Journal of Nuclear Materials 386–388 (2009) 628–630

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Compatibility between SiC and Li ceramics for solid breeding blanket system

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ABSTRACT

Compatibility of high crystalline β-SiC in contact with Li ceramics was studied. Isothermal annealing of the stack of monolithic β-SiC and Li ceramics (Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3 and $LiAlO_2$) in helium was carried out at the temperature of 900 and 1000 °C for 100 h. No bonding between β-SiC and Li ceramics occurred for all the experiment conditions. Adhering substance composed by Li_2SiO_3 on the β-SiC surface and diffusion of ^{7}Li and ^{16}O to bulk β-SiC was clearly observed for the β-SiC specimen in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3. While, small amount of the adhering substance and small diffusion those elements were observed for the β-SiC specimen in contact with $LiAlO_2$.

1. Introduction

Helium cooled solid breeder (HCSB) blanket using silicon carbide (SiC) fiber reinforced SiC matrix composite (SiC/SiC composite) as a structural material is one of the promising blanket design concept because of its high temperature operation and almost no-need to consider the effect of electromagnetic force and corrosion [1]. There remain compatibility issues to be solved in the HCSB blanket, for example, a compatibility of SiC/SiC composite in contact with Li ceramics as a solid breeder and Be-based material as a neutron multiplier.

This study focuses on the compatibility of SiC/SiC composite in contact with Li ceramics. The open literatures [2–4] reported a limited study for the compatibility issue between SiC/SiC composite (SiC) and Li ceramics. The compatibility of SiC/SiC composite with Li ceramics (lithium orthosilicate (Li_4SiO_4), lithium metazirconate (Li_2ZrO_3) and lithium aluminate ($LiAlO_2$)) at 600 and 800 °C [2], the compatibility of SiC/SiC composite with a lithium metatitanate (Li_2TiO_3) and a Li_4SiO_4 at 800 °C [3] and the compatibility of α-SiC and Li ceramics (Li_4SiO_4, Li_2ZrO_3 and Li_2TiO_3) at 700 °C [4] were reported. However, the HCSB blanket is expected to be operated above 800 °C for higher efficiency, therefore, the compatibility at higher temperature above 800 °C should be studied.

The purpose of this study is to clarify the compatibility of monolithic β-SiC in contact with Li ceramics at the temperature above 800 °C. High crystalline β-SiC and four kinds of Li ceramics (Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3 and $LiAlO_2$) were examined in this work.

2. Experimental

Sintered Li ceramics pellet (Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3, $LiAlO_2$, shape: 10 $mm^{\phi} \times 1$ mm^{t}) was used, which was fabricated by TYK Corp. The sintering temperature and time was 1300 °C for 5 h for Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 and 1000 °C for 5 h for $LiAlO_2$. The measured density was 2.69 g/cm^3 for Li_2TiO_3, 1.69 g/cm^3 for Li_4SiO_4, 2.43 g/cm^3 for Li_2ZrO_3 and 1.25 g/cm^3 for $LiAlO_2$, respectively. Though these Li ceramics might contain some sintering agent, their details have not opened by the fabricator. Monolithic β-SiC was fabricated using a chemical vapor deposition (CVD) process by Rohm and Haas. The CVD temperature was 1000 °C. These materials were machined into a disc shape specimen with diameter of 3 mm. Contact surface of these specimens were polished into flat and mirror state.

Specimens were stacked in an inconel tube capsule. Fig. 1 shows the schematic illustration of the capsule including stack of β-SiC and Li ceramics. This capsule was sorted in helium filled quartz tube. The isothermal annealing for the helium filled quartz tube was carried out at the temperature of 900 and 1000 °C for 100 h. The helium pressure during the annealing was 1 atm.

Observation of the contact surface after the compatibility test was performed using an optical microscope and a scanning electron microscope (SEM, JSM-5310LV, JEOL Ltd.). Compositional analysis of contact surface was examined using an X-ray diffraction method (XRD, JDX-3530, JEOL Ltd.) and a secondary ion mass spectroscopy (SIMS, Model 6600, ULVAC-PHI Inc.).

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doi:10.1016/j.jnucmat.2008.12.225

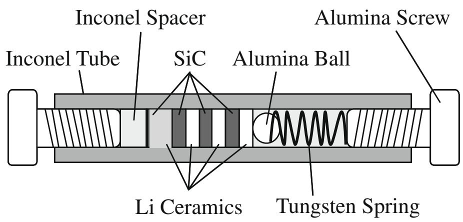

Fig. 1. Schematic illustration of the inconel tube capsule including stack of β-SiC and Li ceramics.

3. Results and discussion

Fig. 2 shows the contact surface of β-SiC and Li ceramics after annealing for 100 h. Adhering substance was observed on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3, while small amount of the adhering substance was observed on the β-SiC surface in contact with $LiAlO_2$. The adhering substance should be reaction layer occurring during the isothermal annealing. Thus, bonding between β-SiC and Li ceramics must have occurred at some level, especially for the β-SiC/Li_2TiO_3, β-SiC/Li_4SiO_4 and β-SiC/Li_2ZrO_3 diffusion couples, though it wasn't mechanically strong. Table 1 summarizes the XRD analysis for the contact surface of β-SiC after annealing for 100 h. Formation of Li_2SiO_3 was clearly observed for the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3, while nothing except for SiC was detected for the β-SiC surface in contact with $LiAlO_2$.

