Proceedings of the 18th International Symposium on the
Packaging and Transportation of Radioactive Materials
PATRAM 2016
September 18-23, 2016, Kobe, Japan

Paper No. 1059

Development of Aluminum Boron Carbide Neutron Absorber Structural Material

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Abstract

Neutron absorber is a material used as a divider to prevent the criticality between fuel assemblies in dry cask storage and/or transportation. Until recently, stainless steel/boron carbide neutron absorbers have been widely used as both neutron absorbers and structural material for casks due to stainless steel's reliable tensile strength and proof stress. However, spent fuel nowadays tend to have higher burn-up than before, and this trend has led to higher requirements for heat removal that cannot always be achieved by traditional stainless steel structure. This has made aluminum/boron carbide neutron absorbers, which have high thermal conductivity, more desirable for dry cask storage.

Up to now, the main aluminum/boron carbide neutron absorber used for both neutron absorption and structural material in dry casks has been based on Al-Mg-Si system alloy (6000 series aluminum alloy) since it has excellent extrudability and mechanical properties. However, it has been found that Al-Mg-Si system alloy's tensile strength and proof stress dramatically deteriorate over time due to the over aging effect, which can limit its future applicability in casks with high burn-up spent fuel. To improve aluminum/boron carbide neutron absorber structural material, an extruded neutron absorber made by powder metallurgy was developed that uses a specially dedicated Al-Mn system alloy (3000 series aluminum alloy) combined with boron carbide, whose tensile strength and proof stress remain stable over time even after being exposed at high temperature for a long time. This neutron absorber structural material is a metal matrix composite (MMC) formed by extruded billets that are prepared by cold isostatic pressing (CIP) and sintering. In order to get the best possible results from the Al-Mn system alloy, its chemical composition was enhanced and the parameters of each manufacturing process, from CIP to extrusion, were fine tuned. This paper shall show how the Al-Mn system alloy is the better candidate over Al-Mg-Si system alloys for neutron absorber structural material.

Introduction

A typical structure for dry casks is shown on the left side of Figure 1. The fuel is inserted into a cell whose walls are made from three different kind of material: stainless steel, aluminum/boron carbide, and an aluminum filler. The stainless steel is the structural element that provides strength; the aluminum/boron carbide sheet provides neutron absorption capability and helps maintain sub-criticality within the cask; the aluminum filler has high thermal conductivity and helps dissipate heat out of the cask. The concept of a single aluminum/boron carbide material, as shown on the right side of Figure 1, consists in replacing three materials by one, that can still meet the three required functions – strength, sub-criticality, and heat transfer.

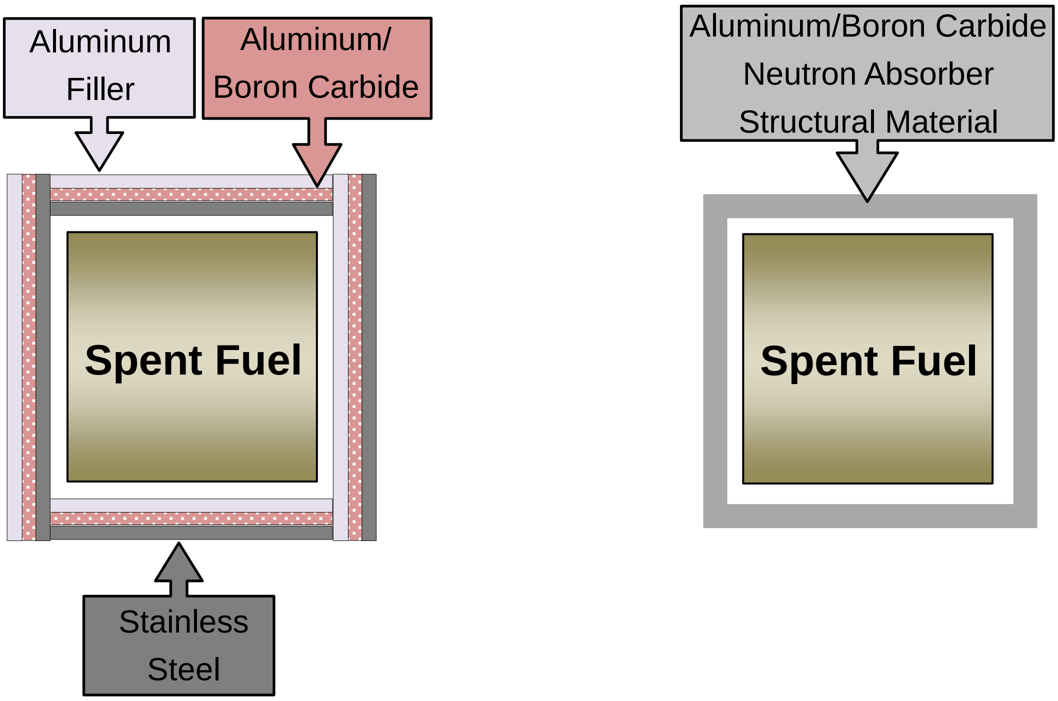

Figure 1 Typical structure of a dry cask (left) and new concept (right)

