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Development of Aluminum-based Neutron Absorber Material Containing Gadolinium

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Abstract

Neutron absorber materials are used to prevent the criticality of fuel assemblies in dry casks and spent fuel storage racks. Currently, aluminum/boron carbide metal matrix composite (Al/B_4C composite) is used for neutron absorber materials, which can be produced via a powder metallurgy process.

In general, neutron absorbing performance (^{10}B areal density) of Al/B_4C composite is controlled by boron carbide (B_4C) concentration and thickness of the composite. If the thickness is limited by the design, the concentration needs to be increased to ensure the performance. However, this results in reduced ductility of the composite. The addition of another element that has a larger thermal neutron absorption cross section than boron may improve the ductility while keeping the same neutron absorbing performance since the volume fraction of the compound would be lower than using boron alone.

Gadolinium was selected as a candidate element because it has a large thermal neutron absorption cross section and has been used in the nuclear industry. Aluminum/gadolinium oxide metal matrix composite (Al/Gd_2O_3 composite) was manufactured. From the evaluation of this new material, it was found that the amount of particles in the composite could be reduced by using of Gd_2O_3 instead of B_4C when neutron transmission rate is same between Al/Gd_2O_3 composite and Al/B_4C composite. In addition, it was shown that the elongation of the Al/Gd_2O_3 composite is higher than that of the Al/B_4C composite, which has a neutron absorbing performance as high as the developed composite.

The aluminum-based neutron absorber material containing gadolinium, which has high neutron absorbing performance and good formability, was developed for spent fuel storage application.

Introduction

Neutron absorber materials made of aluminum / boron carbide metal matrix composite (Al/B_4C composite), borated aluminum alloy or borated stainless steel are used to prevent the criticality of dry casks and spent fuel storage racks. MAXUS®, one of the Al/B_4C composite for neutron absorber material, is produced via a powder metallurgy process by Nikkeikin Aluminium Core Technology Co., Ltd.

In general, the neutron absorbing performance of Al/B_4C composite, that is ^{10}B areal density, is controlled by B_4C concentration and thickness of the composite. If the thickness is limited by the design, the B_4C concentration is increased to ensure the performance. However, it causes the reduction of ductility of the material [Reference 1]. The addition of gadolinium, which has a larger thermal neutron absorption cross section than boron, may improve the ductility of the material while keeping the same neutron absorbing performance since the volume fraction of the compound would be smaller than the case of using boron. Thus an aluminum-based neutron absorber material containing gadolinium was developed.

Material and manufacturing process

Al-Gd alloy and Al/Gd_2O_3 composite were selected as candidate materials for aluminum-based neutron absorber material containing gadolinium. Al-Gd alloy uses Gd metal for its raw neutron absorbing material, while Al/Gd_2O_3 uses Gd_2O_3 powder. Gd metal and Gd_2O_3 powder have natural abundance of isotope of gadolinium.

The manufacturing processes are shown in Figure 1.

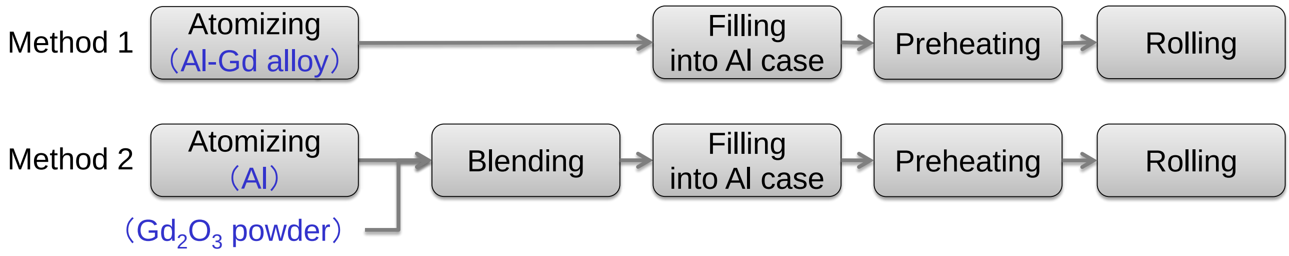


Figure 1. Manufacturing process for the materials

In Method 1, Atomized Al-Gd alloy powder was filled into aluminum alloy cases. Subsequently, those cases were subjected to hot rolling into sheets. In Method 2, pure Al powder and Gd_2O_3 powder were mixed using a blender. The mixed powder was then filled into aluminum alloy cases. Subsequently, those cases were hot rolled into sheets.

