Development of High-Strength Aluminum Alloys for Basket in Transport and Storage Cask for High Burn-up Spent Fuel

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1. Introduction

Mitsubishi Heavy Industries, Ltd. (MHI) has developed high-strength borated aluminum alloys (high-strength B-Al alloys), suitable for application to baskets in transport and storage casks for high burn-up spent fuels.

Aluminum is a suitable base material for the baskets due to its low density and high thermal conductivity. The aluminum basket would reduce weight of the cask, and effectively release heat generated by spent fuels. MHI had already developed borated aluminum alloys (high-toughness B-Al alloy) [1], and registered them as ASME Code Case “N-673”. However, there has been a strong demand for basket materials with higher strength in the case of MSF (Mitsubishi Spent Fuel) casks for high-burn up spent fuels, since the basket is required to stand up to higher stress at higher temperature. The high-strength basket material enables the design of a compact cask under a limitation of total size and weight. MHI has developed novel high-strength B-Al alloys which meet these requirements, based on a new manufacturing process. The outline of mechanical and metallurgical characteristics of the high-strength B-Al alloys is described in this paper.

2. Targets of Development

High-strength B-Al alloys had been developed to satisfy following requirements.

(1) High boron content (over 3 mass% as boron)
(2) High strength (0.2% proof stress of over 100MPa at 250°C)
(3) Enough toughness to withstand impact force
(4) Stable mechanical properties during transport and storage at high temperature
(5) High thermal conductivity

3. Manufacturing Process

Mechanical alloying (MA) has been adopted as a key process in manufacturing high-strength B-Al alloy. Raw materials, aluminum alloy powder and boron carbide (B_4C) powder, are premixed by a blender in the desired boron content (up to 9 mass% as B_4C). Then, in the MA process, the premixed powders are mechanically milled with alumina balls using a high-speed steel attritor in a sealed and argon purged steel chamber. After the milling, the powders are consolidated into billets by pressing and vacuum sintering. Tubes for baskets are produced by hot extrusion of the sintered billets. As shown in Fig.1, two types of the tube for MSF casks are designed. “Square tube” is designed mainly for BWR spent fuels. The square tubes are arranged parallel to each other in the cask, and each of the tube holds one spent fuel. On the other hand, the “double-hollow tube” is designed mainly for PWR fuels. The double-hollow tubes are assembled into lattice structure, and each square of the lattice holds one spent fuel.

(a) (b)

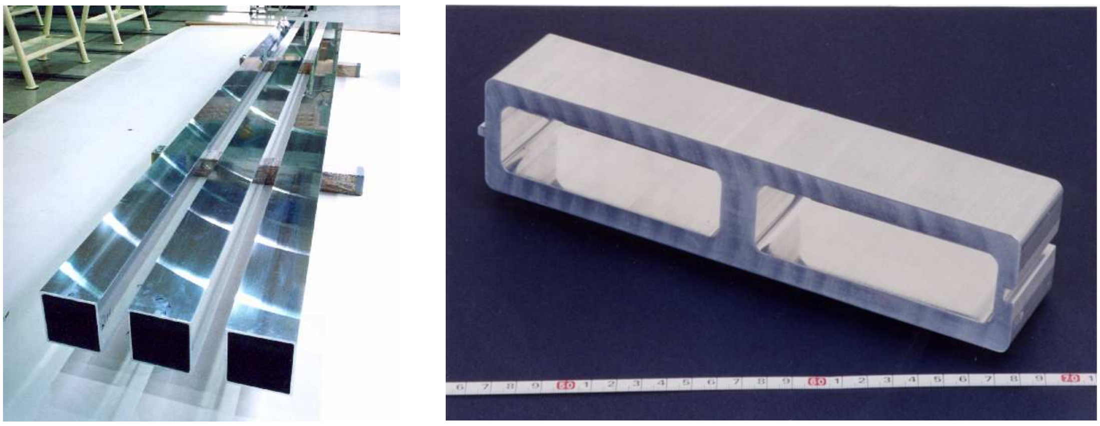

Fig.1 Outer appearance of (a)”square tube” high-toughness B-Al alloy and (b)”double-hollow tube ” high-strength B-Al alloy

4. Material Characteristics of High-Strength B-Al Alloy

4.1 Chemical composition

Chemical compositions of the high-strength B-Al alloy have been determined by modifying that of the high-toughness B-Al alloy. Chemical compositions of three types of high-strength B-Al alloy MBL05, MBL07 and MBL09 are shown in Table 1. The B_4C contents in these alloys are 5, 7 and 9 mass%, respectively. Alloying elements are magnesium and silicon. However, it has not been intended that the alloys are age hardened as AA6xxx series Al-Mg-Si alloy.

