

In-situ Preparation of Boride-Particle-Dispersion-Strengthened Aluminum Alloys by Mechanical Alloying

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ABSTRACT Mechanical alloying (MA) and reaction-heat-treatment were employed to prepare the particle-dispersion-strengthened aluminum alloys using elemental aluminum, transition metal, M ($M = \text{Ti, Zr, Nb}$ or Ta) and boron powders as starting materials. The powders mechanically alloyed for 72~180 ks were heat-treated in the temperature range from 573 to 873 K for 3.6 ks in vacuum.

In all systems, very fine particles of TiB_2 , ZrB_2 , NbB_2 and TaB_2 were precipitated and dispersed *in-situ* by heating the MA powders at 773~873 K in Al-Ti-B, Al-Zr-B, Al-Nb-B and Al-Ta-B systems, respectively. The boride particle sizes dispersed in the Al matrices ranged up to about 50 nm. The grain sizes of Al matrices ranged from 100 to 500 nm.

Keywords: *mechanical alloying, aluminum alloy, transition metal diboride, particle-dispersion-strengthening, in-situ preparation*

1. INTRODUCTION

Many aluminum alloys [1] have been developed up to date as Duralumin, Superduralumin and Extra superduralumin. These alloys were strengthened by the age-hardening effect of the precipitation of metastable phases such as Guinier-Preston zones and intermediate precipitates. However, these alloys degrade by the growth of precipitates i.e. over aging phenomenon when they are put in the elevated temperature environment.

In order to improve this shortcoming and to develop the aluminum alloys which can hold the high strength after the exposure in elevated temperature, the particle-dispersion-strengthened aluminum alloys have been studying of which particles are oxide, carbide, boride and nitride having the rigidity even in the high temperature range. Usually, they are built up by mechanical alloying (MA) [2] using the oxide, carbide, boride and nitride particles as starting powders with matrix material.

If these particles could not be crushed finely during MA, they could not work as dispersoids sufficiently. To obtain the finely dispersion of the particles easily, MA and reaction-heat-treatment are adopted for *in-situ* preparation of the particle-dispersion-strengthened alloys which were mechanically alloyed using elemental powders followed by reaction-heat-treatment. In the present study, the authors intend to prepare the transition metal diboride particle-dispersion-strengthened aluminum alloys by combining MA and reaction-heat-treatment because the transition metal diborides hardly resolve to aluminum matrix.

2. EXPERIMENTAL PROCEDURE

The starting materials used in the present study were elemental powders of aluminum (average particle size; $150 \mu\text{m}$), titanium ($14.0 \mu\text{m}$), zirconium ($5.4 \mu\text{m}$), niobium ($21.0 \mu\text{m}$), tantalum ($1.6 \mu\text{m}$) and amorphous boron ($0.7 \mu\text{m}$). Assuming that the particles of TiB_2 , ZrB_2 , NbB_2 and TaB_2 could be precipitated by heating in vacuum in the mechanically alloyed Al-Ti-B, Al-Zr-B, Al-Hf-B, Al-Nb-B and Al-Ta-B system powders, the starting powders were mixed in the ratios of 10, 20 and 30 volume percentages. MA was carried out up to 180 ks using a Spex 8000 mixer mill under an argon atmosphere with stainless steel vessel and steel balls. In order to prevent the agglomeration of powders in the course of mechanical alloying, small amounts of methyl alcohol were added each batches as the process control reagent (PCR). The mechanically alloyed powders were heated for degassing in vacuum at 873 K for 3.6 ks and for converting into the boride-particle-dispersion-strengthened aluminum alloys.

The powders obtained were identified the phases by X-ray diffractometry with the monochromatized $\text{Cu K}\alpha$ radiation. A part of the powders obtained by degassing was canned in aluminum capsules and extruded into the rods at 873 K with extrusion ratio of 16. The microstructures, particle morphology and distribution were investigated with a transmission electron microscope (TEM), (JEOL; JEM-2000FX) operating at 200 kV.

