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EFFECT OF TiB_2 CONTENTS ON MICROSTRUCTURE AND THERMAL CONDUCTIVITY OF Al MATRIX COMPOSITES PREPARED BY SPARK PLASMA SINTERING

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ABSTRACT

Pure aluminum (Al) matrix composites with changing the volume fraction of TiB_2 particle content from 10 to 30% was fabricated by spark plasma sintering (SPS) process. Then the microstructure, density, dispersibility and thermal conductivity of the composites was estimated. As increasing TiB_2 content, the dispersibility of particles and the density of the composites degraded slightly. And, the effect of the dispersibility of TiB_2 particles on the thermal conductivity was quite little. The thermal conductivity of the composites was less than the theoretical value calculated by Maxwell-Eucken equation considering the effect of the density. It is due the influence of the interface heat transfer rate, because these composites have many small sized particles and have high interface density.

KEYWORDS

Titanium diboride, Dispersibility, Thermal conductivity, Pure aluminum, Tensile strength

INTRODUCTION

Recently, metal matrix composites (MMCs) have been used widely in electric / electronic components, architectural materials, transportation equipment, aerospace and a precision instruments, because it's superior properties have been greatly watched. Demand of MMC for industrial use has been increasing gradually. Furthermore, multifunction properties are required for the composites strongly. By integrating the dissimilar materials, the composites have various properties that we cannot get from monolithic materials. On the other hand, the response to the thermal management is required for materials, recently, and the need for the structural material controlling the thermal conductivity has been increasing. Especially, superior thermal conductivity and good mechanical properties from room temperature to high temperature are required for heat sink and heat exchanger at the same time. In late years, a study on the composites combining carbon materials and aluminum and copper has been developing, and actually the composites with high thermal conductivity which is not provided in the conventional materials is developed. On the other hand, non-oxide ceramics particles such as SiC and AlN, TiB_2 have good thermal conductivity. Particularly, as TiB_2 has good electrical conductivity, TiB_2 containing MMCs is expected as innovative materials used as electric/ electronic components. Furthermore, as TiB_2 has low density, high melting point, low thermal expansion and good deformation resistance, the development of high performance composites with multi-functional properties is expected. As thermal conductivity of pure Al and TiB_2 are 225 and 66.4 W/mK, respectively, it is expected that the degradation of thermal conductivity by adding dispersant in Al matrix is suppressed. The authors have carried out a thermal conductivity, electrical conductivity, mechanical properties of 20 vol.% TiB_2 dispersed Al composites (Sugio, 2016, Sasaki, 2016). In this study, the influence of quantity of addition of TiB_2 on microstructure, thermal conductivity was studied minutely.

EXPERIMENTAL PROCEDURE

Starting materials used are pure aluminum powders with 3 μm in average diameter and 99.9% in purity and TiB_2 powders with 2.62 μm in average diameter, which were served by Kojundo Chemical Laboratory Co., Ltd. and New Metals Co., Ltd. in Japan, respectively. After measuring the volume fraction of TiB_2 as 10, 20, and 30 vol.%, pure Al and TiB_2 powders were mixed in ethanol with alumina ball in V shape type mixer, which condition was 50 rpm in rotation speed for 16 h in mixing time. After mixing, ethanol was removed by filter, and then dried at 120°C for 2 days by drying machine. Mixed powder was sintered by spark plasma sintering (SPS), and then, TiB_2 particle dispersed Al matrix composites were obtained. After having pulse electric discharge with 0.6 ks interval, the mixed powder was sintered under the condition of 500 MPa in applied pressure, 793–873 K in temperature, vacuum under 10^{-2} Pa and over 0.6 ks in keeping time. The density of the composites was measured by the Archimedes method, and its microstructure was observed by scanning electron microscopy (SEM). The dispersibility of the TiB_2 particles in composites was evaluated quantitatively by localized number in 2 dimension method (LN2DR) (Sugio, 2007) and SEM image. LN2DR is defined as the number of bary center of particle included in measuring circle, which is expressed following equation:

$$\frac{7}{\pi R_{2D}{}^{2}} = \lambda_A \quad \Rightarrow \quad R_{2D} = \left(\frac{7}{\pi \lambda_A}\right)^{1/2} = \frac{1.493}{\lambda_A{}^{1/2}}$$

where, R_{2D} is the radius of the measuring circle, and λ_A is the number density.

Generally, as the number of LN2DR decreases, the dispersibility of dispersant increases. The theoretical value of uniform distribution of particles is increasing as decreasing the volume fraction of particles in composites such as 0, 4.5 and 3.0 of LN2DR for 0, 10 and 20 vol.% particles in composites.

The particle size distribution was measured by image analysis software. Thermal conductivity was measured by steady state method. After crippling the composites by hot plate and cold plate, a thermocouple was set in three places in composites. The steady state was obtained by heating a hot plate and cooling a cold plate. Then, temperature was measured at 1 s intervals for 300 s in steady state by each thermocouple.