All methods produced rolled sheets of 2.5 mm thickness, 150 mm width and 1,000 mm length. Al-8 mass% Gd was manufactured by Method 1, while Al/ 8 mass% Gd_2O_3 composite was manufactured by Method 2.

From these two methods, one was selected based on the manufacturability, uniformity of gadolinium containing particles in the material's matrix (determined through optical microscopic inspection of the cross section), density and relative density[Reference 2] (measured using Archimedes' principle), elongation after fracture (tensile testing), whether the material can be mixed with B_4C, and adjustability of the chemical composition.

Neutron absorbing performance evaluation based on neutron transmission testing

The neutron transmission testing, which uses a research reactor, was performed to evaluate the neutron absorbing performance of the material. The reactor has been used for the evaluation of neutron absorber material containing ^{10}B. The neutron transmission testing measures the number of neutrons that pass through the material from the reactor to the neutron detector (BF_3 neutron detector). The neutrons that are exposed to the material are moderated to the energy level of thermal neutrons by heavy water. Neutron transmission rate, which is the index of neutron absorbing performance, is calculated by number of neutrons that go through the material and the number of neutrons that go through when there is no material.

Composition effects on properties of the Al/Gd_2O_3 composite

After the selection of the manufacturing method, materials containing differing amounts of Gd_2O_3 and B_4C were manufactured and evaluated for their effects on the properties. Table 1 shows all the test materials that were manufactured.

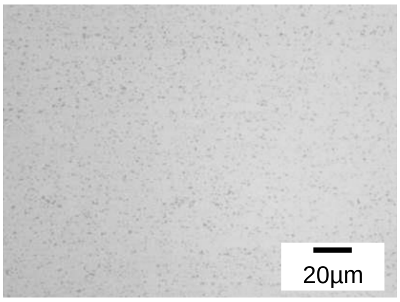
Table 1. Composition of the test materials

	Gd_2O_3 (mass%)	B_4C (mass%)	Al (mass%)	Volume fraction of Gd_2O_3 and B_4C (vol%)
1	8	0	92	3
2	8	20	72	25
3	15	0	85	6
4	10	10	80	15
5	5	20	75	24
6	30	0	70	13
7	20	20	60	32
8	10	40	50	49
9	6	0	94	2
10	4	10	86	12
11	2	20	78	22
12	12	0	88	5
13	4	40	56	43
14	0	0	100	0
15	0	10	90	11
16	0	20	80	21
17	0	30	70	32

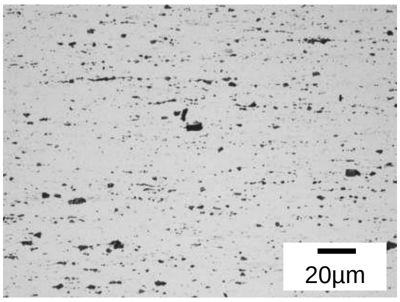
Results and discussion

Results of considering for material and manufacturing

It was found that there were no major issues with either manufacturing processes after testing each one. The microstructures for each method are shown in Figure 2. There were no porosity or no particle clusters found. Further, the particles in (a) were mainly either Al_3Gd or Al_4Gd, while the particles in (b) were Gd_2O_3.



a) Al-Gd alloy



b) Al/Gd_2O_3 composite

Figure 2. Microstructures of Al-Gd alloy and Al/Gd_2O_3 composite

Table 2 compares the composition, particle uniformity, relative density, elongation after fracture, and chemical composite adjustability. The particle uniformity for each manufacturing method was determined from the microstructure evaluation (Figure 2).

Considering that it is easier to adjust the blended amount of materials for manufacturing Method 2 compared with the adjustment of compounds during atomizing for manufacturing Method 1, Method 2 is found to be the better manufacturing process.

Table 2. Comparison of method 1 and method 2

	Composition	Gd-Particle uniformity	Relative density	Elongation	Chemical Composite adjustability
Method 1	Al-8%Gd	Good	100%	25%	Not Easy (Controlled by Atomizing)
Method 2	Al/8%Gd_2O_3	Good	99%	17%	Easy (Controlled by Blending)

Evaluation of neutron absorbing performance by the neutron attenuation testing

Figure 3 shows the relationship between Gd_2O_3 or B_4C concentration and neutron transmission rate, which is the number of neutrons that pass through when there is material divided by the number of neutrons that pass through when there is no material. Figure 3 shows that the amount of particles in the chemical composite could be reduced by using of Gd_2O_3 instead of B_4C when neutron transmission rate is same between Al/Gd_2O_3 composite and Al/B_4C composite.