3. RESULTS AND DISCUSSION

3.1 Al-Ti-B system

Figure 1 shows the change in the X-ray diffraction patterns of Al-Ti-B (30vol% TiB_2) by MA. All the powders of this system became the α -Al solid solution by the MA up to 180 ks. In this system powders, the self propagating high temperature synthesis (SHS) reaction between Ti and B or Al and Ti did not take place but α -Al supersaturated solid solutions were obtained by MA up to 180 ks. It was obvious that the diffraction lines of Al shifted toward the higher diffraction angle by the solution of Ti to Al after MA for 72 ks. The lattice parameter of Al obtained from Al(311) diffraction line was 0.3986_6 nm in Al-30vol% TiB_2 . This result accords with the values edited by Pearson[3] in which the lattice parameter of Al was decreased by the solution of Ti to Al matrix up to about 0.35 mass%Ti. In contrast, it was recognized that the large expansion of solubility limit of Ti to Al was

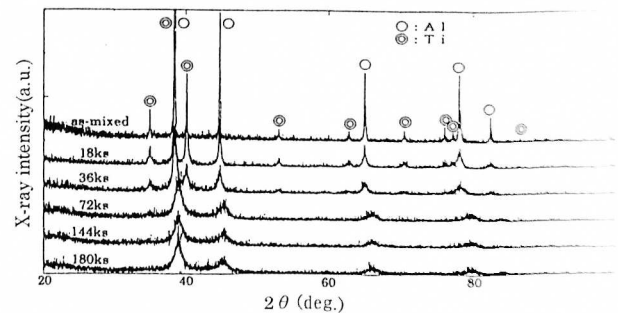


Fig.1 Change in the X-ray diffraction patterns of Al-Ti-B (30vol% TiB_2) by mechanical alloying

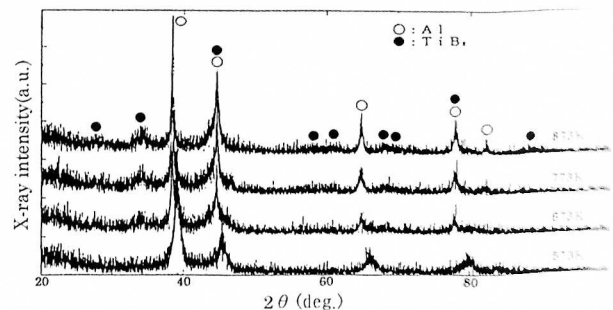


Fig.2 Change in the X-ray diffraction patterns of Al-Ti-B (30vol% TiB_2) powders mechanically alloyed for 180 ks followed by heating at various temperatures for 3.6 ks.

caused by MA in the present study.

Figure 2 shows the change in the X-ray diffraction patterns obtained from the Al-Ti-B(30vol%TiB₂) powders which were mechanically alloyed for 180 ks followed by the heat-treatment at 573~873 K for 3.6 ks. The heat-treatment at 573 K did not give any phase transformation except the release of strain accumulated by MA. In the temperature range of 673~873 K, TiB₂ phase precipitated from the supersaturated solid solution made by MA. In accord with the precipitation of TiB₂ it was observed that the diffraction angle of Al alloy returned back to pure Al.

Figure 3 shows the typical TEM images and selected area diffraction(SAD) patterns obtained from Al-5, -10 and -20 vol%TiB₂ mechanically alloyed for 72 ks followed by extruding at 873 K. The grain size of Al matrix became finer with increasing of the concentration of alloying elements, which were about 500, 300 and 200 nm in Al-5, -10 and -20vol%TiB₂ alloys, respectively. It was obvious from SAD patterns that the TiB₂ particles were precipitated and dispersed uniformly in the Al matrix. The morphology of the TiB₂ precipitates was rectangular parallelepiped. It was considered that very fine microstructure was attributed to the prevention of grain growth caused by dispersion of very fine particles.

