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Tritium trapping in silicon carbide in contact with solid breeder under high flux isotope reactor irradiation

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ABSTRACT

The trapping of tritium in silicon carbide (SiC) injected from ceramic breeding materials was examined via tritium measurements using imaging plate (IP) techniques. Monolithic SiC in contact with ternary lithium oxide (lithium titanate and lithium aluminate) as a ceramic breeder was irradiated in the High Flux Isotope Reactor (HFIR) in Oak Ridge, Tennessee, USA. The distribution of photo-stimulated luminescence (PSL) of tritium in SiC was successfully obtained, which separated the contribution of ^{14}C β-rays to the PSL. The tritium incident from ceramic breeders was retained in the vicinity of the SiC surface even after irradiation at 1073 K over the duration of ~3000 h, while trapping of tritium was not observed in the bulk region. The PSL intensity near the SiC surface in contact with lithium titanate was higher than that obtained with lithium aluminate. The amount of the incident tritium and/or the formation of a Li_2SiO_3 phase on SiC due to the reaction with lithium aluminate under irradiation likely were responsible for this observation.

1. Introduction

Silicon carbide (SiC) is a promising reduced activation structural material for deuterium–tritium fusion blankets because of its high-temperature operation capability and chemical inertness, especially in the form of SiC fiber-reinforced SiC–matrix composites [1–3]. To establish an effective tritium recovery scheme, it is important to examine the behavior of tritium in SiC under irradiation with neutrons and energetic particles, including tritons created by nuclear reactions of $^6Li(n,\alpha)T$ in breeder materials. The behavior of hydrogen isotopes, including tritium in SiC and its composites, has been discussed in the literature [4–8]. Data from these studies shows six orders of magnitude deviation of the diffusion coefficient. In these studies, the hydrogen isotopes retained in the bulk region of SiC have not been directly observed because the gaseous tritium, which is thermally released in the form of HTO, T_2O, HT and/or T_2, is detected in most cases. Because hydrogen isotopes in materials undergo several processes (e.g., trapping–detrapping, diffusion, and recombination) before their release, the direct observation of tritium in the materials can extensively elucidate the underlying tritium behavior, including the effects of irradiation with energetic particles. The goal of this study is to evaluate tritium distribution in SiC under irradiation at high temperature using the imaging plate (IP) technique, which is a powerful tool for measuring the tritium distribution on and in the bulk region of materials [9].

Studies on tritium injected as energetic particles into SiC under neutron irradiation at high temperatures assuming fusion reactor conditions (e.g., the proposed design of the helium-cooled ceramic breeder blanket) are not available to date. Recently, monolithic beta-phase SiC and lithium ceramics were irradiated in the High Flux Isotope Reactor (HFIR) to elucidate the compatibility, irradiation effects, and behavior of tritium created in lithium ceramics under neutron irradiation and injected into SiC as MeV energetic particles. In 2011, we reported that SiC exhibited better compatibility with lithium aluminate (LAO) and lithium titanate (LTO) than with lithium zirconate and silicate [10]. In the present study, trapping of tritium in situ injected into SiC samples from LAO and LTO during irradiation in the HFIR was examined using the

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IP technique. To the best of our knowledge, this is the first study to successfully evaluate tritium in radioactive ceramic materials irradiated with neutrons and tritium.

2. Experimental

This study used high purity monolithic SiC disk samples (3 mm diameter and ~1 mm in thick), fabricated by Rohm and Hass Co. Ltd. by chemical vapor deposition. The SiC samples were loaded in a helium-filled titanium–zirconium–molybdenum (TZM) capsule in contact with disks of lithium aluminate ($Li_{1-x}AlO_{2-x}$:LAO) and lithium titanate ($Li_{2-x}TiO_{3-y}$:LTO) on one side. Irradiation was performed in the HFIR which is located at Oak Ridge National Laboratory. The irradiation temperature was 1073 K and the fast neutron ($E > 0.1$ MeV) fluence was 5.9×10^{25} n/m^2, corresponding to 5.9 displacements by the fast neutrons per atom (dpa) for SiC [11].

After irradiation, the SiC disk samples were ultrasonically cleaned. They were then heated to 423 K for 0.5 h to remove contaminants on the surface, prior to being cut in the cross-sectional direction and polishing. The cross-sectional face of the SiC sample was exposed to an IP film (GE Healthcare Japan, BAS-TR2040), as depicted in Fig. 1. The exposure time was 1 h. The photo-stimulated luminescence (PSL) intensity distribution was measured by an IP reader (Fujifilm, FLA-7000). In the exposure process, a thin polymer film (1.2 μm thick, ING Corporation) was placed between the SiC sample and IP film to avoid contamination tritium or other radioactive constituent on the IP film. Because the range of tritium β-rays in the polymer film was approximately 5 μm, the contribution of the thin polymer film to the PSL intensity distribution could be negligible. To separate the contribution of tritium β-rays to PSL signals from that of ^{14}C β-ray excitation, IP measurements were also performed on a reference SiC sample irradiated in the HFIR outside the TZM capsule at the same fluence and temperature to prevent irradiation with tritium created in ceramic breeder materials. The IP technique is useful for detecting β-rays from tritium because of its wide dynamic range, which has been demonstrated in various metal materials by Otsuka et al. [9]. However, the application of this technique to the measurement of tritium in radioactive materials has been limited because radiation from radionuclides can affect the memorizing system of IP film. Even at a high fluence of neutron irradiation, the low activity of SiC enabled the evaluation of tritium distribution owing to the lower background radiation. Structural changes on the SiC face in contact with the breeding materials were analyzed by X-ray diffraction (θ–2θ XRD), using an X'Pert Pro diffractometer (PANalytical).

