

Keywords: *VGCF, aluminum, composites, thermal conductivity, spark plasma sintering*

1. Introduction

As the problems on heat generation of semiconductor and IC have been developed recently, the demand of metal matrix composites with good thermal conductivity and low thermal expansion has been increasing. Usually, matrix used in practical use for composites is pure aluminum and copper. The dispersive material in metal is SiC powder, but it is hard material and has relative low thermal conductivity. In actual, carbon materials seem to be suitable for the dispersive materials, because carbon materials have good formability and thermal conductivity. Unfortunately, carbon materials are low wettablity for molten aluminum and have high reactivity. Carbon materials react with aluminum to produce the needle kike aluminum carbide (Al_4C_3), which degrades the mechanical and functional properties of the composites. The reactivity of carbon materials to aluminum is completely different for the surface structure of carbon materials. Basal plane of carbon without defects and impurity have low reactivity for aluminum [1]. Vapor grown carbon fiber (VGCF) seems to be suitable for the dispersive materials for composites because of perfect surface structure [2]. Furthermore, VGCF has good mechanical and thermal conductivity, low thermal expansion and high cost-performance compared with other carbon materials.

In this study, in order to suppress the reaction between carbon and aluminum and obtain high performance composites, VGCF and spark plasma sintering process was used as starting material and manufacturing process, respectively. Spark plasma sintering (SPS) is suitable process for the preparation of dense composites at lower temperature. On the other hand, well dispersed VGCF/ Al composites have low electrical conductivity because of strain in matrix introduced by the difference of thermal expansion between VGCF and aluminum [3]. The same damage is considered for the thermal conductivity. Thereby, the effect of size of starting aluminum powder on the microstructure and properties such as the mechanical and functional properties in this study.

2. Experimental procedure

Starting materials for matrix were three kinds of aluminum powder with 1μm, 3μm and 30μm in average diameter and 99.9 % in purity, and dispersing materials was VGCF with 150 nm in average diameter and 10-20 μm in length, which is produced by SHOWA DENKO Inc. Japan. At first, VGCF was stirred in ethanol with ultrasonic vibration for 30 min in order to sleave the aggregation of VGCF and disperse uniformly. Then, aluminum powders was poured in VGCF containing ethanol, and mixed for 60 min by ultrasonic vibration. Volume fraction of VGCF used in this study was 10 vol. %. This mixed powder with ethanol put in aluminum can with alumina boll and boll-milled for 12h. Finally it was dried in atmosphere. The mixed powder was sintered by SPS method under the condition of 823K in sintering temperature, 50 MPa in applied pressure, 1.0-1.5 A/m^2 in applied current density and under 10^{-2} Pa in vacuum. After sintering, we can obtain the composites with over 99.7 % in relative density. In order to compare with monolithic alloy, aluminum block was fabricated by SPS in same conditions. Vickers hardness and tensile strength was measured in order to estimate the mechanical properties, which were measured by Vickers hardness tester with the load of 500g and the holding time of 15s, and tensile testing machine with cross head speed of $2.0x10^{-3}s^{-1}$, respectively. Thermal expansion and thermal conductivity (TC) at room temperature was measured in order to estimate the functional properties. TC of samples was measured by the laser flash thermal contacting measuring apparatus (TC-700, ULVAC-RIKO Inc., Japan) at room temperature. The coefficient of thermal expansion (CTE) of the samples was measured by the dilatometer (DIL 402C, NETZSCH inc., Germany) Furthermore, the microstructure was observed by optical microscopy and scanning electron microscopy (SEM).

3. Results and Discussion

Fig. 1 shows the microstructure of composites fabricated by aluminum powder with different average diameter of 1μm, 3μm and 30μm (1μm /Al composite, 3μm /Al composite, 30μm /Al composite). In 1μm /Al composite, VGCF are dispersed uniformly in al matrix without obvious aggregation. The composite has fine and isotropic structure. As increasing the diameter of aluminum powder, VGCF became to aggregate. In 30μm /Al composite, this tendency was emphasized, and VGCF aggregates shows a preferential orientation, which results from the deformation of aluminum powders under uniaxial pressure during the hot-pressing in SPS.

