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Fabrication of TiB_2 Particle Dispersed FeAl-based Composites by Self-propagating High-temperature Synthesis

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FeAl–TiB_2 composites have been produced from mixtures of Fe, Al, Ti and B powders using the Self-propagating High-temperature Synthesis (SHS or combustion synthesis) method. When the powder mixture was heated in vacuum to approximately 900 K, an abrupt increase in temperature was observed, indicating that the SHS reaction occurred in the powder mixture. The treating time from the start of heating to the end of the exothermic reaction was only approximately two minutes. X-ray diffraction analyses revealed that the SHS sample consisted of only FeAl and TiB_2 without any elemental metals and any other compounds. Metallographic investigations using a scanning electron microscope and an electron probe microanalysis revealed that fine TiB_2 particles were dispersed in FeAl matrix phase. As the volume fraction of the TiB_2 particles was increased from 0.3 to 0.8 by controlling the powder mixture composition, the average TiB_2 particle size increased from 1 to 7 μm and the average Vickers hardness of the composites increased from 800 to 1 600.

KEY WORDS: ceramic; intermetallic; hard metal; powder metallurgy; combustion synthesis; SHS.

1. Introduction

An iron aluminide FeAl has a low density and an excellent corrosion and oxidation resistances, and it consists of inexpensive raw materials of iron and aluminum.[1] Therefore, FeAl can be a candidate material for engine components. It has been reported that the dispersion of hard ceramic particles in FeAl improves the mechanical properties such as hardness, wear resistance, compressive strength and so on. Schneibel *et al.*[2] produced FeAl–ceramic composites using the liquid-phase sintering method and reported they had excellent mechanical properties. Subramanian and Schneibel[3] produced FeAl–TiC and FeAl–WC composites using the melt infiltration processing and investigated the mechanical properties of the composites. They reported that as the carbide content increased, hardness also increased while bend strength and fracture toughness decreased. They also reported that the FeAl–TiC and FeAl–WC composites had much more excellent oxidation resistance than a Co–WC composite, which is well known as the cemented carbide. Durlu[4] produced FeAl–TiC composites using the reaction sintering of powder mixtures of Fe, Al and TiC and reported the density, hardness and bend strength. Krasnowski *et al.*[5] produced FeAl–30%TiC composite using the mechanical alloying and hot-pressing consolidation.

The above-mentioned methods, namely, the liquid-phase sintering method, the melt infiltration processing and the mechanical alloying and hot-pressing consolidation, are all useful for producing hard intermetallic-ceramic composite materials. However, it seems that there is still great room for improvement from viewpoints of efficiency and cost. They all require high temperatures and long periods of time. In this study, we propose a more simple and inexpensive production method for intermetallic-ceramic composites based on the SHS (Self-propagating High-temperature Synthesis) method, using FeAl–TiB_2 hard composites as a demonstration material.

2. Procedure

2.1. Compaction of Powder Mixture

The elemental powders of Fe, Al, Ti and B were mixed by hand in a glass beaker using a steel spoon with the addition of small amount of ethanol, and the powder mixture was pressed into a cylindrical shape of a 25-mm diameter and a 20-mm height using a metal mold and punches under a uniaxial pressure of 600 MPa in air at room temperature. The molar ratios of the elemental metals in the mixed powder were Fe : Al = 1 : 1 and Ti : B = 1 : 2, and the estimated volume fraction of TiB_2 was varied from 0 to 0.8 by changing the composition of powder mixture based on the densities and molecular weights of the elements and the compounds.

2.2. SHS under Atmospheric Pressure

The powder compact was heated in a graphite vessel using an infrared furnace at a heating rate of 10 K/s in an argon gas of 1 atmospheric pressure. The change in the temperature of the powder compact was monitored using a B-type platinum thermocouple covered by a 6-mm diameter alumina protective tube. When sudden and dramatic temperature rise indicating the onset of the SHS reaction was observed, the electric power supply to the furnace was

switched off, and then the sample was cooled in the furnace.

The SHS samples were sectioned and polished for metallographic observations using an optical microscope (OM) and a scanning electron microscope (SEM). The X-ray diffraction (XRD) analysis and the electron probe microanalysis (EPMA) were performed for the SHS samples to identify the reaction products. The density, porosity and hardness were measured using the Archimedes method, an image analyzer and a Vickers hardness tester, respectively.