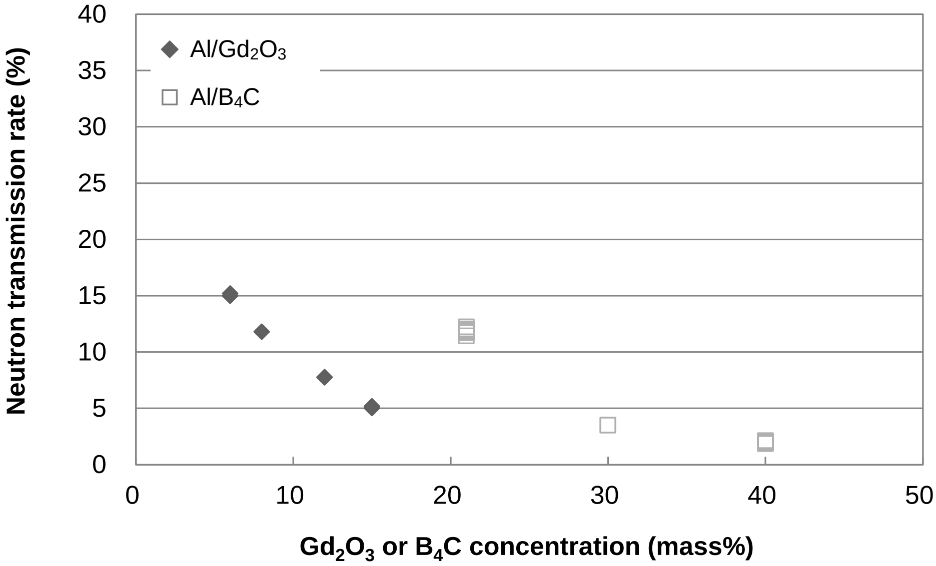
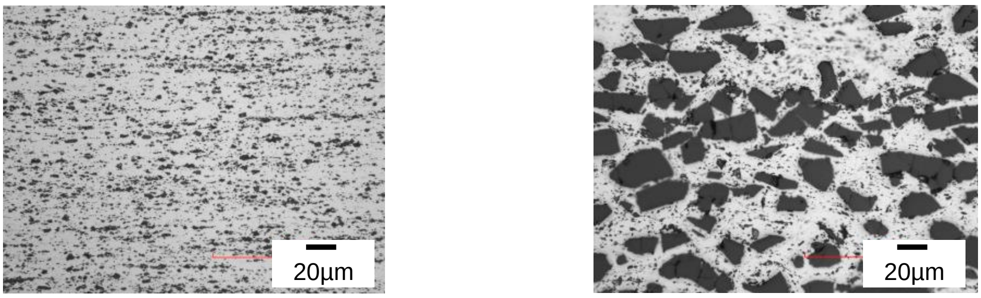


Figure 3. Relationship between Gd_2O_3 or B_4C concentration and neutron transmission rate.

Composition effects on properties of the Al/Gd_2O_3/B_4C composite

It was determined that manufacturing Method 2 is best for neutron absorber material containing gadolinium, so material with different amounts of Gd_2O_3 and B_4C were manufactured. The materials were able to be manufactured and there were no major issues with the material itself. The microstructures of typical materials are shown in Figure 4. None of the materials showed any porosity or particle clustering.



a) Al/30mass%Gd_2O_3 b) Al/20mass%Gd_2O_3/20mass%B_4C

Figure 4. Microstructures of Al/30mass%Gd_2O_3 and Al/20mass%Gd_2O_3/20mass%B_4C composite

Figure 5 shows the relationship between the replacement rate of Gd_2O_3 for B_4C and elongation after fracture for the materials which have the same neutron absorbing performance. The replacement rate is the percentage of Gd_2O_3 when Gd_2O_3 is added to replace the neutron absorbing performance of B_4C with consideration that the neutron absorbing performance of Gd_2O_3 is two times more than that of B_4C. 0% replacement rate is the elongation of Al/B_4C composite, while 100% replacement rate is the elongation of Al/Gd_2O_3 composite. The elongation increases as the replacement rate of Gd_2O_3 rises.