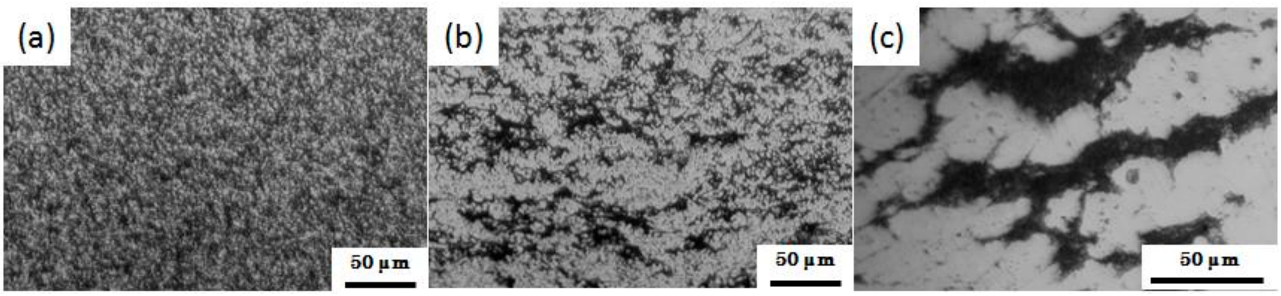

Fig. 1 SEM images of 10vol% VGCF/Al composites with average diameter of (a) 1 μm Al powders, (c) 3 μm Al powders and (d)30 μm Al powders, respectively.

Fig. 2 shows the comparison of Vickers hardness of the composites and Al blocks fabricated by different Al powders. As decreasing the size of Al powders, both of the corresponding composites and Al blocks possess higher hardness, which is attributed to the grain refinement of Al matrix. Vickers hardness of monolithic aluminum block was enhanced by addition of VGCFs. It is caused by the high hardness of VGCF and the arrest of grain growth for aluminum by well dispersed VGCF.

Fig.3 shows the tensile strength of VGCF/Al composites and Al blocks fabricated by different size of Al powders. Fig. 4 shows stress-strain curves of VGCF/Al composites fabricated by different size of Al powders. The strength increased with decreasing the size of Al powders, which is caused by the grain refinement of Al matrix. Morever, the oxide film on Al powder particles could affect to the interfacial bonding between Al powder particles. On the othrehand, the strength of the composites were lower than that of the corresponding Al blocks. The tensile strength of the composites is usally influenced by the interfacial bonding between the fiber and matrix. As increasing the size of Al powder in composites, the strein is decreased dramaticallly. The lower strength of he composites can be explained by weak bonding at the interface between VGCF and Al matrix because of the poor wettability bewteen VGCF and Al. The decrement rate of the composite strength for Al block increased as increasing the size of aluminum powder. Espacially, the the decrement of 30μm /Al composite is remarkable. The decrement rate of the composite strength is closely related to the dispersion of VGCF in Al matrix. For 30μm /Al composite, obvious aggregation of VGCFs was observed in Al matrix.

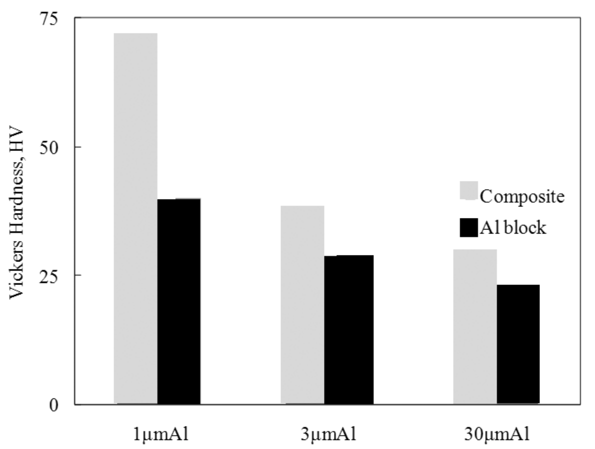

Fig. 2 Vickers hardness of VGCF/Al composites and Al block fabricated by Al powders with different diameter

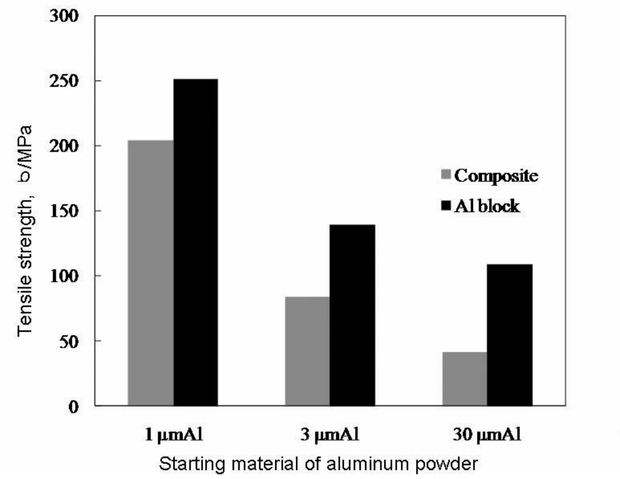

Fig. 3 Tensile strength of VGCF/Al composites and Al block fabricated by Al powders with different diameter