2.3. SHS under High Pressure

The SHS reaction under a high pressure was also performed using the pseudo Hot Isostatic Pressure (pseudo-HIP) method. A schematic illustration of the pseudo-HIP equipment is shown in **Fig. 1**. As shown in the figure, the sample was heated using a resistance wire made of an Fe–Cr–Al alloy in a pressing media of alumina powder in vacuum. A pressure of 100 MPa was initially applied and the pressure was immediately raised to 600 MPa. Then, the electricity supply to the wire was stopped when the sudden temperature rise due to the SHS reaction was observed. After holding for 5 min at this pressure, the sample was removed from the equipment and cooled. The same measurements, observations and examinations as those described in Sect. 2.2 were also performed for the samples produced under a high pressure.

3. Results and Discussion

3.1. Products of SHS Reaction

Figure 2 shows a temperature-time curve for a sample having a TiB_2 volume fraction (f_v) of 30%. The temperature gradually rose due to external heating. When the temperature reached approximately 700°C, the temperature rise was slightly slowed down due to melting of aluminum. When the temperature reached approximately 800°C, the temperature suddenly and dramatically increased to approximately 1 600°C in approximately 10 s. This sudden temperature rise implies that the SHS reaction occurred between the liquid aluminum and solid elemental powders of Fe and Ti. Similar sudden temperature rise was observed in all compacts having different compositions. After the sample was taken out from the pseudo-HIP equipment shown in Fig. 1, it was subjected to the XRD analysis. The analysis revealed that the sample consisted of only FeAl and TiB_2, as shown in **Fig. 3**. Other samples having different compositions also consisted of only FeAl and TiB_2. The results indicate that a reaction expressed by Eq. (1) occurred.

$$Fe+Al+Ti+2B=FeAl+TiB_2 \quad \text{.....(1)}$$

The samples were sectioned and polished for the metallographic observations. **Figure 4** shows the microstructures of the samples. In all samples, dark gray particles were dispersed in bright matrix phase. According to the EPMA results, the dark gray particles are stoichiometric TiB_2, while the matrix is FeAl containing titanium and boron at 3 to 4 mol%. Judging from the 2-D shape of the TiB_2 particles on the cross-sections, they seem to have a 3-D shape of a rectangular paralleleleppiped. The TiB_2 particle diameter increased with the increase in volume fraction of TiB_2. The

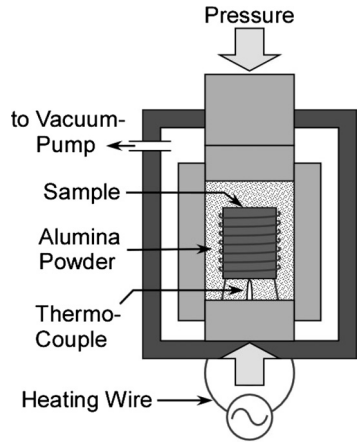

Fig. 1. Schematic illustration of the pseudo-HIP equipment.

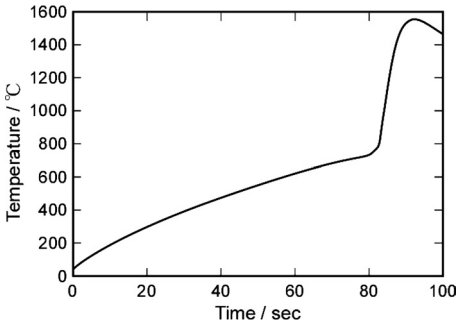

Fig. 2. Temperature change during heating of a powder compact without pressure, f_V=30%.

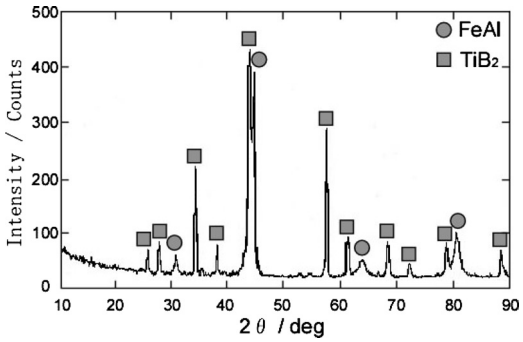

Fig. 3. XRD spectrum of an SHS sample, f_V=30%, without pressure.

average particle diameter was measured as the area equivalent diameter and the results are shown in **Fig. 5**. The average particle diameter increased from 1 to 7 μm as the volume fraction increased from 0.3 to 0.8.

