Effect of Heat Cycling on Microstructure and Thermal Property of Boron Carbide Sintered Bulk as a Shielding Material for Fusion Blanket*)

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In the Force Free Helical Reactor (FFHR) design activity in NIFS, metallic carbides and hydrides are considered as candidate shielding materials for the fusion blankets. These materials are expected to have some advantages on neutronic and thermo-physical properties. In order to promote the blanket design, it is necessary to clarify thermal properties of the candidate materials. We studied microstructure and thermal property of boron carbide (B_4C), which is one of the promising candidates shielding materials, including the effect of heat cycling. By the laser-flash method, thermal diffusivity, which is one of the properties necessary for evaluating thermal conductivity, was measured precisely for B_4C samples. The thermal diffusivity of B_4C around 200 °C decreased to 1/3 (5×10^{-6} m^2S^{-1}) compared with that at room temperature. The sintering density of B_4C bulk was decreased slightly by the thermal cycling. It was suggested that the B_4C bulk has high thermal stability and soundness of microstructure during the life-time of blanket system.

Keywords: shielding material, boron carbide, thermo-physical property, laser-flash method, thermal cycling

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1. Introduction

The thermal and radiation shielding is one of the important components of breeding blankets of the fusion power plant. In the design of the helical typed fusion reactor (Force Free Helical Reactor; FFHR), a combination of reduced activation ferrite steel (RAFM) and boron carbide (B_4C) is chosen as main radiation shielding material [1]. In the neutron engineering design of FFHR, tungsten carbide (WC), which is higher sintering density and radiation shielding compared with B_4C, will also be installed behind the blanket (between blanket and superconducting magnet) [1]. In addition to the carbide materials, hydride materials such as ZrH_2 and TiH_2 are also studied for the shielding application [2].

In the FFHR design, the temperature of the shield is kept between 100 °C and 200 °C under the steady state operation. However, it is also requested that the shield shows high safety performance in the case of temperature rise induced by the loss of He coolant [1]. The both thermo-physical and neutronic properties are most important physical parameters for the shielding material for the fusion blanket. However, the carbide materials such as B_4C and WC are categorized into advanced functional materials and the data-base of the both thermo-physical and neutronic properties necessary for evaluating their performance during the use in fusion blanket is quite limited.

Generally, there are two types of thermal conductivity measurement methods, namely steady-state and transient methods. The temperature gradient method, which is one of the typical steady-state methods, is a simple and easy process to evaluate the thermal conductivity directly [3–5]. This method, however, has various disadvantages, such as long time and large sized sample necessary for the precise evaluation and narrow applicable temperature range typically being from room temperature to 150 °C due to the resolution limit of the sensor. Recently, thermal transient methods are mainly used for evaluation of thermal conductivity in wide range of temperature. The principle of the flash method, which is one of the transient methods, is already established by Parker *et al.* [6]. In the flash method, there are several types of pulse light source such as Xenon flash and laser. The laser flash method has the wide applicable temperature range and can measure the thermal conductivity (λ) precisely. The evaluation is based on the following simple equation,

$$\lambda = \alpha \times Cp \times \rho, \tag{1}$$

where α is the thermal diffusivity, Cp is the specific heat capacity and ρ is the sample density. This equation means that measurement of thermal diffusivity makes it possible to evaluate the thermal conductivity.

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In this study, we evaluated thermal diffusivity of the B_4C bulk samples using the laser flash method. The effect of heat cycling on the microstructure and thermal diffusivity was also investigated in order to evaluate the thermal soundness during operation in fusion blankets.

2. Sample Preparation and Measurements

We prepared high purity (> 99.9%) B_4C bulk samples (Ceradyne inc., USA) using natural boron and carbon powders. The B_4C bulk samples were fabricated by the hot isostatic press (HIP) sintering. The dimension of B_4C bulk samples is 10 mm in diameter and 3 mm thickness. The sintering density of the B_4C sintered bulk exceeded above 95% of the theoretical density.