RESULTS AND DISCUSSION

Relative density of composites increased as increasing sintering time, but the sintering ability degraded as increasing TiB_2 contents. When sintering is 1 h, the relative densities of 10 and 30 vol.% TiB_2/Al composites were 95 and 85%, respectively. By increasing sintering time to 4.5 h, the composites of 94.4% in relative density was obtained. The increasing of TiB_2 content in composites brought a slowdown of densification speed, but the dense composites was obtained by the increasing of sintering time.

Figure 1 shows the microstructure of 10, 20 and 30 vol.% TiB_2/Al composites. It seems that TiB_2 particles was relatively dispersed well but have some aggregation of particles here and there. Then, the dispersibility of TiB_2 particles in composites was estimated by the value of LN2DR. Figure 2 shows the relationship between TiB_2 content in composites and LN2DR. As the volume fraction of particles increased, the value of LN2DR indicating the uniform distribution showed smaller numbers. 4.5, 3.0 and 2.0 are theoretical number of uniformly dispersed number with random for 10, 20, and 30% of TiB_2 contents, respectively. But the experimental values were 5.12, 4.37, 4.17, and it means a tendency to heterogeneity dispersion of particles was observed. The experimental value tends to deviate from the theoretical value of uniform dispersion as increasing TiB_2 contents.

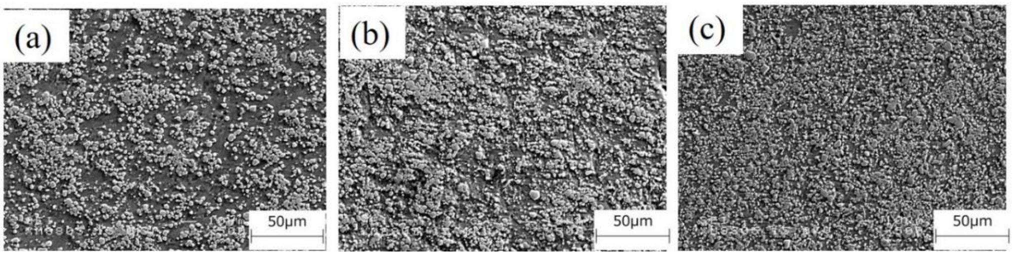

Figure 1. SEM images of (a) 10 vol.%, (b) 20 vol.% and (c) 30 vol.% TiB_2/Al composites

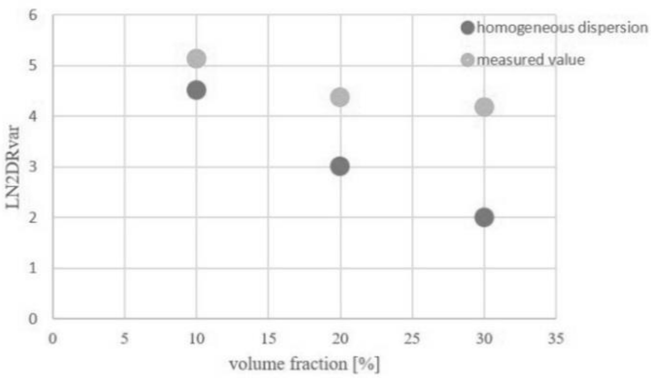

Figure 2. Relationship between $LN2DR_{var}$ and volume fraction of TiB_2 in composites

Figure 3 shows the thermal conductivity obtained by the steady state method. The thermal conductivity of the composites with TiB_2 content of 10, 20 and 30 vol.% was 147, 140 and 99 W/mK, respectively. As TiB_2 content increased, the thermal conductivity of the composites decreased, which is caused by lower thermal conductivity of TiB_2 (66.4 W/mK) than pure Al (225 W/mK). Then, the theoretical value was calculated by using Maxwell-Eucken equation, which assumes the uniform dispersion of particles, and the equation considering the effect of density. Following is Maxwell-Eucken equation:

$$\lambda = \lambda_{\mathrm{m}} \cdot \frac{(1 + 2 \cdot \Phi_t \cdot A)}{(1 - \Phi_{\mathrm{t}} \cdot A)}$$

$$\mathrm{A} = \frac{(1 - \frac{\lambda_m}{\lambda_t})}{(2 \cdot \frac{\lambda_m}{\lambda_t} + 1)}$$

Where, λ is thermal conductivity, and ϕ is volume fraction.