3.2. Reaction Model

Heating of the powder mixture to about 800°C brought about the production of the TiB_2 particle dispersed FeAl-based composites, as shown in Fig. 4. We considered a model for the production of these composites, as follows. In the heating process, aluminum melts first because it has the lowest melting point in the four elemental metals. The melted aluminum infiltrates into the interfaces between the powders, which leads to the exothermic reactions between the aluminum liquid and powders of Fe and Ti, producing iron aluminides and titanium aluminides. As the temperature in-

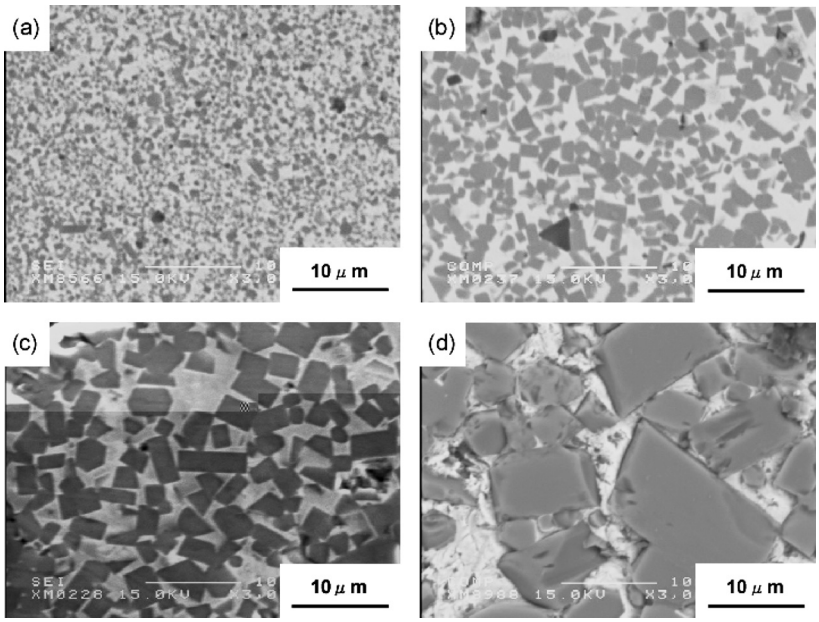

Fig. 4. SEM images of SHS samples with a pressure of 600 MPa, (a) f_V=30%, (b) 50%, (c) 60%, (d) 80%.

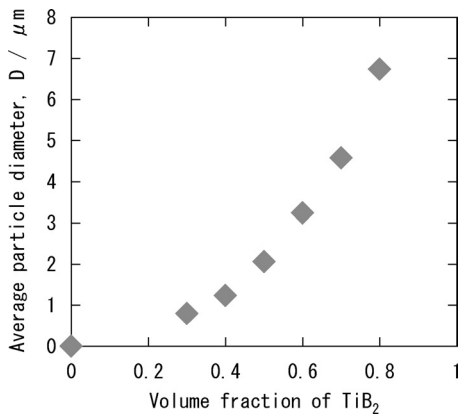

Fig. 5. Effect of the volume fraction of TiB_2 particles on the average particle diameter; samples were produced with a pressure of 600 MPa.

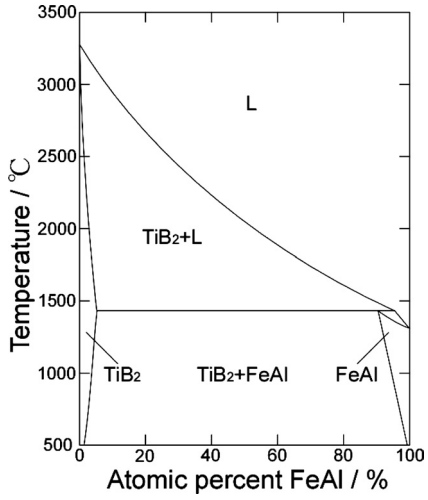

Fig. 6. Hypothetical TiB_2–FeAl pseudo-binary phase diagram.

creases due to these exothermic reactions, a liquid alloy appears. This liquid reacts with B powder, resulting in the generation of additional heat. Finally, an Fe–Al–Ti–B quaternary liquid with an extremely high temperature is produced.