The prepared B_4C sintering bulk samples were carried out the thermal cycling up to 700 °C. This temperature was selected as the typical highest operation temperature of the breeding blanket systems [7]. The B_4C bulk samples were set into the temperature plateau region of the electrical furnace and were heated up to 700 °C of the peak temperature in flowing 6N high purity Ar gas. The temperature was elevated at a rate of 700 °C/hour and then held for 1 hour, followed by furnace cooling to 50 °C. The number of thermal cycling was 100.

Microstructure of B_4C bulk before and after the thermal cycling was investigated using the X-ray diffraction (XRD) analysis and scanning electron microscope (SEM) observation. The thermal diffusivity of B_4C bulk samples was measured using the laser-flash method according to the Japan Industry Standard (JIS) code (R1611-1997). The schematic configuration of the equipment is shown in Fig. 1. At first, B_4C samples were coated with Au thin film by sputtering followed by graphite spraying in order to absorb the flashing laser beam efficiently and to be opaque for the wave-length of the heating laser beam and the corresponding wave-length of the infrared radiation thermometer. The thermal diffusivity (α) is estimated by the following equation so called "Half Time method" [6];

$$\alpha = 0.1388 \times d^2/T_{1/2}, \tag{2}$$

where d is sample thickness and $T_{1/2}$ is the time required for the back surface to reach half of the maximum temperature rise. The measurements were carried out between room temperature to around 800 °C.

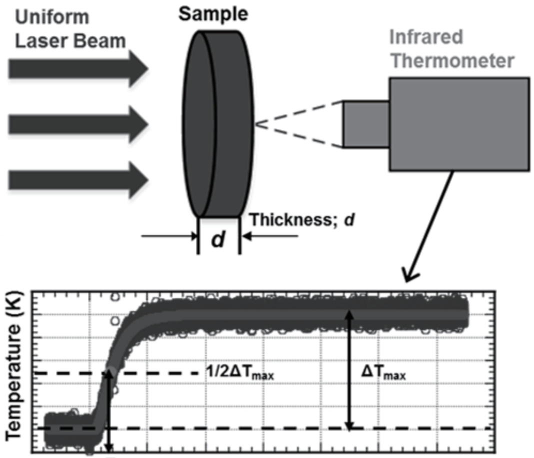


Fig. 1 Schematic configuration and transient temperature curve of the laser-flash method.

3. Results and Discussions

3.1 Microstructure of B_4C bulk samples before and after thermal cycling

Figure 2 shows the bulk density (D_n) degradation of B_4C bulk as a function of the thermal cycling times. D_0 is the sintered bulk density before thermal cycling. The sintered B_4C bulk density was decreased to 98% by the thermal cycling up to 700 °C. This is considered to be caused by the thermal expansions and constrictions of the B_4C bulk.

Typical XRD pattern of the B_4C sintered bulk before and after the thermal cycling is shown in Fig. 3. All main diffraction peaks of the bulk samples before and after thermal cycling indicated the B_4C phase. We confirmed that B_4C sintered bulk did not cause the phase transformation by the 100 thermal cycling up to 700 °C.

The comparisons of Full Width at Half Maximum (FWHM) on the X-ray diffraction of B_4C bulk sample before and after thermal cycling is shown in Fig. 4. The peak intensity count was decreased by the thermal cycling. It is known that FWHM is the measure of crystallinity. The decrease of FWHM means the enhanced crystallinity. In Fig. 3, the FWHM was increased from 0.2 to 0.28 by the thermal cycling. This suggested that the B_4C crystallinity was lowered by the thermal cycling up to 700 °C.