Table 1 Chemical compositions of high-strength B-Al alloy (mass%)

Alloy	B_4C (nominal value)	Si	Mg	Fe
MBL05	5	0.13	0.70	0.26
MBL07	7	0.12	0.53	0.21
MBL09	9	0.12	0.52	0.32
High-toughness B-Al	1.5-9	0.4-0.9	0.4-0.8	0.35max
AA6061	-	0.4-0.8	0.8-1.2	0.7max

4.2 Microstructure and B_4C dispersion

Fig.2 shows an optical micrograph of MBL05 alloy. All the dark parts were identified as B_4C particles by EPMA (Electron Probe Microanalyzer). The size of the particles is from submicron to 20μm. Uniform B_4C dispersion is obtained by mixing of the raw materials using the blender.

Fig.3 shows crystal grain structure of MBL09 alloy observed by TEM (Transmission Electron Microscope). The average size of the grain is under 1μm. This grain size is extremely finer than that of the aluminum alloys produced through melting process or conventional powder metallurgy.

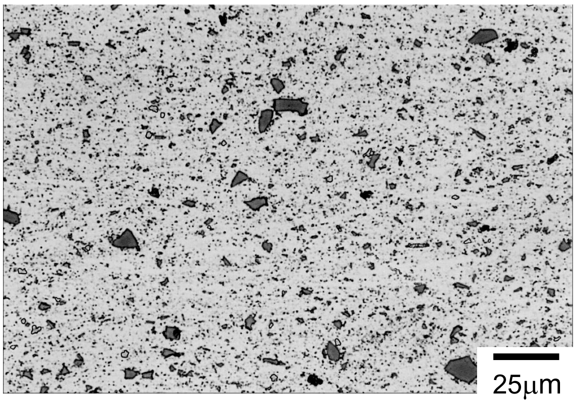

Fig.2 Optical micrograph of MBL05 alloy.

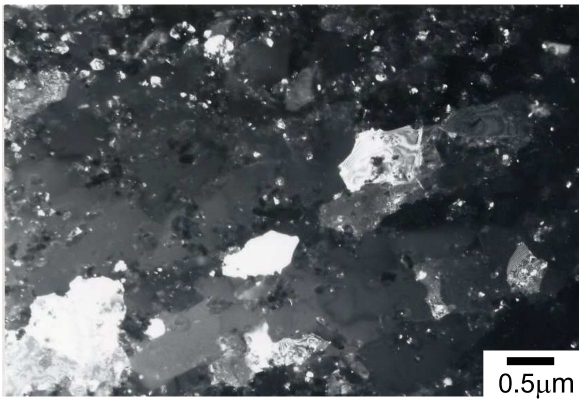

Fig.3 TEM micrograph of MBL09 alloy.

Fig.4 shows SEM (Scanning Electron Microscope) micrograph of the aluminum carbide particle, formed by mechano-chemical reaction during the MA. These particles are considerably fine (about 200nm in size) and dispersed uniformly in the matrix.

Through MA process, crystal grain refinement and dispersion of aluminum carbide particles are obtained. The strength of the alloys is significantly enhanced by these extremely fine crystal grain and aluminum carbide particles.

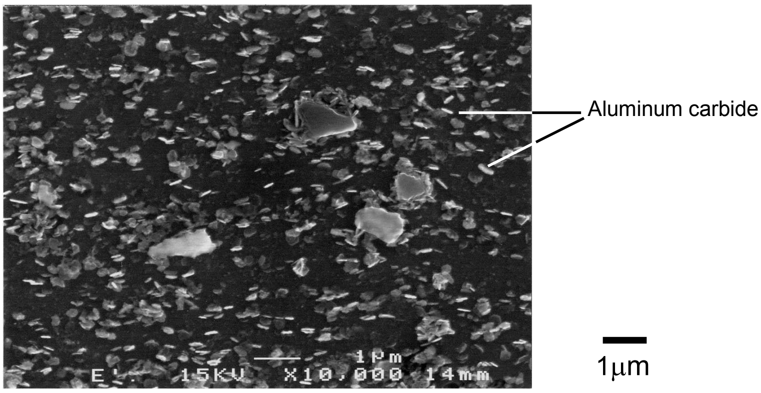

Fig.4 SEM (Scanning Electron Microscopy) micrograph of aluminum carbide particles in MBL09 alloy.