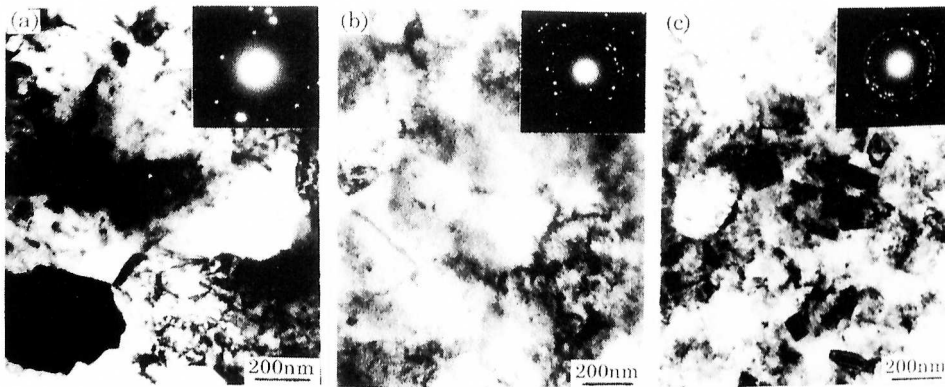


Fig.3 Typical TEM images and SAD patterns obtained from (a) Al-5, (b)-10 and (c)-20vol%TiB₂ alloys extruded at 873 K.

3.2 Al-Zr-B system

Although the starting Zr powder contained small amount of Zr hydride, the mixed powders containing 10vol%ZrB₂ were changed to the mixture of Al and Zr hydride of which amount was increased by the reaction between Zr and methyl alcohol added as PCR in the course of MA.

Figure 4 shows the change in the X-ray diffraction patterns of Al-Zr-B(30vol% ZrB₂) by MA. The Al-Zr-B system powders involving 20 and 30vol% ZrB₂ were changed to a metastable (Al,Zr) phase [4] including small amount of Zr after MA for 72 ks.

Figure 5 shows the change in the X-ray diffraction patterns of Al-Zr-B (30 vol% ZrB₂) powders mechanically alloyed for 72

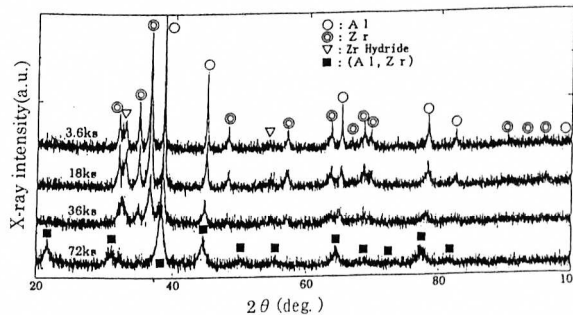


Fig.4 Change in the X-ray diffraction patterns of Al-Zr-B(30vol% ZrB₂) by MA.

ks followed by the heat-treatment at various temperatures for 3.6 ks. Although the heat-treatment at 573 K for 3.6 ks made the X-ray diffraction lines sharpen, it did not give the MA powders any crystallographic phase transformation. By the heat-treatment in the temperature range between 673-873 K for 3.6 ks, ZrB_2 particles precipitate in the MA powders. In addition, the intermetallic compound, Al_3Zr , generated in the powder of Al-30vol% ZrB_2 .

Figure 6 shows the typical TEM images and SAD pattern taken from the Al-10 vol% ZrB_2 alloy mechanically alloyed for 72 ks followed by extruding at 873 K. As shown with arrows in Fig.6(b), the morphology of ZrB_2 precipitates was rod-shaped one with an aspect ratio about 5. It was different from the morphology of TiB_2 particles in rectangular parallelepiped as mentioned in the section 3.1. Although the crystal structure of TiB_2 and ZrB_2 is AlB_2 -type hexagonal of which space group is D_{6h}^1 -P6/mmm [5], the reason for this difference in morphology is not still clear.