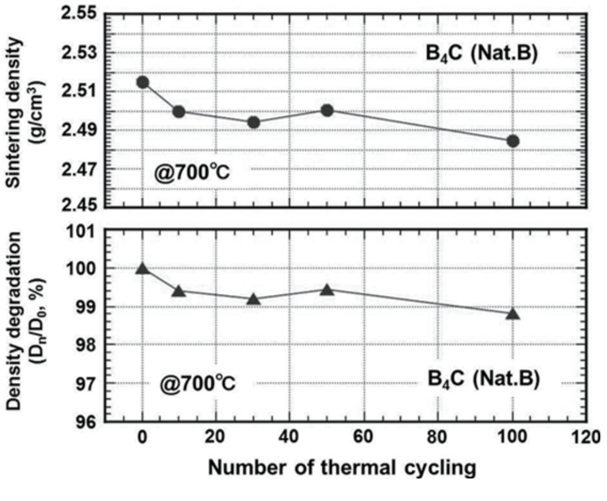


Fig. 2 The sintering density and density degradation of the B_4C bulk sample as a function of the number of the thermal cycling up to 700 °C.

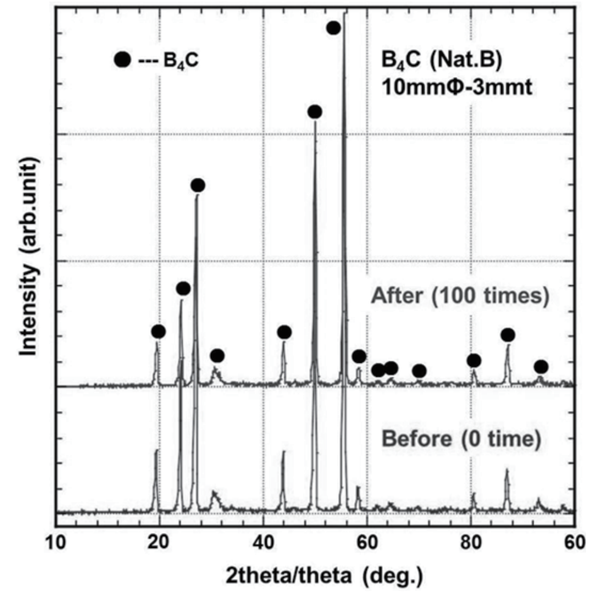


Fig. 3 Typical XRD patterns of the surface area on the B_4C bulk sample before and after thermal cycling.

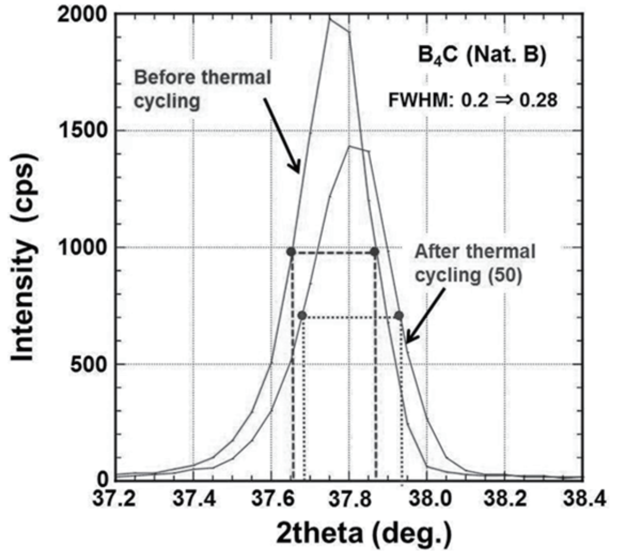


Fig. 4 The comparisons of FWHM on the X-ray diffraction peak of B_4C bulk sample before and after thermal cycling.

Figure 5 shows that typical SEM image of the surface area in a B_4C bulk sample before and after thermal cycling up to 700 °C. The B_4C bulk sample before and after thermal cycling showed high densely structure without voids and cracks. This suggested that the B_4C sintered bulk had highly stable state against the thermal stress.