4.3 Tensile properties

Fig.5 shows 0.2% proof stress and tensile strength of the three high-strength B-Al alloys at various testing temperature levels up to 300°C. All the alloys have 0.2% proof stress of over 250MPa at room temperature (R.T.), and over 100MPa at 250°C. These strength levels are twice as high as that of high-toughness B-Al alloy. Thus, the alloys have been proved to have enough strength as basket material of MSF casks for high burn-up spent fuel. Note that the B_4C content have little influence on the strength of the alloy.

4.4 Toughness

Charpy lateral expansion is one of the indices for toughness, and the level of over 0.5mm is required to register an alloy as ASME Code Case. Fig.6 shows lateral expansion in Charpy impact tests on MBL05, MBL07 and MBL09 alloy. Although toughness is lowered with increase of the B_4C content, all the alloys meet Charpy lateral expansion level of over 0.5mm (at -40˚C). Since this criterion for toughness “lateral expansion of over 0.5mm” is not directly related to fracture of a basket, fracture toughness tests were carried out on MBL09 alloy. MBL09 alloy is considered to be the most brittle according to the results of Charpy impact tests. The result showed that the K_{IC} value of MBL09 is 40.9MPa・$m^{0.5}$ (at -40˚C), and this value is comparative to that of other high-strength wrought aluminum alloys without B_4C[2] as shown in Table 2.