Current solutions often use Al-Mg-Si system alloy (6000 series aluminum) as the matrix that the neutron absorber (typically boron carbide) is uniformly dispersed within. However, it has been found that such material, despite having high mechanical properties in its “as-extruded” state, showed much lower properties after being exposed to high temperature, which makes its use problematic in future.

The purpose of this development was to find an alloy which gives the neutron absorber material high mechanical properties that do not deteriorate over time at high temperatures.

Comparison of 6061 and 3004 aluminum alloys

The chemical composition of one typical Al-Mg-Si system alloy (6000 series) used as a matrix for neutron absorber structural material is shown in Table 1. The main precipitate conferring strength to the material is Mg_2Si.

Table 1 also shows the chemical composition of standard Al-Mn system 3004 alloy. The presence of manganese of up to 1.5% helps increase the mechanical strength stability over time at high temperature thanks to the presence of Al-Mn compounds.

Table 1 Chemical composition of 6061 aluminum and 3004 aluminum (unit: mass%)

	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti
6061	0.4-0.8	≤ 0.7	0.15-0.40	≤ 0.15	0.8-1.2	0.04-0.35	≤ 0.25	≤ 0.15
3004	≤ 0.3	≤ 0.7	≤ 0.25	1.0-1.5	0.8-1.3	-	≤ 0.25	-

The microstructure of both 6061 aluminum and 3004 aluminum is shown on Figure 2.

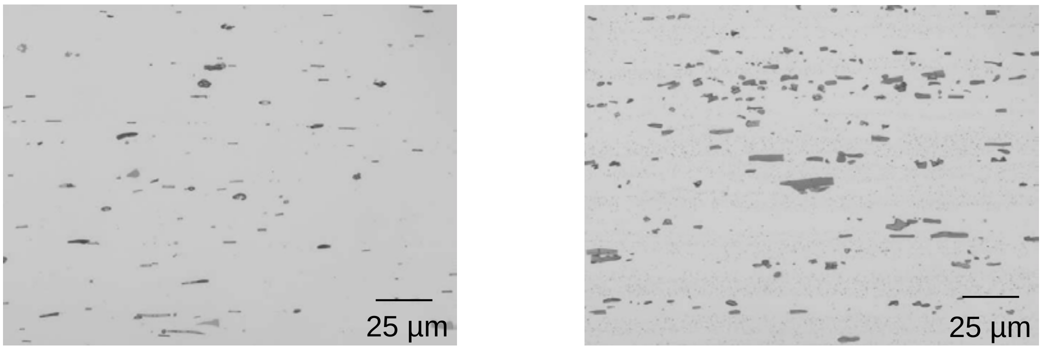

Figure 2 Microstructure of 6061 aluminum (left) and 3004 aluminum (right)

The difference of mechanical behavior between 6061 aluminum and 3004 aluminum [1] is shown on Figures 3 to 5.

The X-axis is the Larson-Miller Parameter (LMP), which is a function of temperature and time for an isothermal condition, as defined by the following formula: LMP = (T+273.15)*(C+log(t)) where T is the exposure temperature (°C), t is the time before failure (hours), and C is a constant which is typically equal to 20 in the case of aluminum. The Y-axis shows the 0.2% proof stress of each alloy in MPa.