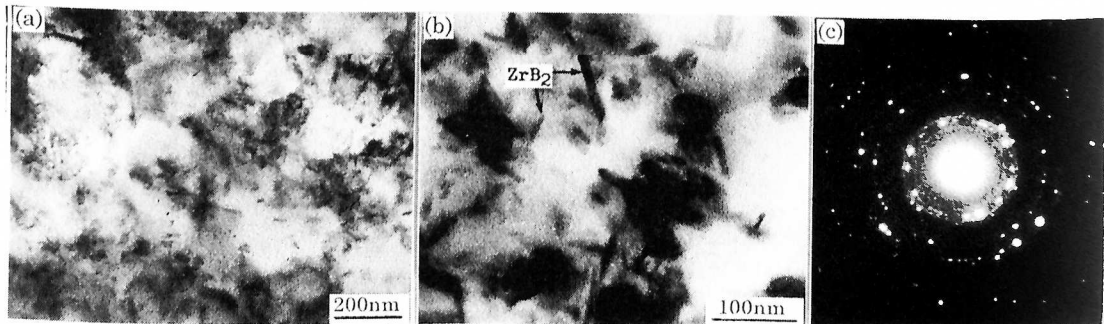


Fig.6 Typical TEM images(a,b) and SAD pattern(c) obtained from Al-10vol% ZrB_2 alloy extruded at 873 K.

3.3 Al-Nb-B system

The mixed powders up to 10 vol% NbB_2 were changed to the mixture of Al and Nb hydride ($NbH_{0.89}$) by MA for 72 ks. Figure 7 shows the change in the X-ray diffraction patterns of Al-Nb-B (30vol% NbB_2) by MA. The powders containing alloying elements corresponding to 20 and 30 vol% NbB_2 changed to Al and $NbH_{0.89}$ by MA for 36 ks, the further MA produced Al_3Nb intermetallic compound by MA for 72 ks.

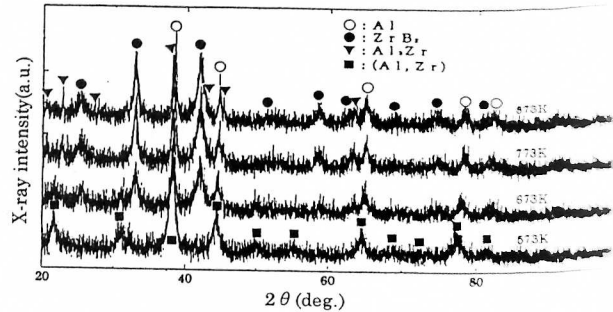


Fig.5 Change in the X-ray diffraction patterns of Al-Zr-B (30vol% ZrB_2) powders mechanically alloyed for 72 ks followed by heating at various temperatures for 3.6 ks.

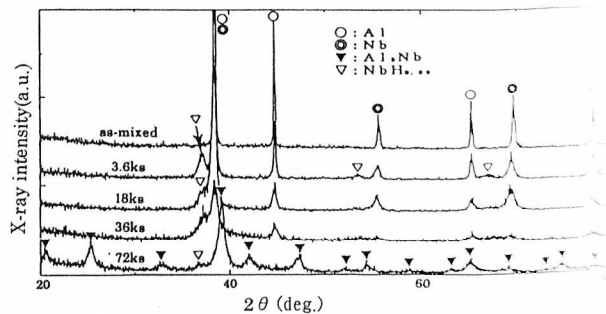


Fig.7 Change in the X-ray diffraction patterns of Al-Nb-B(30vol% NbB_2) by MA.

Figure 8 shows the change in the X-ray diffraction pattern obtained from Al-Nb-B (30 vol% NbB₂) powders mechanically alloyed for 72 ks followed by the heating at various temperatures for 3.6 ks. The precipitation of NbB₂ started by the heat-treatment at 673 K for 3.6 ks, then NbB₂-particle-dispersion-strengthened aluminum alloy powders containing a small amount of Al₃Nb were obtained by the heat-treatment at 873 K for 3.6 ks. The NbH_{0.89} phase generated by MA disappeared by the heat-treatment at 773 K for 3.6 ks.

