Synthesis of Fine Ceramic Particles in Molten Aluminum by Combustion Reaction

Wataru Yoshida¹, Makoto Kobashi² and Naoyuki Kanetake²

¹Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan
²Department of Materials Science and Engineering, Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan

Ceramic particles (TiB₂ and TiC) dispersed aluminum alloy was synthesized by a combustion reaction. Starting materials were aluminum, titanium, boron and boron carbide. The heat of reaction between Ti/B and Ti/B₂C was too high to maintain the original configuration of the blended powder compact. Aluminum was added to the blended powder mixture to control the adiabatic temperature of the reaction. Aluminum could successfully control the adiabatic temperature and prevented the collapse of the precursor. The average size of the TiB₂ and TiC particles strongly depended on the amount of aluminum added as the diluents of the heat of reaction. By increasing the aluminum addition to the powder phase, the average size of TiB₂ and TiC particles synthesized by the combustion reaction decreased. The TiB₂ and TiC particles were extracted from aluminum matrix, and confirmed to have submicron size under the suitable conditions.

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1. Introduction

In general, a chemical reaction that synthesizes intermetallics or ceramics generates a large amount of heat of reaction. Combustion reaction is a process that makes use of the strong exothermic heat to induce the sequential chemical reaction at the neighboring part of the reacted zone. The following reactions synthesize titanium diboride (TiB₂) and/or titanium carbide (TiC) particles and generate large quantity of heat of reactions.

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\begin{align*}
\text{Ti} + 2\text{B} & \rightarrow \text{TiB}_2 & (1) \\
3\text{Ti} + \text{B}_2\text{C} & \rightarrow 2\text{TiB}_2 + \text{TiC} & (2)
\end{align*}
\]

When these reactions take place together with aluminum, aluminum matrix composite with fine ceramic particles dispersion can be fabricated. Since the combustion reaction process uses heat of reaction, only a small amount of energy is required to synthesize ceramics or intermetallics. Hence, combustion reaction is an attractive process in terms of productivity and cost-effectiveness.

By dispersing hard particles of submicron order, deformation resistance of material is strengthened by the effect of pinning dislocations. Therefore, composites dispersing a fine particle have superiority of high-temperature strength and creep resistance.

The objective of this research is to synthesize fine ceramic particles dispersed in aluminum by the combustion reaction process. In this paper;

(1) Controlling adiabatic temperatures by changing powder blending ratio
(2) Controlling the size of ceramic particles are investigated and also discussed thermodynamically

2. Experimental Procedure

Titanium powder (45μm, 99.9%), aluminum powder (45μm, 99.99%), boron powder (45μm, 99%) and boron carbide powder (10 μm, 99%) were used as starting materials. Molar blending ratio of starting materials and theoretical volume fractions of aluminum matrix are shown in Table 1. The mole blending ratio of titanium and boron powder was fixed to B/Ti = 2, and the mole blending ratio of titanium and boron carbide powder was fixed to Ti/B₂C = 3. The volume fraction of the aluminum powder was varied from 40 to 70 vol% in both cases. Cylindrical powder compacts (10 mm in diameter, 8 mm in height) were made by an indirect mixing pressure of 150 MPa. The experimental apparatus is illustrated in Fig. 1. The powder compact was heated in an induction furnace under an argon atmosphere to induce the combustion reaction. After the reaction, the cross section of the specimen was observed by a scanning electron microscope (SEM) and analyzed by an X-ray diffraction (XRD) method. Detailed observations of ceramic particles were carried out after extracting particles by immersing the composite in hydrochloric acid solution for 20 s.

3. Results and Discussion

3.1 Microstructure of Al-Ti-B system

Figure 2 shows the microstructure of specimens after the reaction of Al-Ti-B system. TiB₂ particles are uniformly dispersed in all specimens. The volume fraction of aluminum matrix was remaining as an intermediate product. Figure 2 shows the microstructure of Al-Ti-B system. TiB₂ particles are uniformly dispersed uniformly in all specimens. The size of titanium and boron carbide powder was analyzed by an intermediate product. Figure 2 shows the microstructure of Al-Ti-B system. TiB₂ particles are uniformly dispersed uniformly in all specimens. The average diameters of TiB₂ particles are 1.25 μm, 1.12 μm, 0.57 μm and 0.31 μm, respectively. By increasing the fraction of aluminum in specimen, the average...
diameter of TiB₂ particles decreased. The formation of Al₃Ti together with TiB₂ was observed in 70 vol%Al specimen. As for the Al-Ti-B reaction system, the first-stage reaction between aluminum and titanium took place immediately after the melting point of Al. This reaction raised the temperature of the specimen and triggers the second-stage reaction between titanium and boron. Therefore, when the amount of aluminum was high (70 vol% Al), the temperature of the specimen did not increase high enough by the first reaction, and the TiB₂ formation did not occur completely. High-magnification photographs of extracted TiB₂ particles of each specimen are shown in Fig. 3. A frequency distribution of TiB₂ particle size of each specimen was calculated as shown in Fig. 4. As increasing the aluminum volume fraction, irregularity of TiB₂ particle size became smaller.