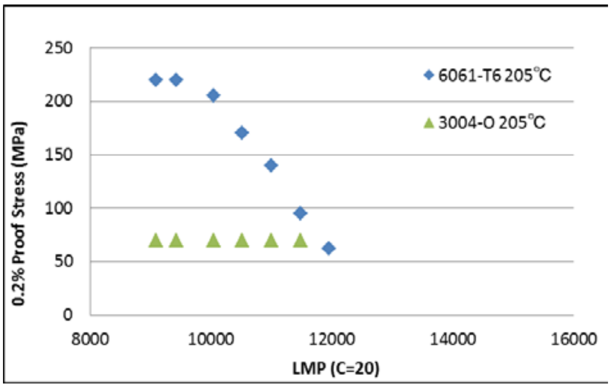

Figure 3 Reference data [1]: 0.2% proof stress in function of LMP at 205°C

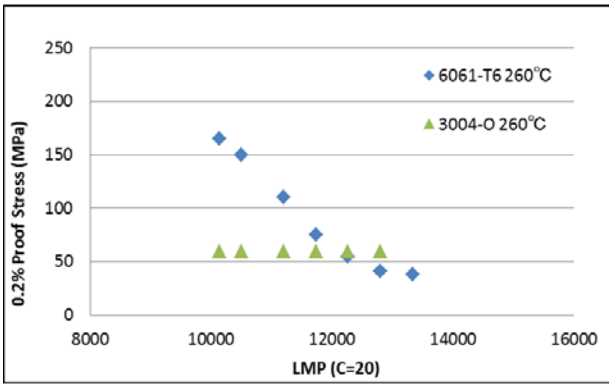

Figure 4 Reference data [1]: 0.2% proof stress in function of LMP at 260°C

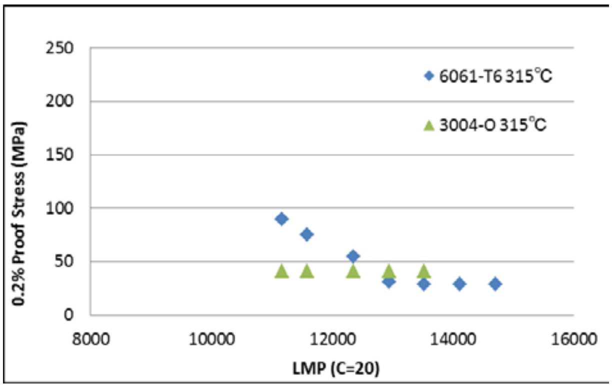

Figure 5 Reference data [1]: 0.2% proof stress in function of LMP at 315°C

Although 6061 aluminum has a higher strength than 3004 aluminum in its "as-extruded" condition, i.e. when the LMP is low, it becomes clear that 3004 aluminum benefits a much better stability over time at high temperature, and its strength eventually exceeds that of 6061 aluminum after long-term high-temperature exposure.

In order to corroborate these literature data, experiments have been conducted internally and gave similar results, as shown on Figures 6 to 8.

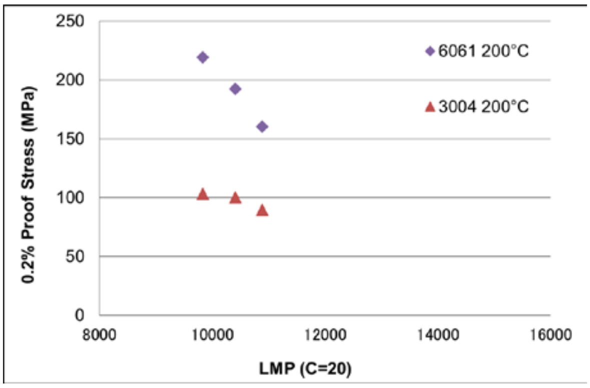

Figure 6 Experimental data: 0.2% proof stress in function of LMP at 200°C

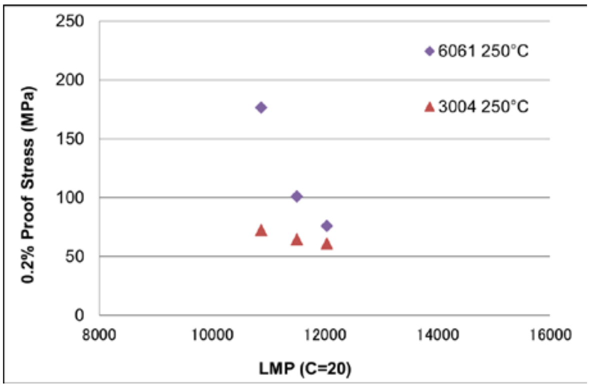

Figure 7 Experimental data: 0.2% proof stress in function of LMP at 250°C

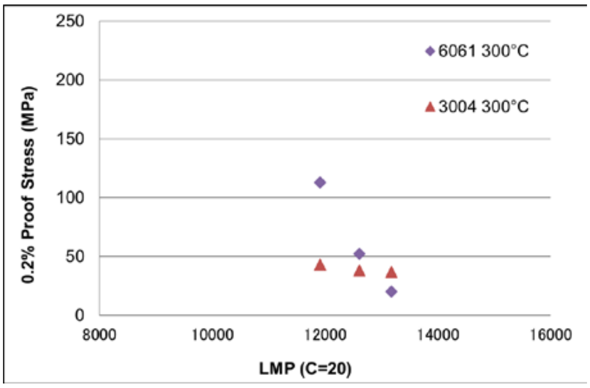

Figure 8 Experimental data: 0.2% proof stress in function of LMP at 300°C

In order to improve the stability over time at high temperature of standard 3004 aluminum's mechanical properties even more, a specifically enhanced alloy was developed. Chemical composition is shown in Table 2. Chemical compositions of 6061 aluminum and standard 3004 aluminum are also shown to allow for easier comparison.