Although detailed Fe–Al–Ti–B quaternary phase diagrams have not been published, a TiB_2–FeAl pseudo-binary eutectic or peritectic phase diagram shown in **Fig. 6** may be proposed based on Ko and Hanada's hypothetical TiC–Fe_3Al pseudo-binary eutectic phase diagram.[6] According to this diagram, during cooling of the liquid produced by the SHS reaction, TiB_2 crystallizes as the primary crystal, and then FeAl crystallizes by a peritectic reaction as the secondary crystal. Moreover, when TiB_2 concentration is higher, the primary TiB_2 crystal appears at higher temperatures and it coexists with the residual liquid for longer periods during cooling, which promotes the growth of TiB_2 particle at higher TiB_2 concentrations. This may be a reason why TiB_2 particles were larger when TiB_2 concentration was higher, as shown in Figs. 4 and 5.

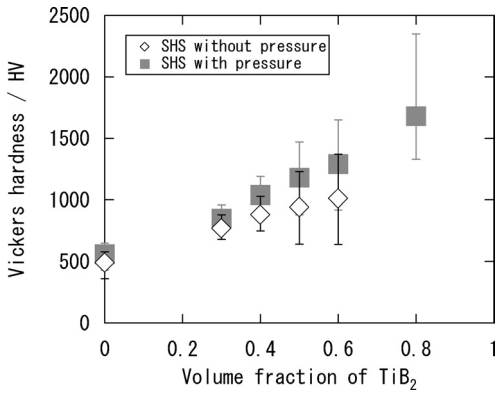

Fig. 7. Effects of the volume fraction of TiB_2 particles on the Vickers hardness of the SHS composites.

3.3. Vickers Hardness

Vickers hardness increased with an increase in volume fraction of TiB_2, as shown in **Fig. 7**. Schneibel *et al.*[2] reported that their liquid-sintered FeAl–50%TiB_2 composites exhibited a Vickers hardness number of 730. Our compos-

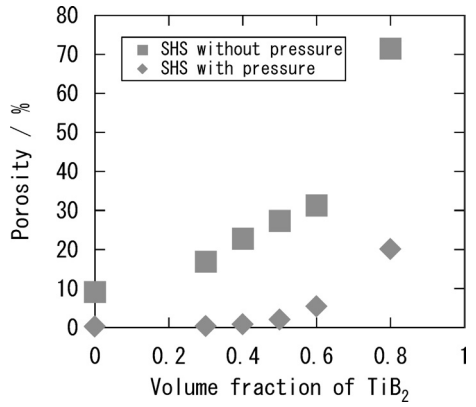

Fig. 8. Effects of the volume fraction of TiB_2 particles on the porosity of the SHS composites.

ite having the same TiB_2 concentration exhibited a similar hardness for SHS samples without pressure, as shown in Fig. 7. However, the hardness increased to approximately 1 000 for SHS samples with pressure. In all composites having different TiB_2 concentrations, the application of pressure during the SHS brought about an increase in Vickers hardness. This improvement in hardness was due to the decrease in porosity caused by the pressure application during the SHS reaction, as shown in **Fig. 8**. Vickers hardness could not be measured for a sample having $f_V=80\%$, because it had a high porosity of 75% as shown in Fig. 8. However, the porosity was reduced to 20% by applying a pressure of 600 MPa. Increase in pressure applied during the SHS reaction will reduce the porosity in the reaction product.

4. Summary

When powder mixtures consisting of Fe, Al, Ti and B having molar ratios of Fe : Al = 1 : 1 and Ti : B = 1 : 2 were heated to approximately 800°C, the SHS reaction occurred and TiB_2 particle dispersed FeAl-based composites were produced in approximately 10 s. As the volume fraction of the TiB_2 particles increased, the diameter of the particle and the Vickers hardness of the composites increased. The application of a pressure during the SHS reaction led to a decrease in porosity and an increase in Vickers hardness.

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