3.2 Microstructure of Al-Ti-B₄C system

Figure 5 shows the microstructure of specimens after the reaction of Al-Ti-B₄C system. TiB₂ and TiC particles are dispersed uniformly in all specimens. Figure 5 shows the microstructures of 40 vol%Al, 50 vol%Al, 60 vol%Al, and 70 vol%Al specimens, and the average diameters of TiB₂ particles are 0.68 µm, 0.49 µm, 0.44 µm, and 0.21 µm, respectively. High-magnification photographs of extracted particles from each specimen are shown in Fig. 6. Similar to the Al-Ti-B system, the size of particles generated in aluminum matrix decreased by increasing aluminum volume fraction. The ceramic particles synthesized from B₄C powder became smaller than that of Al-Ti-B system. This is probably because B₄C needed to decompose in prior to the reaction and created finer nucleation sites of TiB₂ and TiC.

3.3 Calculations of adiabatic temperature

The adiabatic combustion temperatures of blending powders containing boron and boron carbide were calculated by
In the Al-Ti-B system, the adiabatic temperatures of the 40 and 50 vol%Al specimens can be calculated as 2723 K, which is the boiling point of aluminum. Hence, aluminum vapor raised the internal pressure of powder compacts and made the specimen broken into small pieces. The adiabatic temperatures of 60 vol%Al and 70 vol%Al specimen are 2461 K and 2158 K, respectively. In these specimens, the adiabatic temperatures are below the boiling point of aluminum, and the original configuration of powder compact was maintained. As for the Al-Ti-B₄C system, the adiabatic temperature of 40, 50, 60 and 70 vol%Al specimens were 2723 K, 2573 K, 2243 K and 2008 K, respectively. Figure 7 shows the theoretically calculated adiabatic temperature and the practically measured combustion temperature as a function of aluminum addition. The theoretical data is higher than the practically measured data at all range of aluminum addition. This is because some fraction of reaction heat was emitted to the ambient atmosphere in practice. The temperature was effectively controlled in the wide range of about 800 K both in theory and in practice. When ceramic particle was synthesized by the combustion reaction, the size of the ceramic particle was affected by the combustion temperature because of larger growth of ceramic particle at higher temperatures. Thus, the addition of aluminum could be one of the effective methods to control the size of TiB₂ and TiC particles. Relation between a ceramic particle size and measured combustion temperatures is summarized in Fig. 8. The particle size was effectively reduced to sub-micron levels by decreasing the combustion temperature to around 1800 K or lower. It was also confirmed that the finer ceramic particles were synthesized from the Al-Ti-B₄C system.

\[
\begin{align*}
    &\ xAl + 3Ti + B_4C \rightarrow xAl + 2TiB_2 + \Delta H_f(TiB_2) \\
    &\ \\
    &\ \\
    &\ \\
\end{align*}
\]

the equations (3) and (4), respectively. The initial temperature was assumed as 300 K.

The reactions of (3) and (4) can be expressed as:

\[
\begin{align*}
    &\ xAl + Ti + 2B \rightarrow xAl + TiB_2 + \Delta H_f(TiB_2) \\
    &\ \\
    &\ \\
\end{align*}
\]

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\begin{align*}
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This is probably because $B_4C$ needed to decompose in prior to the reaction and created finer nucleation sites of $TiB_2$ and $TiC$.

4. Conclusion

Ceramic particle dispersed aluminum alloy was synthesized by combustion reaction and the effect of aluminum addition as diluents on the particle size was also investigated. The following results were obtained through the experiment.

1) The combustion temperature of samples becomes lower by increasing aluminum volume fraction.
2) Particle size of the ceramic generated by combustion reaction is smaller as the combustion temperature becomes lower.
3) The smallest size of the ceramic particles is about $0.3 \mu m$ in 70 vol% Al sample.

4) In comparison with the Al-Ti-B system, the size of particles synthesized in Al-Ti-B$_4$C system is smaller.

REFERENCES