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DEVELOPMENT AND RELIABILITY VERIFICATION OF ALUMINUM ALLOYS FOR BASKET OF TRANSPORT AND STORAGE CASK FOR SPENT NUCLEAR FUEL

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ABSTRACT

Long-term reliability of basket material "N-673" for transport and storage cask has been evaluated, taking effect of temperature and irradiation during storage into consideration. Creep tests, thermal aging tests and neutron irradiation tests were conducted, using test pieces from full-scale basket. Consequently, it was confirmed that strength of N-673 is not significantly affected by thermal aging during long-term storage. Neutron irradiation of 10^{16}n/cm^2 at room temperature also does not affect mechanical properties. And creep strain is negligibly small at basket temperature.

1. INTRODUCTION

Aluminum is a highly desired material for basket in spent nuclear fuel casks, for its small density and high thermal conductivity. These characteristics make huge contribution to realization of lightweight, large payload cask. Therefore, various sorts of aluminum-based materials have been proposed. Most of them are produced through conventional ingot metallurgy (casting process). However, there are two problems in application of ingot metallurgy to basket material. One is segregation of boron compound due to density difference between aluminum and boron compound, and the other is loss of ductility by formation of brittle intermetallic compounds in the material over melting point. In order to solve these problems completely, Mitsubishi Heavy Industries, Ltd. has developed basket material by applying powder metallurgical process without using casting process, and registered it as ASME Code Case N-673. "N-673" is produced from powder of a commercial 6000 series aluminum alloy and boron carbide (B_4C), and usually supplied in the form of extruded tube profiles. The maximum boron content is 9mass%, and its distribution is quite homogeneous as shown in Figure 1 due to powder metallurgical process. In spite of such high B_4C content, N-673 has good impact properties. For instance, 5mass%B_4C has Charpy lateral expansion of 1.5-2.2mm at temperature range of -100 to 300˚C, N-673 with, exceeding the minimum requirement of 20mils (0.508mm) by ASME Sec.III Division 1 NG.

The authors have already been reported basic mechanical, physical and metallographic characteristics of the material in its initial state soon after production[1]. However, application of the new materials to actual cask basket requires evaluation of long-term change in material characteristics caused by severe condition during storage, which include mechanical load, elevated temperature and neutron irradiation. Therefore additional researches have been done for the sake of verification of long-term stability of N-673.

For this study, 14 charges of materials were produced by changing B_4C content (2.5, 3, 3.5, 5, 7 and 9 mass%) and three kinds of matrix composition within allowance of the composition range described in the Code Case. These materials were prepared through completely same procedure as actual basket production. These basket tubes have square cross section with wall thickness of 5.5mm or 15mm, and inner hollow of 151mm square.

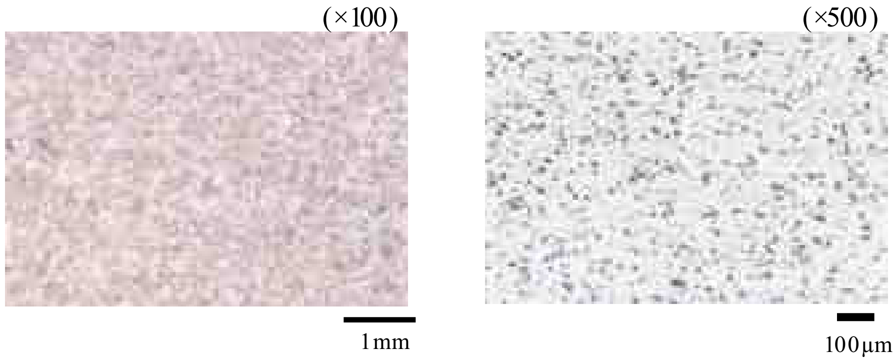


Figure 1 Microscopic boron distribution in N-673 (7%B_4C)

2. EFFECT OF THERMAL AGING ON MECHANICAL PROPERTIES

According to result of thermal assessment of MSF-57B cask with a heat power of 48.8kW, maximum temperature of basket reaches 230˚C during storage. Stability of mechanical strength has to be evaluated in detail in order to use aluminum alloys at this temperature level. Generally, aluminum alloys for structural components are used after strength enhancement by age-treatment such as T4(solution treatment, quench and then aging at room temperature(R.T.)) or T6(solution treatment, quench and aging at higher temperature than R.T.). However, strength of age-treated aluminum alloys significantly decreases when held at elevated temperature, finally to reach initial strength before the age treatment. This is called “over aging” caused by microstructural change during the holding. Specifically, fine precipitates, formed during age treatment and greatly contribute to strength, rapidly grow coarser at higher temperature, and gradually lose its strengthening effect.