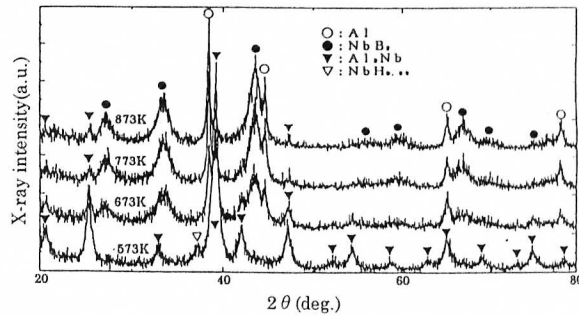


Fig.8 Change in the X-ray diffraction patterns of Al-Nb-B(30vol%NbB₂) powders mechanically alloyed for 72 ks followed by heating at various temperatures for 3.6 ks.

Figure 9 shows the typical TEM images and SAD pattern taken from the Al-10vol%NbB₂ mechanically alloyed for 72 ks followed by extruding at 873 K. It was observed that the NbB₂ particles were precipitated as very fine particles in Al matrix. The grain sizes of NbB₂ and Al matrix were about 50 and 200~300 nm, respectively.

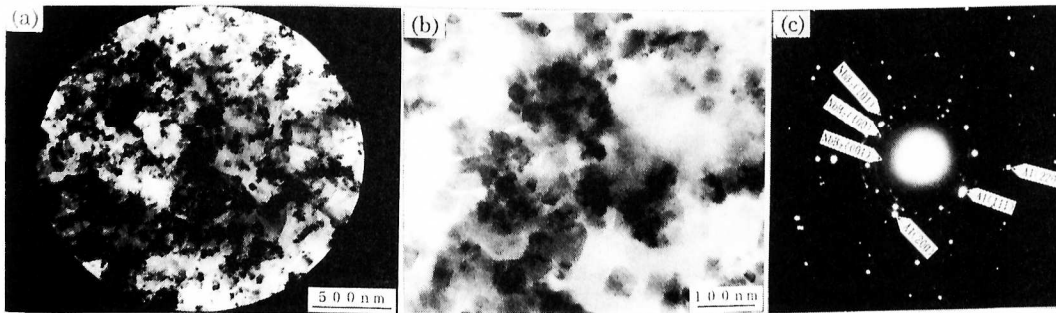


Fig.9 Typical TEM images(a,b) of Al-10vol%NbB₂ alloy extruded at 873 K and SAD pattern(c) obtained from the area in figure(a).

3.4 Al-Ta-B system

Figure 10 shows the change in X-ray diffraction patterns obtained from the Al-Ta-B(20vol%TaB₂) powder. It was obvious that the mixtures of Al and Ta hydride (TaH_{0.8}) were obtained by MA for 72 ks. Whereas the mixed powder containing 30 vol% TaB₂ changed Al₃Ta intermetallic compound by the SHS caused by MA up to 72 ks.

In the powders containing up to 10vol%TaB₂, the mixture of Al and TaH_{0.8} was obtained by the heat-treatment at 673 K. The TaB₂-dispersion-strengthened aluminum alloy powders were obtained by the heat-treatment over 773 K.

In the powders containing 20vol%TaB₂,

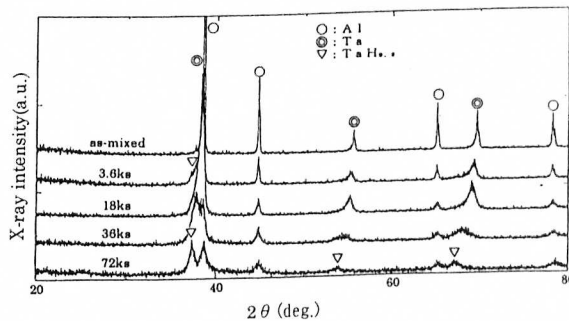


Fig.10 Change in the X-ray diffraction patterns of Al-Ta-B(20vol%TaB₂) by MA.

the TaB₂-dispersion-strengthened aluminum alloy powders were obtained by the heat-treatment over 673 K. Figure 11 shows the change in the X-ray diffraction pattern obtained from Al-Ta-B (30vol%TaB₂) powders mechanically alloyed for 72 ks followed by the heat-treatment at various temperatures for 3.6 ks. The powders consisting Al₃Ta and TaB₂ were obtained by the heat-treatment over 673 K for 3.6ks.