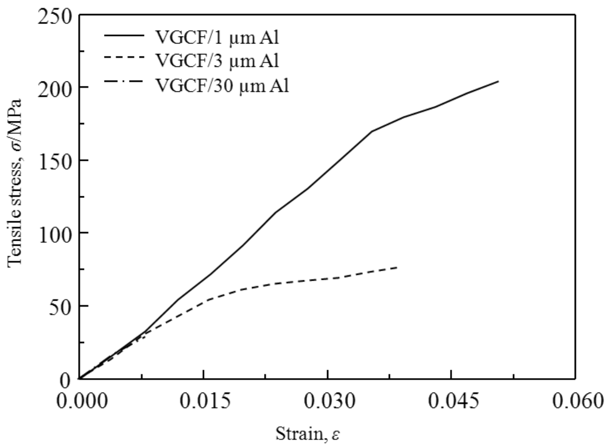

Fig. 4 Stress-strain curves of VGCF/Al composites fabricated by different size of Al powders.

Fig.5 shows the fracture surface obtained from tensile strength for the composites. For 1μm /Al composite, ductile structure of Al matrix was observed mainly. It seems the compsite strength is related by Al matrix, strongly. Furthermore, , as single VGCF was observed around ductile ftacture, VGCFs is seem to de-bond from Al matrix. For 3μm /Al composite, both of the uniformly dispersed VGCF and aggregated VGCF were observed in fracture surface. For 30μm /Al composite, only the aggregated VFCFs were observed. It seems fracture was occured in aggragated VGCF. From these result, it was concluded that the decrement rate of tensile strength of the composites depend on the quality of VGCF aggregaretes in th ecomposites, strongly.

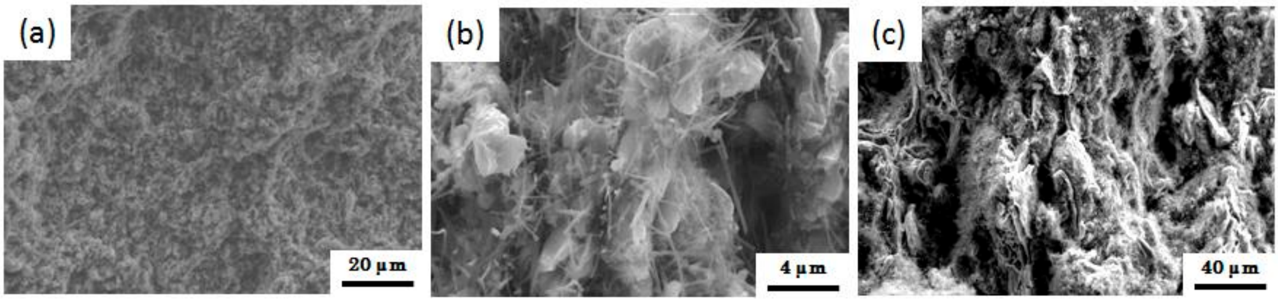

Fig. 5 SEM images of fracture surface for 10vol% VGCF/Al composites with average diameter of (a) 1 μm Al powders, (c) 3 μm Al powders and (d)30 μm Al powders, respectively.

Fig. 6 shows TC of 3 μm Al block and 1μm /Al, 3μm /Al, 30μm /Al composites. TC of Al block was lower than the standered value of Al (273 W/m·K). It seems the small amount of the oxde layer bewteen Al particles degrade TC of composite in spite of the brakedown of oxide layer by the spark by SPS. TC of the composites were higher than that of Al block. It is contributed to the reinforcement of VGCF in composites, which is 150-175 W/m·K. Diffreence of TC for the difference of powder size was small. Although TC of the composites were reduced remarkably than the theoretical value of 1200 W/m·K and 237 W/m·K for VGCF and aluminum, respectively. It is cause by the thermal resistance at VGCF/Al and VGCF/VGCF interfaces. As increasing the particle size of Al, the number of VGCF/Al interface decreases, and the number of VGCF/VGCF intreface increases. Cosequently, it is considered that the difference of TC for particle size keep the barance for low TC.