Here, in the case of the FFHR design in NIFS, the life-time design of blanket system is 30 years [8–10]. If the maintenance of blanket system is carried out once a year, major thermal cycling of more than 30 times but with much lower temperature range than the present tests is expected during the life-time of blanket system. We thought that B_4C bulk was sufficient thermal durability throughout the blanket life time.

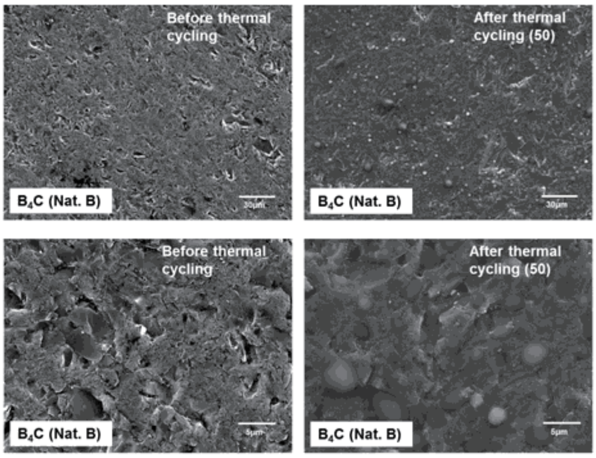


Fig. 5 Typical SEM image of surface area in B_4C bulk sample before and after thermal cycling.

The plate-like structured B_4C grains were obtained before thermal cycling (as-HIP sintering). However, a part of the plate-like structured B_4C grains changed to the spheroidized structure after the thermal cycling. The lowering of B_4C crystallinity shown in Fig. 3 thought to be caused by the microstructure change.

From the results of XRD analysis and SEM observation shown in Figs. 3, 4 and 5, we concluded that B_4C bulk has high thermal stability and soundness of microstructure during the life-time of blanket system. However, it is necessary to investigate the thermal and fast neutron irradiation effects of the B_4C bulk for the shielding blanket application.

3.2 The thermal diffusivity of the B_4C bulk samples measured by the laser-flash method

Figure 6 shows the temperature dependence of thermal diffusivity before thermal cycling of the B_4C bulk sample measured by the laser-flash method. The thermal diffusivity of the B_4C bulk sample around room temperature was about $1.5 \sim 1.6 \times 10^{-5}\,\mathrm{m^2S^{-1}}$. The thermal diffusivity of B_4C bulk was lowered with increasing temperature, which is typical behavior of temperature dependence of ceramic materials. In the case of ceramic material, heat is mainly carried by phonon and the thermal diffusivity is proportional to the heat capacity, the phonon velocity, and the mean free path of phonons. The temperature dependence shown in Fig. 6 was caused by the shorter mean free path with phonon-phonon scattering under high temperature. The thermal diffusivity for the temperature range from 100 °C to 300 °C is highly relevant to the shielding blanket design. The thermal diffusivity for this temperature range region was obtained to be about $5 \sim 7 \times 10^{-6}\,\mathrm{m^2S^{-1}}$ and was about 1/3 of that at the room temperature.

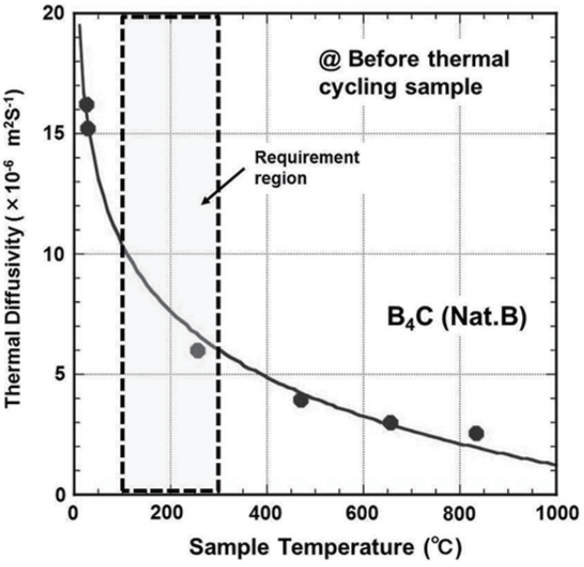


Fig. 6 The thermal diffusivity before thermal cycling of the B_4C bulk sample as a function of the sample temperature.