Figure 12 shows the typical TEM images and SAD patterns taken from the Al-5 and -10 vol%TaB₂ mechanically alloyed for 72 ks followed by extruding at 873 K. It was observed that the TaB₂ particles were precipitated as very fine particles in Al matrix. The grain sizes of aluminum matrices were about 500, 300 and 200 nm in the Al-5, -10 and -20 vol%TaB₂ alloys respectively. The particle sizes of TaB₂ were about 20 nm in Al-10 vol%TaB₂ alloy.

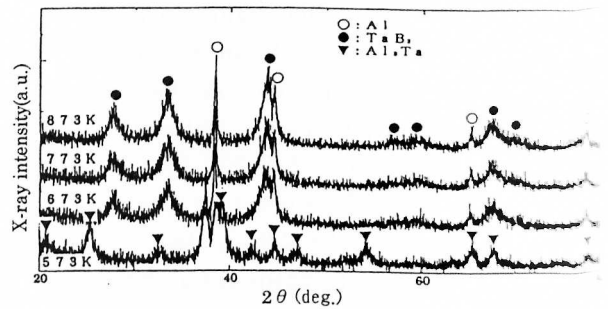


Fig.11 Change in the X-ray diffraction patterns of Al-Ta-B(30vol%TaB₂) powders mechanically alloyed for 72 ks followed by heating at various temperatures for 3.6 ks.

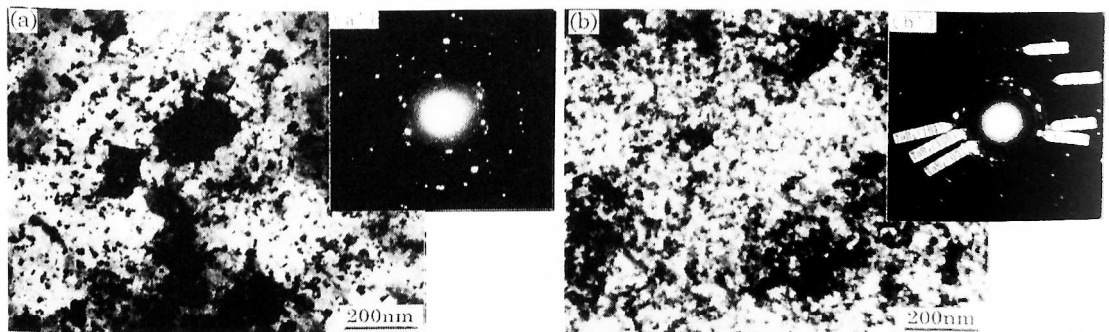


Fig.12 Typical TEM images and SAD patterns obtained from (a) Al-5 and (b)-10 vol%TaB₂ alloys extruded at 873 K.

4. CONCLUSION

MA and reaction-heat-treatment were employed to prepare the particle-dispersed strengthened aluminum alloys using elemental aluminum, transition metal and boron powders as starting materials. The powders mechanically alloyed for 72~180 ks were heat-treated in the temperature range from 573 to 873 K for 3.6 ks in vacuum.

In all systems, very fine particles of TiB₂, ZrB₂, NbB₂ and TaB₂ were precipitated and dispersed *in-situ* by heating the MA powders at 773~873 K in Al-Ti-B, Al-Zr-B, Al-Nb-B and Al-Ta-B systems, respectively. The boride particle sizes dispersed in the Al matrices ranged up to about 50 nm. The grain sizes of Al matrices ranged from 100 to 500 nm.

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