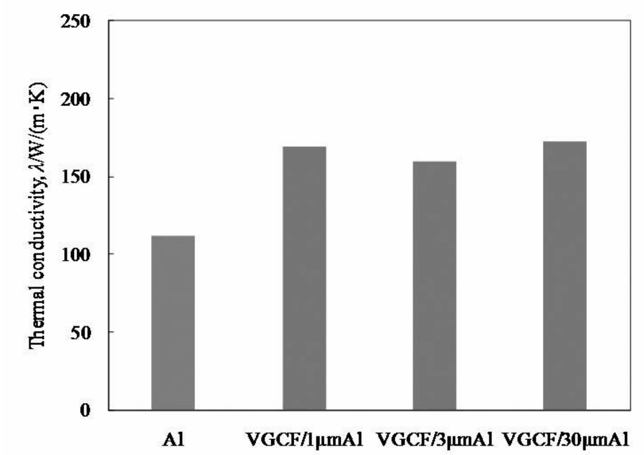

Fig. 6 Thermal conductivities for Al and VGCF/Al fabricated by different diameter Al powders at room temperture.

Fig. 7 shows CTE of the composites and Al blocks. CTE of the composites is higher than the theoretical value ($21.1 x10^{-6}$/K) calculated from the rule of mixture. The high CTE of the composites is caused by the weak interfacial bonding of VGCF/Al and VGCF/VGCF interfaces. For 1μm /Al composite and 3μm /Al composite, CTEs are a little smaller than that of Al blocks. As VGCFs in these composites were dispersed uniformly (or partially uniform) in Al matrix, the expanding of Al matrix by heating was suppressed, and thus leading to smaller CTEs. However, CTE of 30μm /Al composite is higher than that of Al block. VGCFs in this composites were aggregated at the interface between Al particles in the composites, resulting VGCFs was difficult to suppress the expanding of Al matrix because of the sliding between VGCFs.

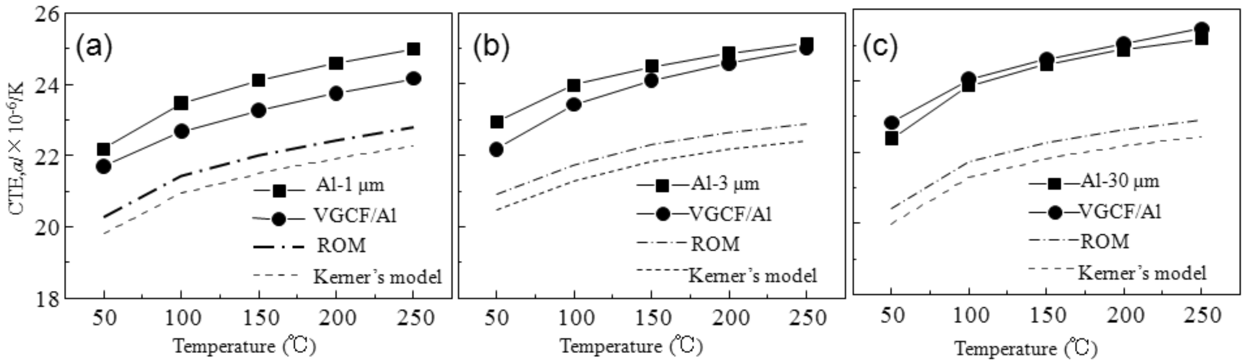

Fig. 7 Coefficient of thermal expansion of VGCF/Al composites and Al block fabricated by (a)1μm Al powder, (b) 3μm Al powder and (c) 30μm Al powder.

4. Conclusion

In order to obtain the material with high thermal conductivity and low thermal expansion, VGCF/Al composites was fabricated by SPS in order to suppress interfacial reaction. In order to estimate the effects of microstructure on the mechanical and thermal properties, 10 vol % VGCF dispersed pure aluminum matrix composites was fabricated using by different aluminum particle sized aluminum as starting material. Followings are our conclusions;

(1) Vickers hardness of monolithic aluminum block was enhanced by addition of VGCFs. As decreasing the size of aluminum powder, the hardness became to improve dramatically.

(2) The tensile strength of composites were lower than that of monolithic aluminum block due to a week bonding at VGCF/Al matrix interface and VGCF/VGCF interface.

(3) TC of composites is 150-175W/m·K, and the obvious dependence of powder particle diameter was not observed.

(4) CTE of composites decreased as decreasing a diameter of aluminum powder due to the sliding between VGCFs.

Acknowledgements

This study was supported by the light metal educational foundation in Japan and Grant-in-Aid for Scientific Research (c) (21560771) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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