On the other hand, strength decrease does not significantly occur in the N-673 basket material, since it is used without any age-treatments to obtain fine precipitates for strengthening.

Effect of thermal aging on mechanical properties was investigated by conducting tensile tests after heat treatment at 200, 250 and 300˚C for 5000 or 20000hours, and comparing the results with those of the materials before heat treatment. Heat treatment at 300˚C for 5000hours is equivalent to heat treatment for 60 years at 255˚C, according to Larson-Miller parameter (LMP) with the constant of 20, which is considered applicable parameter for evaluation of accelerated aging behavior of 6000 series aluminum alloys[2]. The tensile tests were carried out at 200, 250 and 300˚C for evaluation at actual basket temperature.

Figure 2 shows 0.2% proof stress at various temperature levels. Heat treated materials have slightly lower 0.2% proof stress compared to as-extruded material at 200 ˚C. This small decrease is caused by coarsening of extremely fine precipitates inevitably formed after hot extrusion. On the other hand, 0.2% proof stress is consistent from condition to condition at 250 and 300˚C. The reason for this behavior can be explained that tensile test since the test pieces are held at test temperature for 30minues and over. Consequently, it is concluded that the B-Al basket material has enough performance in terms of thermal aging.

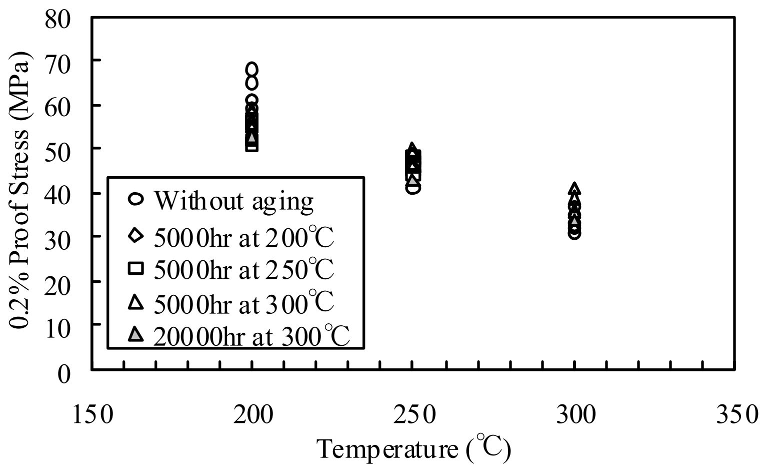


Figure 2　　Effect of thermal aging on strength of N-673 (3-9%B_4C)

3. CREEP AND CREEP RUPTURE PROPERTIES

As basket temperature is high enough to cause creep deformation, it is also essential to evaluate creep properties. Creep and creep rupture tests of N-673 were conducted at 150, 200, 250 and 300˚C in air, with stress range from 17 to 57MPa. For these tests, seven charges of the materials with different B_4C contents (2.5, 3, 3.5, 5 and 9mass%) and different chemical compositions are used.

Figure 3 shows a stress-rupture plot of the N-673. Rupture strength shows fine linear relationship with LMP. As indicated on the horizontal axis, LMP equivalent to 60 years at 230˚C (highest temperature in basket assembly) is included in range of the testing data. Thus, estimation of creep strength for the assumed storage period and temperature is possible by means of interpolation through these data. Larson-Miller parameter curve of creep rupture strength is shown in Figure 4. Average creep rupture strength for rupture in 10^5 hours and 60 years was estimated based on the creep test data using following formula, given by linear regression of the creep rapture data in Figure 4.

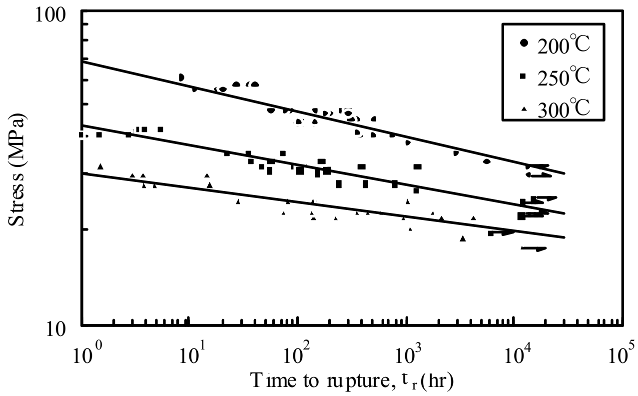
$$\log \sigma = 2.97 - 0.125 \times 10^{-3} \mathrm{LMP}$$

where σ is stress, LMP is Larson-Miller parameter, LMP=T ($\log(t_r)$ + 20).