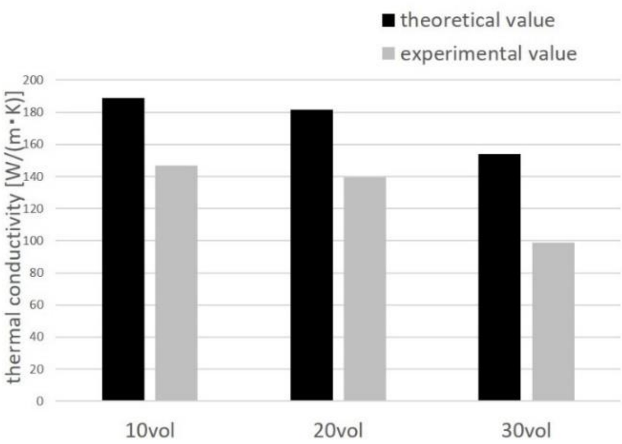

Figure 3. Thermal conductivity of 10 vol.%, 20 vol.% and 30 vol.% TiB_2/Al composites

Then, these composites have different relative density because of different sintering condition, we have to consider the effect of density before using Maxwell-Eucken equation. Following is density correction formula (EMPT model):

$$\lambda_{\mathrm{eff}} = \frac{1}{4}[\lambda_p(3v_p - 1) + \lambda_{\mathrm{c}}(3v_c - 1) \ + \left([\lambda_{\mathrm{p}}(3v_p - 1) + \lambda_c(3v_c - 1)]^2 + 8\lambda_{\mathrm{p}}\lambda_c\right)^{\frac{1}{2}}\right]$$

Where, λ_c is thermal conductivity of composites, λ_p is thermal conductivity of pore, ν_c is the relative density of composites, and ν_p is rate of pore.

By using above formulae, the theoretical value was obtained as 189, 181 and 153 W/mK for the composites with TiB_2 content of 10, 20 and 30 vol.%, respectively, which is higher than the experimental value. TiB_2 particle in this composites have a tendency of some heterogeneous dispersion, but it seems the influence of its structure on the thermal conductivity is small. In 20 vol.% TiB_2/ Al composites with 3.0 and 4.37 of LN2DR, the theoretical value of thermal conductivity was calculated as 181 and 181 W/mK, respectively. The difference of thermal conductivity between uniform distribution and some heterogeneous dispersion obtained from the sintered composites was not observed. Therefore, it is guessed that the interface heat transfer affect to the degradation of the thermal conductivity because of high grain boundary density. When the particle size is smaller than 0.48 μm, the effect of heat transfer rate at the interface is enhanced dramatically (Yamada, 2015). As a result, it seems the thermal conductivity of the experimental results became smaller than the theoretical value.

Figure 4 shows the distribution of particle diameter for TiB_2 in Al composites with 10, 20 and 30 vol.%TiB_2. With increasing volume fraction of TiB_2, the average particle size became small with 1.85, 1.71 and 1.46 μm, and there were many the particles smaller than 0.48 μm. It seems that the increment of TiB_2/Al interface density by the refinement of TiB_2 particles is caused by the difference between theoretical and experimental values.

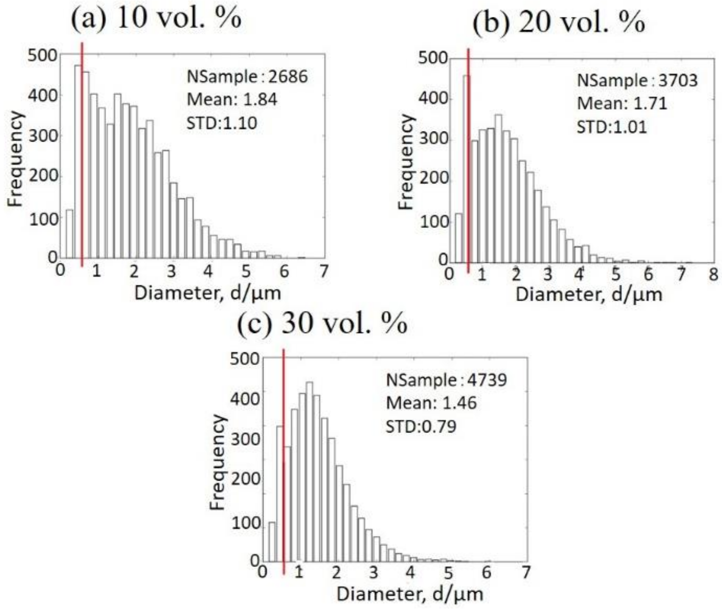

Figure 4. Particle size distribution of (a) 10 vol.%, (b) 20 vol.% and (c) 30 vol.% TiB_2/Al composites

CONCLUSIONS

Al matrix composites with volume fractions of TiB_2 particle content ranging from 10 to 30% were fabricated by SPS method. Then, the microstructure and the density of the composites were estimated, and the dispersibility and the distribution of particle size were estimated quantitatively from the SEM images. Following are the results of this study.

1. TiB_2 particle in the composite had weak heterogeneous dispersion. And with increasing the volume fraction, the degree of heterogeneous dispersion increased gradually.
2. The thermal conductivity varied from 147 to 99 W/mK with increasing the volume fraction of TiB_2 particle from 10 to 30 vol.%. This experimental value was less than the theoretical value calculated by Maxwell-Eucken equation considering the effect of the density. It is caused by the influence of the interface heat transfer rate, because these composites have many small sized particles and have high interface density.

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