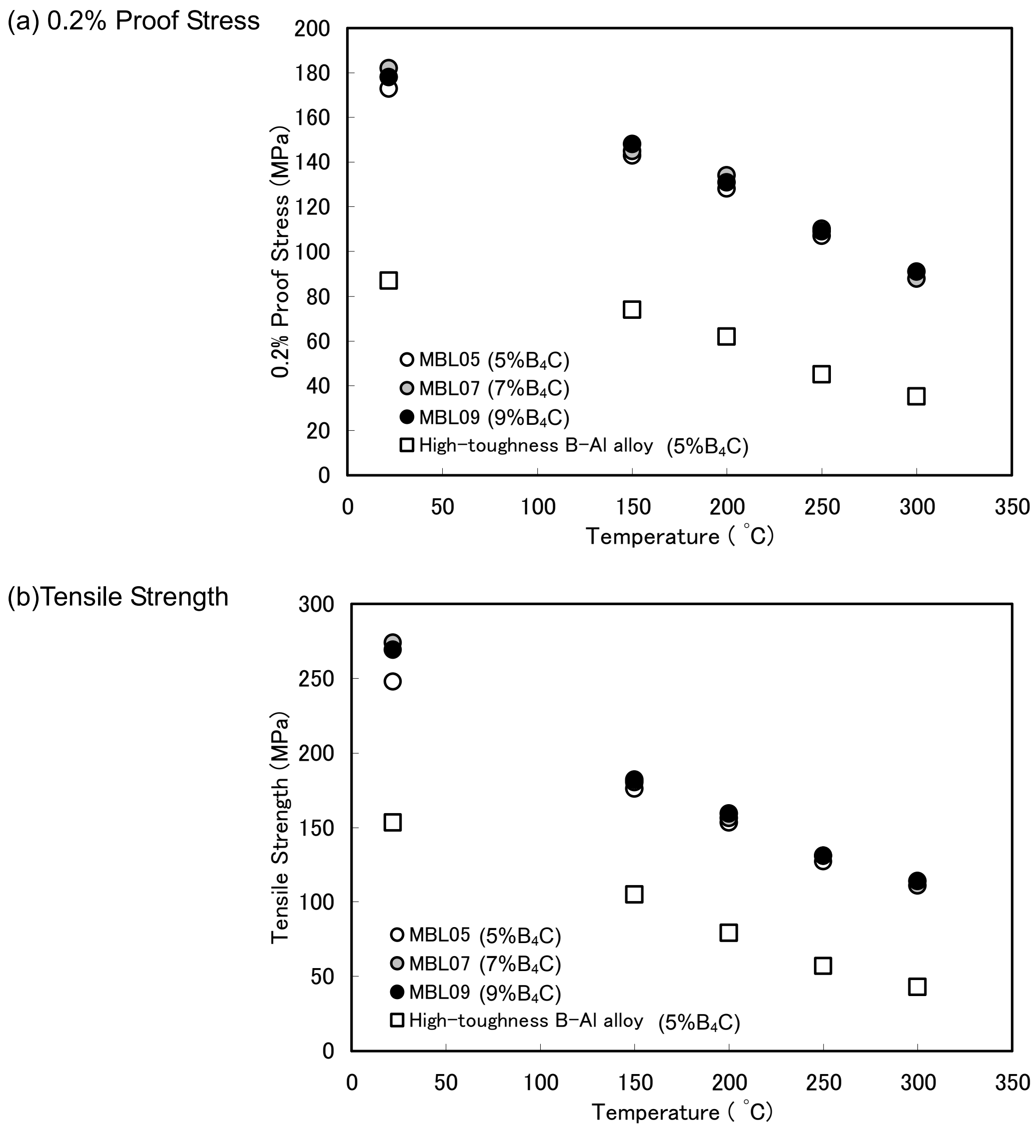

Fig.5 0.2% proof stress and tensile strength of high-strength B-Al alloys

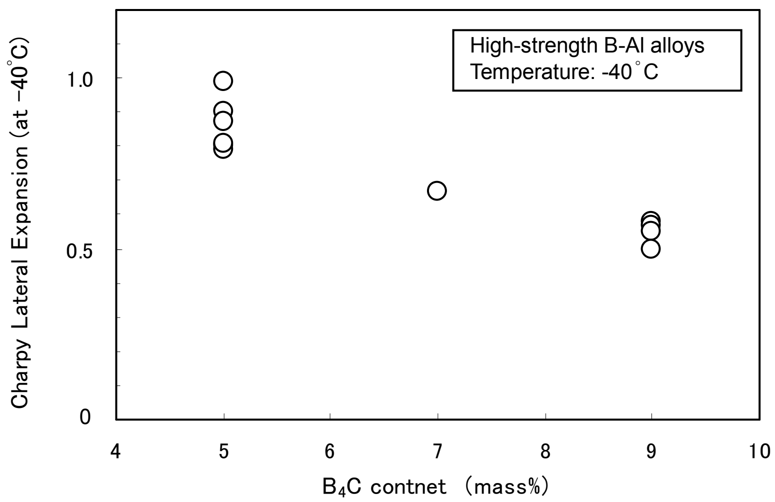

Fig.6 Dependence of Charpy lateral expansion of high-strength B-Al alloy on B_4C content (tested at -40˚C)

Table 2 Facture toughness of several aluminum alloys

Alloy	Fracture Toughness K_{IC} (MPa・$m^{0.5}$)
High-strength B-Al alloy (MBL09)	40.9
AA7075-T651	28.6
AA2024-T351 (super duralumin)	34.0
High-toughness B-Al alloy	67.2

4.5 Stability of mechanical properties against heat treatment

In general, all the age hardened high strength aluminum alloys such as AA6061, AA2024 and AA7075 show decease in strength called “over aging” after heat treatment at high temperature, due to coarsening of precipitates by heat treatment. Accordingly, a boron containing alloy produced from such alloys is not applicable to the application for spent fuel cask, in which temperature is high enough to bring “over aging”. In contrast, the developed high-strength B-Al alloys can maintain its strength even after a long term use at temperature of over 200˚C, because the alloy is strengthened by particles of thermally stable aluminum carbide (Fig.4). The carbide particles maintain strength at higher temperature not due only because they are thermally stable, but also because the particles prevent recrystallization coarsening of the fine crystal grains by pinning effect on grain boundaries.

Table 3 shows comparison of mechanical properties before and after the heat treatment for 1600h at 300˚C. This heat treatment is estimated to be comparable to heat treatment at 250˚C for 30 years according to the Larson-Miller parameter. Apparently, the strength and toughness of high-strength B-Al alloy are not affected by the heat treatment. Fig.7 shows strength of AA6061-T6 and MBL05 after heat treatment at 250˚C for sufficiently long duration for strength to saturate. T6 heat treated AA6061 shows considerable strength decrease after heat treatment at 260˚C. For that reason, the high-strength B-Al alloy is much more suitable for the application to the basket in spent fuel casks than the age hardened high strength aluminum alloys.