The direct reaction between Li ceramics and SiC (e.g. Li_2TiO_3 + SiC → Li_2SiO_3 + TiC, Li_2ZrO_3 + SiC → Li_2SiO_3 + ZrC) could create Li_2SiO_3. However, these reactions could occur at the temperature above 1000 °C [4], therefore, Li_2SiO_3 on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 might not formed by the direct reaction.

Table 1
Summary of the XRD analysis for the contact surface of β-SiC after annealing for 100 h.

Temperature	SiC/Li_2TiO_3	SiC/Li_4SiO_4	SiC/Li_2ZrO_3	SiC/$LiAlO_2$
900 °C	SiC Li_2SiO_3	SiC Li_2SiO_3	SiC Li_2SiO_3	SiC
1000 °C	SiC Li_2SiO_3	SiC Li_2SiO_3	SiC Li_2SiO_3	SiC

The reaction between Li ceramics and SiO_2 as follows could occur and create Li_2SiO_3 at the temperature from 600 to 1000 °C [2–4]:

$$Li_2TiO_3 + SiO_2 \rightarrow Li_2SiO_3 + TiO_2 \quad (1)$$

$$Li_4SiO_4 + SiO_2 \rightarrow 2Li_2SiO_3 \quad (2)$$

$$Li_2ZrO_3 + SiO_2 \rightarrow Li_2SiO_3 + ZrO_2 \quad (3)$$

The formation of SiO_2 was observed for the same β-SiC as this study after oxidation test in the in He environment at 1000 °C [5]. Therefore, it is possible that Li_2SiO_3 on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 might be formed by reaction (1)–(3). Additionally, the formation of SiO_2 in Li_4SiO_4 due to Li, O_2 and Li_2O evaporation from solid Li_4SiO_4 was considered [4]. This reaction could enhance the reaction (2).

Sample [2] reported that Li_2SiO_3 formation on the surface of SiC/SiC composite in contact with Li_4SiO_4, Li_2ZrO_3 and $LiAlO_2$ at 800 °C was observed and that Li_4SiO_4 was found to be much more reactive than Li_2ZrO_3 and $LiAlO_2$. Thus, it is possible that small amount of

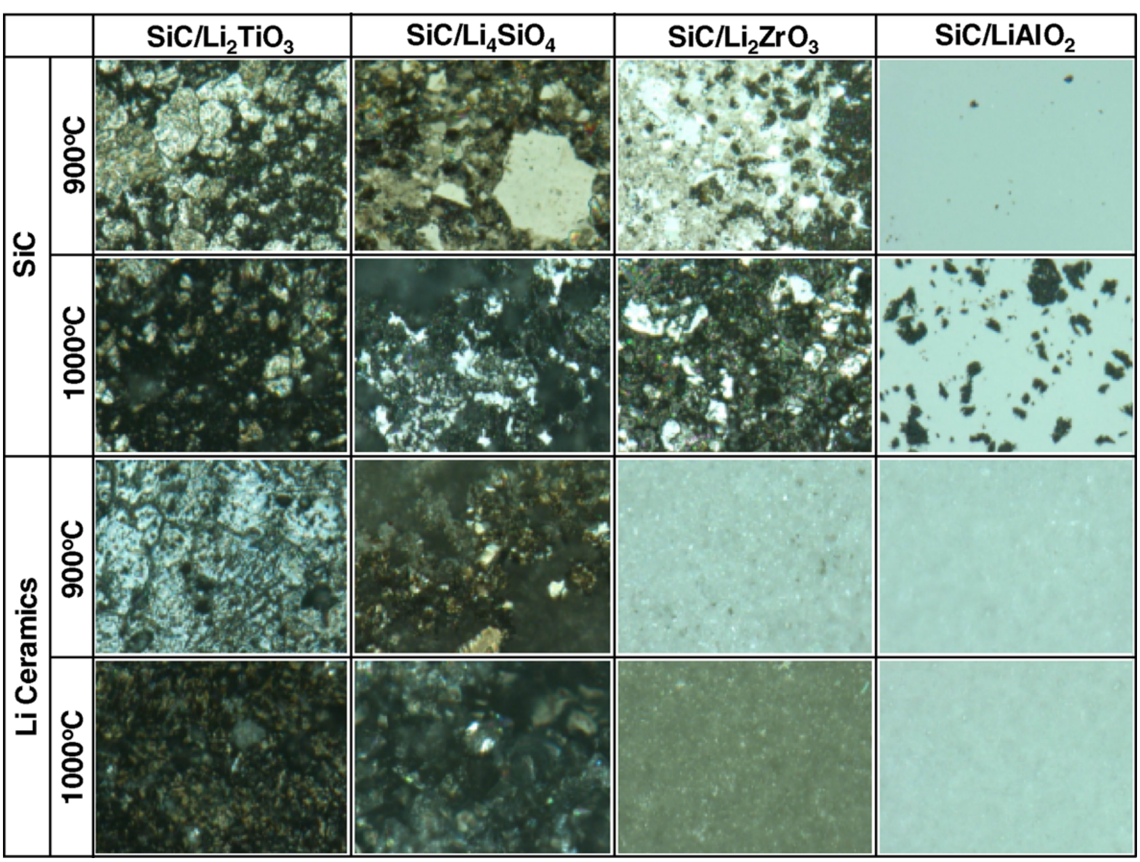

Fig. 2. The contact surface of β-SiC and Li ceramics after annealing for 100 h.