(T : Temperature (K)　t_r : Time to rupture (hour)　σ : Stress (MPa))

The result are shown in Table 1, The minimum stress by which this N-673 material ruptures in 60 years is 19MPa at 250˚C, 28MPa at 200˚C and 40MPa at 150˚C. This result proves creep rupture of basket does not occur during storage, since stress applied to the basket is estimated not to exceed 1MPa in the case of MSF casks.

Figure 5 shows relationship between stress and minimum creep rate. Minimum creep rate $\dot{\varepsilon}$ (percent per hour) is plotted as LMP=T(20-log$\dot{\varepsilon}$). According to this plot, minimum creep rate under stress of 1MPa is calculated to be less than 10^{-20}%/hour and negligibly small. Hence it is estimated that deformation of basket during storage is practically zero.



Symbols with arrow indicate interrupted tests before rupturing.

Figure 3 Stress-rupture plot for B-Al basket material (3-9%B_4C)

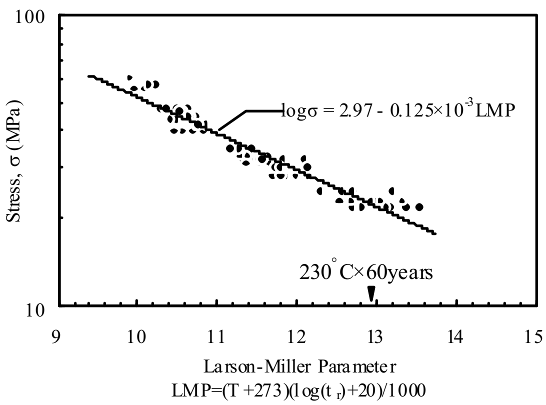


Figure 4 Larson-Miller parameter curve of creep rupture strength for N-673(3-9%B_4C)

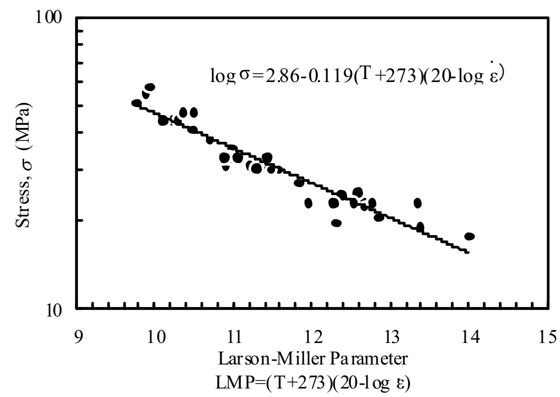


Figure 5 Larson-Miller parameter curve of minimum creep rate for N-673(3-9%B_4C)

Table 1 10^5hours and 60 years rupture strength of N-673

Temperature (°C)	10^5hours rupture strength (MPa)	60 years rupture strength (MPa)
150	44	40
200	31	28
250	22	19

4. EFFECT OF NEUTRON IRRADIATION ON MECHANICAL PROPERTIES

For an aluminum alloy with boron addition, embrittlement or loss of ductility can arise from two factors under neutron irradiation. One is irradiation induced defects formed in aluminum matrix, and the other is accumulation of helium gas on grain boundary or interface between aluminum and boron compound. However, no investigation has been made for irradiation embrittlement of this kind of material. Even limited to boron-free aluminum alloy, there are few reports on effect of neutron irradiation on mechanical properties. But it is known that change in tensile properties of 6061-O aluminum alloy appears only when irradiation of neutron fluence reaches 10^{21}n/cm^2 and over[3].

Considering actual fuel condition, initial neutron flux from certain BWR type spent nuclear fuels in the basket is estimated to be 2.6×10^{6} n/s/cm^2. Hence an estimation of fluence of the basket during storage is given as follows, assuming the initial flux lasts for 60 years (approximately 1.89×10^{9} seconds).

$$\phi = (2.40\times10^{6}\ \mathrm{n/s/cm^2}) \times (1.89\times10^{9}\mathrm{s})$$
$$= 4.54\times10^{15}\mathrm{n/cm^2}$$

Thus, fluence of basket is below 10^{16}n/cm^2 at the outside, and this is far smaller than 10^{21}n/cm^2, at which changes in mechanical properties of 6061-O aluminum alloy appear. Chemical composition of matrix of N-673 and 6061-O is quite similar, and it is considered that matrix of N-673 basket material does not show change in mechanical properties during storage period. However, it should be noted that 6061-O does not contain any boron and embrittlement by helium formation is not appeared in the data.

In order to confirm effect of boron, tensile tests of irradiated N-673 were carried out. Tensile tests were carried out after irradiation of approximately 10^{16}n/cm^2. N673 with 9%B_4C were selected by its high boron content, for which most likely to susceptible to helium embrittlement. Neutron fluence for this test is approximately 10^{16}n/cm^2 which is higher than an assumed fluence for 60 years in basket of a BWR cask. Neutron irradiation was performed at room temperature using a test reactor YAYOI, the University of Tokyo.