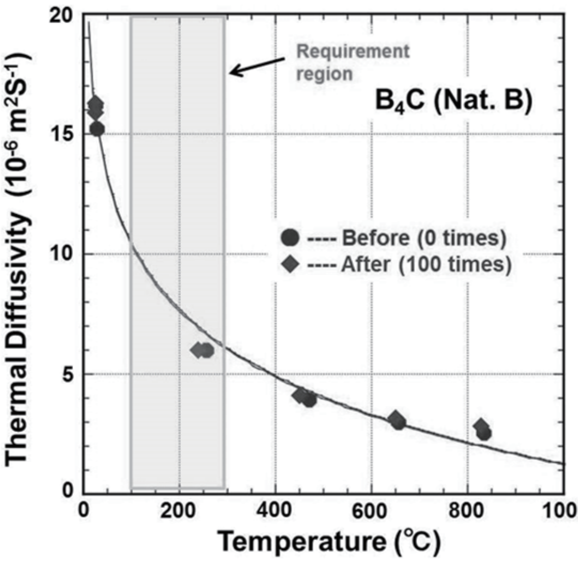
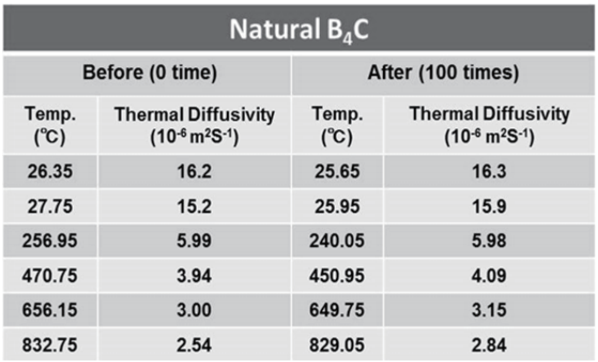


Fig. 7 Temperature dependence of the thermal diffusivity in the B_4C bulk before and after thermal cycling.

Table 1 The thermal diffusivity of the B_4C bulk sample before and after the thermal cycling.



Natural B_4C			
Before (0 time)		After (100 times)	
Temp. (℃)	Thermal Diffusivity (10^{-6} m^2S^{-1})	Temp. (℃)	Thermal Diffusivity (10^{-6} m^2S^{-1})
26.35	16.2	25.65	16.3
27.75	15.2	25.95	15.9
256.95	5.99	240.05	5.98
470.75	3.94	450.95	4.09
656.15	3.00	649.75	3.15
832.75	2.54	829.05	2.84

The effect of thermal cycling on the thermal diffusivity was also investigated. Table 1 and Fig. 7 show the thermal diffusivity of the B_4C bulk samples before and after thermal cycling up to 700 °C. The number of thermal cycling is 100. We found that the thermal cycling did not induce thermal diffusivity degradation and that B_4C bulk had high thermal soundness in the present FFHR design.

4. Conclusions

In this study, the thermal cycling effect on the thermal property and microstructure on the B_4C bulk, which is one of the candidates of thermal and radiation shielding materials of fusion blanket, was investigated. The thermal diffusivity in the expected operation temperature range (100 °C - 300 °C) was obtained to be about $5 \sim 7 \times 10^{-6}$ m^2S^{-1} which is about 1/3 compared with that at the room temperature. Although the density decreased slightly, the thermal diffusivity was not degraded by the thermal cycling. Thus the B_4C bulk is expected to have high thermal stability and soundness of microstructure during the lifetime of blanket system.

It is necessary to investigate the effects of thermal and fast neutron irradiation on the B_4C bulk as the shielding material for fusion blanket.

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