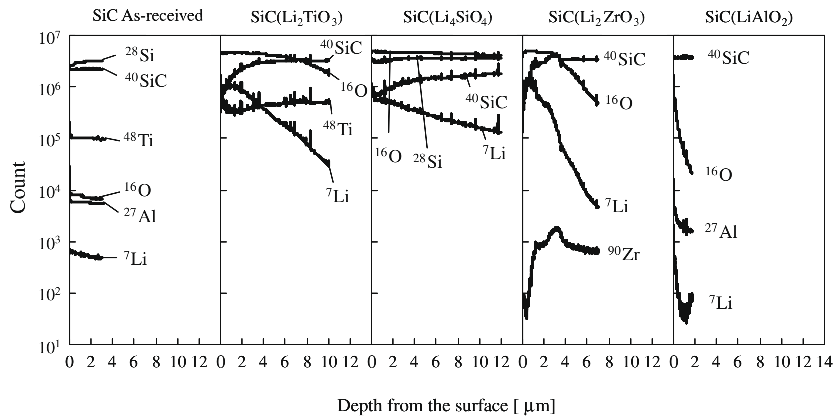

Fig. 3. The SIMS spectrum of the as-received β-SiC and the contact surface of β-SiC after annealing for 100 h.

Li_2SiO_3 was formed on the β-SiC surface in contact with $LiAlO_2$ by the following reaction (4). No detection of Li_2SiO_3 by the XRD analysis might be observed because of the small amount of Li_2SiO_3.

$$2LiAlO_2 + SiO_2 \rightarrow Li_2SiO_3 + Al_2O_3 \quad (4)$$

Fig. 3 shows the SIMS spectrum of the as-received β-SiC and the contact surface of β-SiC after annealing for 100 h. The spectrum of the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 includes not only the measurement of bulk β-SiC but also that of the adhering substance (Li_2SiO_3) layer, while the spectrum of the β-SiC surface in contact with $LiAlO_2$ includes only measurement of bulk β-SiC. Impurity of ^{7}Li, ^{16}O, ^{27}Al, ^{48}Ti and ^{90}Zr in the β-SiC used in this study was below a few ppb [6], therefore, the signal of ^{7}Li, ^{16}O, ^{27}Al, ^{48}Ti and ^{90}Zr of the as-received β-SiC specimen was considered to be the background level of these elements in SIMS measurement of this study. The thickness of the adhering substance layer on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 was considered to be about 4 μm based on the ^{40}SiC signal. Diffusion of ^{7}Li and ^{16}O to the bulk β-SiC was clearly observed for the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3, while small diffusion of ^{7}Li and ^{16}O was observed for the β-SiC surface in contact with $LiAlO_2$. The diffusion depth of ^{7}Li and ^{16}O into β-SiC was above 15 μm for the β-SiC/Li_2TiO_3, β-SiC/Li_4SiO_4 and β-SiC/Li_2ZrO_3 diffusion couples and about 2–5 μm for the β-SiC/$LiAlO_2$ diffusion couple. Therefore, it is possible that the adhering substance layer on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3 might be formed by a solid state diffusion bonding mechanism between β-SiC and Li ceramics.

4. Summary

In order to clarify the compatibility of SiC as a structural material in contact with Li ceramics as a solid breeder in the helium cooled solid breeder (HCSB) blanket, the isothermal annealing of the stack of monolithic β-SiC and Li ceramics (Li_2TiO_3, Li_4SiO_4, Li_2ZrO_3 and $LiAlO_2$) in helium was carried out at the temperature of 900 and 1000 °C for 100 h. The following results were obtained:

(1) No bonding between β-SiC and Li ceramics occurred for all the experiment conditions.
(2) Adhering substance composed by Li_2SiO_3 with a thickness of about 4 μm was observed on the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3, while small amount of the adhering substance was observed on the β-SiC surface in contact with $LiAlO_2$.
(3) Diffusion of ^{7}Li and ^{16}O to bulk β-SiC was clearly observed for the β-SiC surface in contact with Li_2TiO_3, Li_4SiO_4 and Li_2ZrO_3, while small diffusion of ^{7}Li and ^{16}O was observed for the β-SiC surface in contact with $LiAlO_2$.

Acknowledgement

This work was partly supported by the JUPITER-II (Japan-USA Program of Irradiation Testing for Fusion Research II) program.

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