

# Helium Implantation into Highly Microstructure-Controlled B4C-based Ceramics

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## III. 3. Helium Implantation into Highly Microstructure-Controlled B<sub>4</sub>C-based Ceramics

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### Introduction

 $B_4C$  pellets have been used as neutron absorbers in control rods of both boiling water reactors (BWR) and fast breeder reactors (FBR). Volume swelling occurs by accumulation of helium bubbles produced by the  ${}^{10}B(n, \alpha)^{7}Li$  reaction<sup>1-3</sup>), which results in failure of a cladding tube due to extensive mechanical interactions between  $B_4C$  pellets and cladding tubes<sup>4</sup>). To extend the lifetime of control rods and then improve safety performance of fast reactors, it is essential to develop the high-performance  $B_4C$  pellets to overcome the above problem. We have synthesized the highly microstructure-controlled  $B_4C$ -based ceramics for neutron absorbers by controlling the microstructure of  $B_4C$  pellet such as particle size, crystal-orientation, pore-diameter, pore-shape and pore-orientation. This highly controlled microstructure could release helium gas produced during neutron absorption without excessive accumulation of helium, and thereby suppress volume swelling. The purpose of this research is to mimic helium generation in a  $B_4C$  pellet by implanting helium ions, instead of neutron irradiation in a fast reactor, and evaluate accumulation and release of helium gas of highly microstructure-controlled  $B_4C$ -based ceramics.

#### **Experimental procedure**

The B<sub>4</sub>C/ carbon nanotube (CNT) composite was used as target sample in this research. Commercial B<sub>4</sub>C and CNT mixed with Al powder, sintering additive, were used as starting materials. Powder mixture with a composition of 85 vol% B<sub>4</sub>C, 10 vol% CNT and 5 vol% Al was pressed into  $22\times35\times1$  mm rectangular plate. The fabrication of B<sub>4</sub>C/CNT

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composite was performed with a hot-press apparatus (FVPHP-R-5, Hi-Multi-5000, Fuji Dempa Kogyo Co., Ltd., Japan) at around a pressure of 60 MPa at 1950°C for 1 hour under Ar gas flow (2 L/min). The <sup>10</sup>B isotopic composition of the B<sub>4</sub>C sample was the natural abundance ratio (19.8%). In addition to the B<sub>4</sub>C/CNT sample, a B<sub>4</sub>C pellet, which had been irradiated with neutrons as a control rod CR0901 of the fast reactor, JOYO, was prepared for comparison. The burnup was estimated about  $80 \times 10^{20}$  captures/cc from calculation with the code HESTIA<sup>5</sup>).

A fabricated B<sub>4</sub>C/CNT sample was bombarded with He ions from a 930 AVF cyclotron of CYRIC. The implantation energy of helium ions was chosen to be 30 MeV. From calculation using the ion transport code SRIM<sup>6</sup>, the implantation depth from the surface is 300  $\mu$ m, deep enough that highly controlled microstructure well forms. A target holder was made for irradiation of a B<sub>4</sub>C-based ceramics as shown in Fig. 1. The B<sub>4</sub>C/CNT sample was set to the target station the course 1 of the first target room and irradiated with 30 MeV He<sup>2+</sup> beam at an average beam current around 1  $\mu$ A for 8 hours. The front surface of the sample was continually cooled with helium gas flow and the target holder was cooled with circulating water during implantation.

Helium gas release behavior of the He-implanted B<sub>4</sub>C/CNT sample and the B<sub>4</sub>C pellet (JOYO) was evaluated with a thermogravimetry mass spectrometer (TG-MS: JMS-Q1500GC, JEOL). Prior to the TG-MS analysis, these samples were pulverized using a B<sub>4</sub>C mortar. In order to evaluate the dependence of the release behavior of helium gas on the grain size, two powder samples of the B<sub>4</sub>C pellet (JOYO) with different grain sizes, about 50-400 and 1-10  $\mu$ m, were prepared (Fig. 2).

## **Results and discussion**

In the helium ion implantation, helium ions were implanted up to  $1.1 \times 10^{17}$  ion/cm<sup>2</sup>, sufficient dose to evaluate the release behavior of helium gas. TG-MS analysis for the B<sub>4</sub>C (JOYO) showed the dependence of the release behavior of helium gas on the grain size (Fig. 3). The helium gas was released promptly at lower temperature in fine powder than coarse powder. Thus, we prepared fine powder from He-implanted B<sub>4</sub>C/CNT sample. Its release behavior of the helium gas had relatively good agreement with that of B<sub>4</sub>C pellet (JOYO) as shown in Fig. 4, but the helium gas release was observed at higher temperature than B<sub>4</sub>C pellet (JOYO).

### Conclusion

In the present work, 30 MeV helium ions were implanted into  $B_4C$ -based ceramics using a He beam from a 930 AVF cyclotron at CYRIC. The release behavior of helium gas in the He-implanted  $B_4C$ -based ceramics was successfully evaluated with TG-MS analysis. It is found that helium gas release rate depends on the grain size, and He-implanted  $B_4C/CNT$  sample showed relatively similar behavior to a JOYO's  $B_4C$  pellet.

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Figure 1. A target holder for irradiation of a B<sub>4</sub>C-based ceramics



Figure 2. The pulverized B<sub>4</sub>C pellet (JOYO) with different grain sizes; (a) coarse powder and (b) fine powder



Figure 3. TGA curves and mass chromatograph of pulverized B<sub>4</sub>C pellet (JOYO) with different grain sizes; (a) coarse powder and (b) fine powder



Figure 4. Mass chromatograph of pulverized B<sub>4</sub>C pellet (JOYO) and He-implanted B<sub>4</sub>C/CNT sample