Table 2 Chemical composition of enhanced 3004 aluminum (unit: mass%)

	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti
6061	0.4-0.8	≤ 0.7	0.15-0.40	≤ 0.15	0.8-1.2	0.04-0.35	≤ 0.25	≤ 0.15
3004	≤ 0.3	≤ 0.7	≤ 0.25	1.0-1.5	0.8-1.3	-	≤ 0.25	-
Enhanced 3004	≤ 0.5	0.3-1.0	0.4-0.8	1.5-2.5	1.0-1.8	≤ 0.30	≤ 0.20	≤ 0.01

In this new enhanced 3004 aluminum, Mg has been increased by an absolute 0.2 to 0.5 mass%, and Cu has been increased by an absolute 0.15 to 0.55 mass%, to support higher mechanical strength. Fe and Mn have also been increased, respectively by an absolute 0.3 mass%, and 0.5 to 1.0 mass%, to support better thermal stability of mechanical properties over time at high temperature, especially per the presence of Al-Fe-Mn compounds (e.g. $Al_6(Fe, Mn)$) and Al-Fe-Mn-Si compounds (e.g. $Al_{12}(Fe, Mn)_3 Si$).

Extrusion trial and results

In order to observe the actual mechanical behavior of the new material, a concentration of 10.5 volume% (i.e. 9.8 mass%) of boron carbide (B_4C) was introduced in the composition, by mixing B_4C powder and enhanced 3004 aluminum powder in a rotating blender. Concentration of each element in the billet as analyzed by ICP is shown in Table 3, except concentration of B_4C, which is a calculated value directly issued from the measured amount of boron.

Table 3 Chemical composition of actual billet No.1 (unit: mass%)

	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	B	B_4C
Billet 1	0.18	0.46	0.46	1.74	1.35	0.002	0.014	0.004	7.77	9.93

Mixed powder was filled into a rubber mold and subject to Cold Isostatic Pressure (CIP) in order to compact the powder and form a billet. The billet was then sintered at high temperature before it was preheated and extruded through a flat-bar shape extrusion die.

Mechanical properties of the material were tested "as-extruded" at eight different temperatures: -40°C, RT (20°C), 150°C, 200°C, 250°C, 300°C, 350°C and 400°C. Mechanical properties were also tested at 150°C, 250°C and 400°C after two different high temperature aging treatments: 350°C-3h and 350°C-100h. Results of tensile strength and 0.2% proof stress are plotted on Figure 9 and Figure 10, respectively. Each plot corresponds to the average value of 3 measurements. A few observations can be made from these graphs:

- In the as-extruded state, no significant difference in strength could be observed within the -50°C / 150°C range of temperature.
- No significant difference in strength could be observed between 3h aging and 100h aging, demonstrating a high thermal stability of the material.
- At 400°C testing, aging has no more influence on the material since there is no difference in strength between the as-extruded state and the 3h and 100h aging states. This makes sense considering the aging treatment was performed at 350°C.