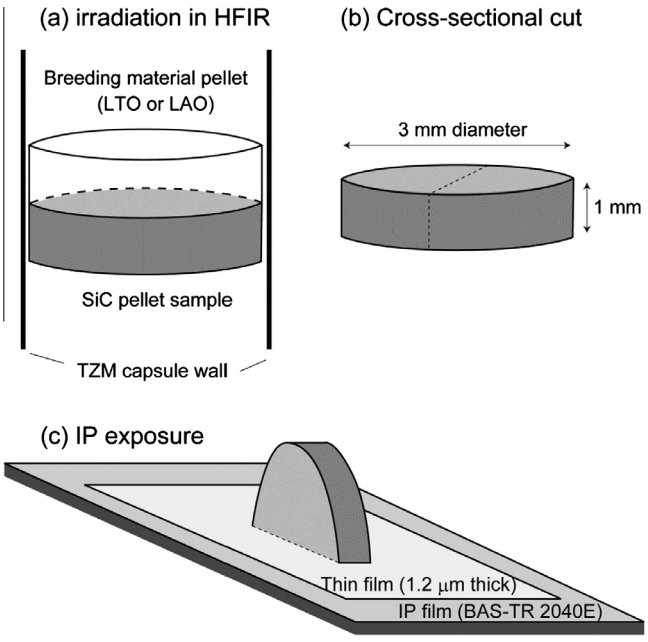

Fig. 1. Schematic of HFIR irradiation and tritium experimental procedures using IP.

3. Results and discussion

IP images of cross sections of SiC irradiated in the HFIR with ceramic breeder materials are shown in Fig. 2. To clarify the effect of tritium injection from solid breeder materials, an image showing the reference SiC sample outside the TZM capsule is included in Fig. 2. The upper areas of samples in images of (a) and (b) correspond to the face of SiC in contact with the solid breeder disk. A

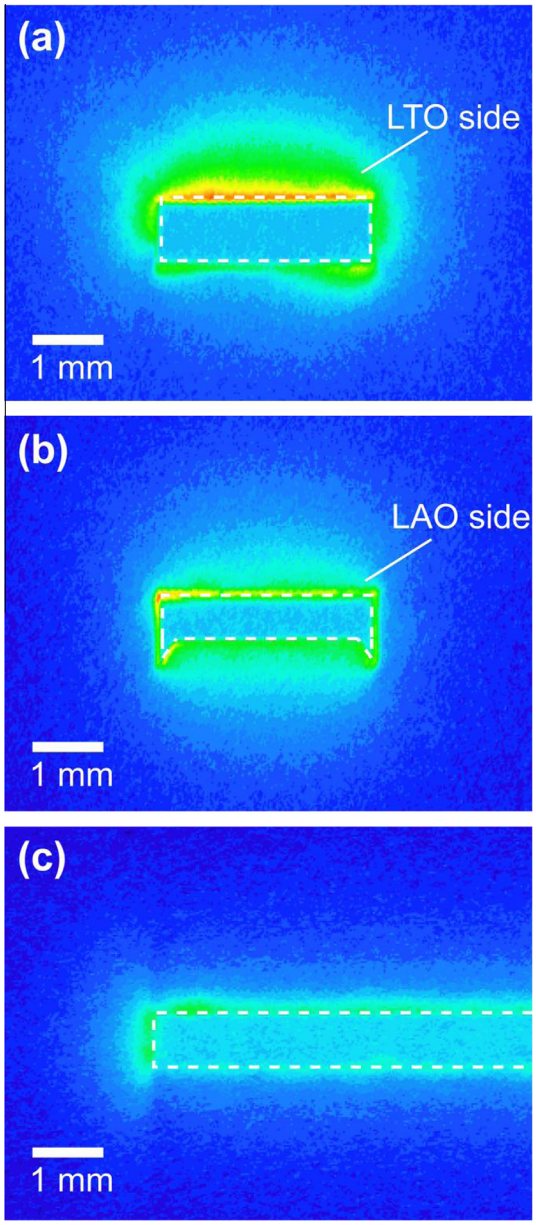

Fig. 2. IP images of cross-section in SiC samples irradiated in HFIR at 1073 K to 5.9 dpa fluence. The edges on the upper side in the images are surfaces that had been in contact with LTO (a) and LAO (b) under irradiation. For comparison, an IP image of a reference SiC is shown in (c).