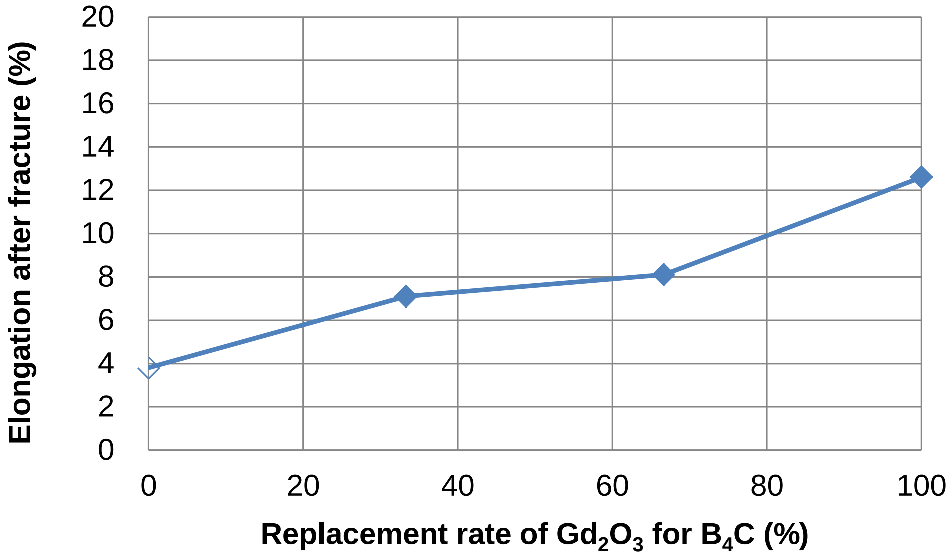


Figure 5. Relationship between replacement rate of Gd_2O_3 for B_4C and elongation

Figure 6 shows the relationship between the concentration of composition and the elongation after fracture. B_4C and Gd_2O_3 are used to calculate the concentration of composition by volume. The lower the concentration, the higher the elongation becomes. It was found that the elongation is adjustable by volume fraction of the B_4C and Gd_2O_3 particles.

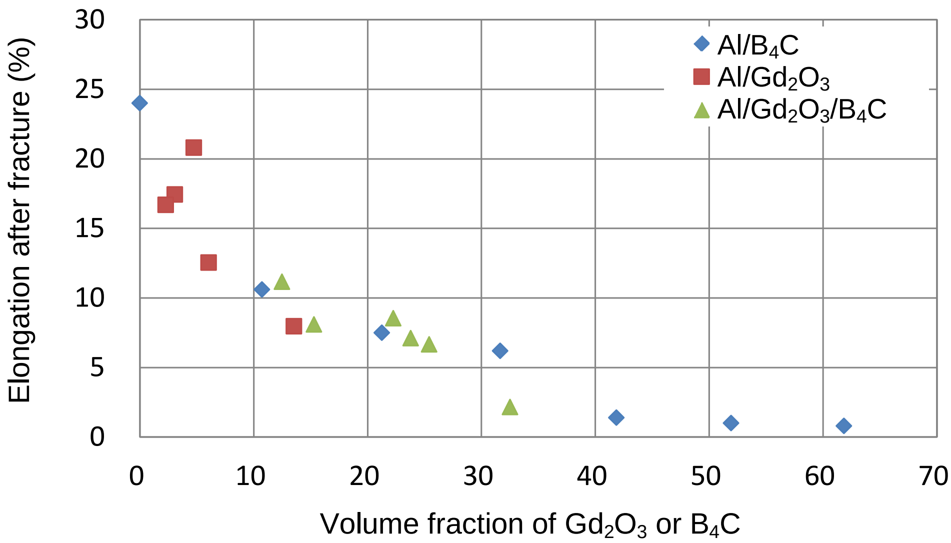


Figure 6. Relationship between volume fraction of Gd_2O_3 or B_4C and elongation

Summary

Neutron absorber material containing gadolinium can have the same effective neutron absorbing performance as boron albeit with a lesser amount. Lower amount of composite material in aluminum-based neutron absorbers allow for higher ductility (elongation after fracture). This means that neutron absorber material containing gadolinium can have better ductility than their equivalent neutron absorbing performing neutron absorber material containing boron.

- It is found that both Al-Gd alloys and Al/Gd_2O_3 composite can be manufactured without any issues.
- Of all the types of manufacturing that were investigated, Al/Gd_2O_3 composite is found to be better from the evaluation.
- Neutron transmission testing has found that the neutron absorbing performance of Al/Gd_2O_3 composite is superior to Al/B_4C composite.
- Al/Gd_2O_3 composite is superior to Al/B_4C for elongation when these materials have the same neutron absorbing performance. Moreover, the volume fraction of Gd_2O_3 and B_4C particles can control the elongation, so a material with an adjusted amount of Gd_2O_3 and B_4C can be manufactured to the required specifications.

Reference

1) Handbook of Neutron Absorber Materials for Spent Nuclear Fuel Transportation and Storage Applications: 2009 Edition. EPRI, Palo Alto, CA: 2009. 1019110, 7-32.

2) ISO/DIS 3252:1998 “Powder metallurgy — Vocabulary”.