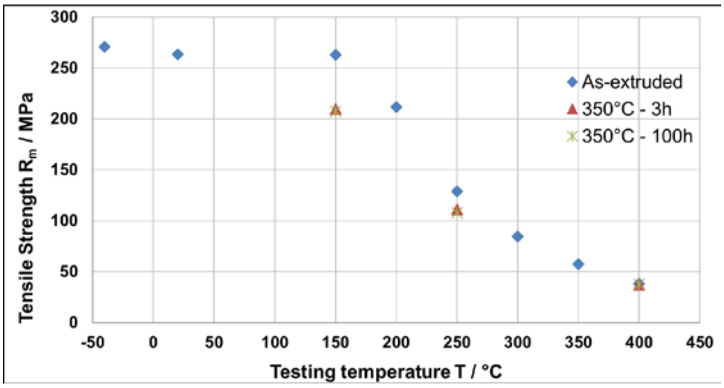

Figure 9 Tensile strength of billet No.1 in function of testing temperature

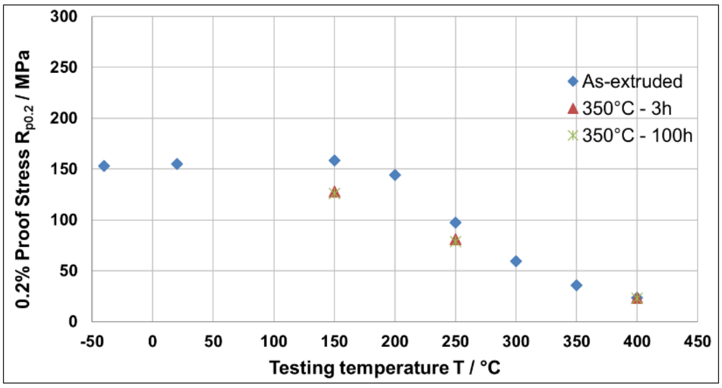

Figure 10 0.2% proof stress of billet No.1 in function of testing temperature

In order to assess the reproducibility of these results, three additional billets were prepared by the same manufacturing process and extruded. The chemical composition of the billets is shown in Table 4. Since the B_4C-Al powder used for the three billets was taken from one lot, only one chemical analysis was conducted.

Table 4 Chemical composition of actual billets No.2, 3 & 4 (unit: mass%)

	Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	B	B_4C
Billet 2	0.10	0.48	0.44	1.76	1.39	0.006	0.004	0.004	7.56	9.66
Billet 3										
Billet 4										

In order to better observe the effect of aging time on the mechanical properties, data were plotted with the aging time (in hours) on the X-axis and a logarithmic scale. Each plot corresponds to the average value of 3 measurements. Although the decrease in strength between the as-extruded condition and the 3-hour aging condition is comprised between 14 to 17%, the decrease is only approximately 1% to 2% between the 3h aging and the 100h aging, which is deemed to be insignificant and proves remarkable thermal stability of the material over time at high temperature.

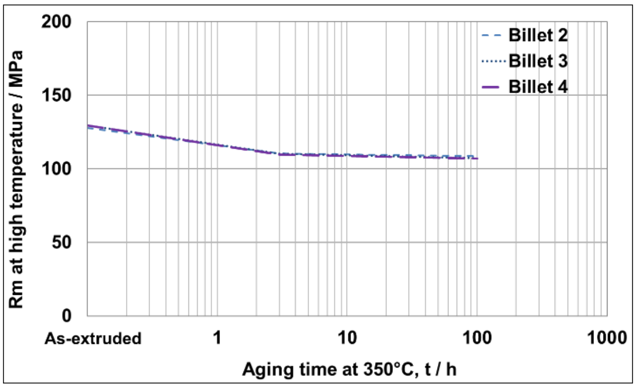

Figure 11 Tensile strength of billets No.2,3 & 4 in function of aging time

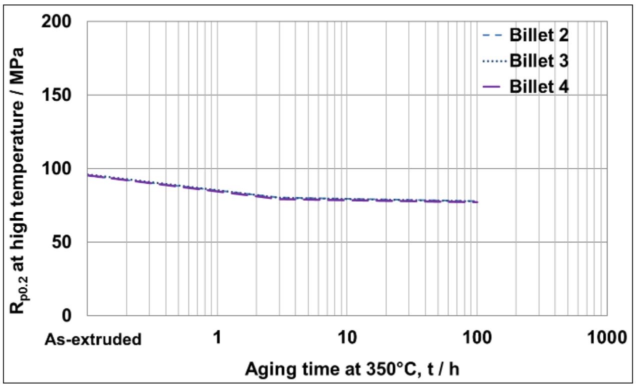

Figure 12 0.2% proof stress of billets No.2,3 & 4 in function of aging time

Conclusions

A new aluminum/boron carbide neutron absorber made by powder metallurgy, containing boron carbide uniformly dispersed in a specifically enhanced 3004 aluminum matrix, has been developed to answer dry cask storage needs. The boron carbide, which has the property to absorb neutrons, prevents over-criticality in the cask, while the aluminum matrix allows for heat transfer and structural strength. This new material features excellent mechanical stability over time at high temperatures, which makes it a groundbreaking candidate for future casks designs in comparison to existing solutions.

Acknowledgments

The authors would like to acknowledge the whole staff who had taken part in this development and made the many extrusion trials and mechanical tests possible.

References

1. J. Gilbert Kaufman, *Properties of Aluminum Alloys: Tensile, Creep and Fatigue Data at High and Low Temperatures*, The Aluminum Association, Inc. and ASM International (1999).