Table 3 Mechanical properties of a high-strength B-Al alloy (5mass%B_4C alloy) before and after heat treatment at 300°C for 1600h

Test	Mechanical Properties	Before Heat Treatment	After Heat Treatment
Tensile Test	0.2% Proof stress (MPa)	184 (R.T.) 125 (250°C)	188 (R.T.) 121 (250°C)
	Tensile strength (MPa)	264 (R.T.) 144 (250°C)	265 (R.T.) 140 (250°C)
	Elongation (%)	14.5 (R.T.) 14.0 (250°C)	20.0 (R.T.) 14.5 (250°C)
Charpy Impact Test	Lateral Expansion (mm)	0.78 (-40°C)	0.88 (-40°C)

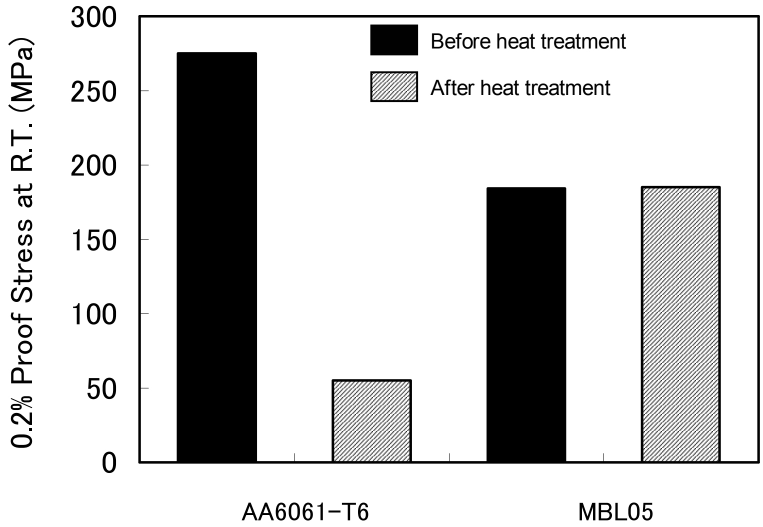

Fig.7 Stable strength of MBL05 and AA6061-T6 after heat treatment at 260°C
(Experimental data for AA6061-T6, estimated value for MBL05 from the data at 300°C)

4.6 Thermal conductivity

The high-strength B-Al alloys are required to have high thermal conductivity equal to or higher than that of other conventional aluminum alloys. Table 4 shows the result of thermal conductivity measurements of the high-strength B-Al alloys [1]. Thermal conductivity of the high-strength B-Al alloys is lower than that of the high-toughness B-Al alloy by about 15%, but much the same as that of AA6061.

Table 4 Thermal conductivity of the high-strength, high-toughness B-Al alloys and AA6061

Alloy	Thermal Conductivity ($W \cdot m^{-1} \cdot K^{-1}$)	
	R.T.	300°C
High-strength B-Al (MBL05)	163	161
High-strength B-Al (MBL09)	169	162
AA6061-T6 [2]	167	-
High-toughness B-Al (5% B_4C)	197	173

5. Summary

The development and research has demonstrated that the high-strength B-Al alloys satisfy the requirements for MSF high-burn-up spent fuel casks. Enhanced strength by fine crystal grain and particles of aluminum carbide is the strong point of the developed alloys. The main characteristics of the high-strength B-Al alloys are summarized as follows:

(1) B_4C content of 5-9 mass% (3.6-6.5 mass% as boron)

(2) Strength is twice as high as that of the high-toughness B-Al alloy

(3) Strength and toughness are stable during long duration at high temperature

Mitsubishi Heavy Industries, Ltd. is planning to apply these high-strength B-Al alloys to the baskets in MSF casks for high-burn up spent fuel casks.

References

[1] Y. SAKAGUCHI, T. SAIDA, T. MATSUOKA, S. KURI, K. OHSONO, S. HODE: Proceedings of the PATRAM2001 (2001)

[2] Japan Aluminum Association: Aluminum Handbook (1994)