Figure 5 shows tensile strength, 0.2% proof stress and elongation before and after irradiation. It is possible that 0.2% proof stress and tensile strength of the irradiated material are slightly higher than those of the non-irradiated material, but no significant difference appeared. Obviously there is no significant difference in elongation as well. Since effect of helium embrittlement usually appears as decrease in elongation, it is concluded that N-673 does not show helium embrittlement at this fluence.

In an actual cask, basket material is irradiated at elevated temperature up to 230°C. Meanwhile, this irradiation test was carried out at room temperature due to limitation of the irradiation facility. However, mechanical property change of matrix caused by irradiation-induced defect formation is greater at room temperature than at elevated temperature, since the defects quickly recover at elevated temperature by accelerated atomic diffusion. For instance, according to result of a research on radiation-induced defect formation by electrical resistivity measurement[4], it was suggested that defects induced by irradiation at 4.2K completely recover during annealing at 300K after irradiation. This means irradiation-induced defects extinct with increase in temperature. For this reason, it is considered that the data obtained by irradiation at room temperature are valid for evaluation of embrittlement of aluminum matrix.

In this study, effect of embrittlement caused by helium accumulation on grain boundary or other interface could not be experimentally verified at elevated irradiation temperature. However, at irradiation temperature lower than 30% of melting point(K), low mobility of irradiation defect makes helium accumulation on grain boundary difficult[5]. Considering that irradiation temperature of basket is approximately 25% of the melting point of matrix alloy, it is speculated that helium embrittlement behavior is not greatly differ from that of test result at RT obtained here.

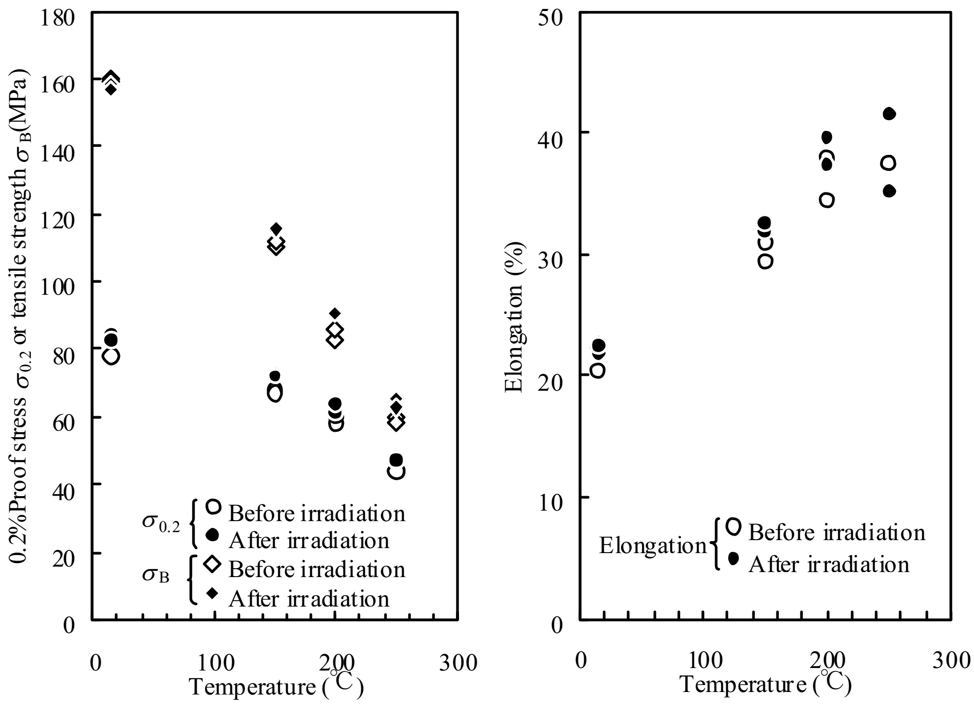


Figure 6 Tensile test result of N-673 with 9wt%B_4C before and after irradiation of 10^{16}n/cm^2

5.CONCLUSIONS

For the sake of long-term integrity evaluation of basket material N-673, creep tests, thermal aging tests and neutron irradiation tests were conducted. The results are summarized as follows:

(1)Strength of N-673 is not significantly affected by thermal aging during long-term storage.

(2)Neither creep rupture nor creep strain large enough to lead loss of subcriticality occurs under assumed stress in basket.

(3)Neutron irradiation of 10^{16}n/cm^2 at room temperature does not affect mechanical properties of N-673.

These research results exemplify that N-673 has fine long-term integrity as a basket material, and currently the application of the material to actual cask is underway.

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