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Investigation of the residual stress in reaction-bonded SiC under irradiation

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ABSTRACT

The development of reaction-bonded SiC (RB-SiC) for an application in nuclear reactor environments has been suspended for many years because of the possible swelling mismatch between SiC and Si under irradiation. Therefore, we studied the residual stresses in RB-SiC due to the irradiation-induced swelling mismatch and the mismatch between the coefficients of thermal expansion. The maximum irradiation-induced swelling stress for the amorphous state was obtained by irradiation with 2 MeV Au^{2+} ions (1×10^{16} ions/cm^2) at room temperature. Utilizing Raman piezo-spectroscopy, nanoindentation tests and thermal expansion coefficient measurements, we demonstrate that the irradiation-induced swelling stress is remarkably high at low temperatures, whereas the thermal stress becomes the dominant residual stress at medium (415 °C < T < 700 °C) or higher temperatures. By optimizing the residual Si content in the RB-SiC, the thermal stress and the irradiation-induced swelling stress can be adjusted and the initiation of cracks can be prevented.

1. Introduction

The reaction bonding process, also called liquid silicon infiltration (LSI) or melt infiltration (MI), is an effective process to produce high-density near net shape SiC components via a reaction of C with molten Si at temperatures above 1450 °C in vacuum [1–3]. Reaction-bonded (RB)-SiC exhibits a low porosity, an excellent hermeticity and a high thermal conductivity, which is very competitive when compared with SiC components intended for nuclear applications that are produced through other techniques, such as chemical vapor infiltration (CVI) and polymer impregnation and pyrolysis (PIP) [4–7]. Recently, the high-temperature performance of RB-SiC has been extensively investigated by controlling the preforms, porosity, and the free Si and free C content. However, the softening of the excess free Si (with a melting point of 1410 °C) at operating temperatures above ~1400 °C, which decreases the strength and toughness of RB-SiC components is a major drawback [8,9]. There is little report on the irradiation damage and its effect on the properties of RB-SiC, and the corresponding failure mechanism is still unclear. Some researchers argued that the excess Si in RB-SiC segregates to the grain boundaries, indicating a radiation instability of the non-stoichiometric structure [10,11]. Other researchers believed that the anisotropic lattice expansion between Si and SiC is the major cause for the decline of the fracture strength by nearly 50% after irradiation [12–14]. These comments seem ambiguous. However, the segregation, phase transformation, grain growth, thermal expansion, and irradiation-induced swelling all contribute to volume changes in the SiC and Si phases, which can be monitored by measuring a single parameter, i.e., σ, the residual stress [15,16].

An exacerbating and uneven distribution of the residual stresses is known to have a significant impact on the mechanical properties, the structural integrity and the lifetime of reactor components [17]. Considering the challenging requirements for an application in advanced fission and future fusion reactors, the design criteria of SiC matrix components includes an upper operating temperature between 600 and 1200 °C, a maximum damage level of 150–200 displacements per atom (dpa) and a lifetime exceeding 30 years (15 MW a m^{-2}) [18,19]. In this case, the residual stress not only includes the familiar thermal stress (σ_T) during the heating-cooling cycle but also a contribution due to the irradiation because the degree of swelling in the different phases may not be the same (σ_S). Unfortunately, the latter residual stress due to the irradiation damage has never been quantitatively calculated.

The aim of this paper is to improve the properties of RB-SiC components for a potential application in nuclear reactors through an optimization and management of the residual stress states

$$\sigma = \sigma_T + \sigma_S + \sigma_{ex}, \quad (1)$$

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Table 1
Comparison of the bulk density and open porosity values determined for the SiC/C preform and the as-received reaction-bonded SiC.

Samples	Bulk density (g/cm^3)	Open porosity (vol.%)
SiC/C preform	1.80	31.4
Reaction-bonded SiC	3.01	0.4

where σ_{ex} is a potential excess stress, which depends on the specific environmental conditions and arises, for instance, from phase transformation, or an external force. σ_T was assessed by studying the variation of the coefficient of thermal expansion (CTE) and the Young's modulus of the RB-SiC. The maximum value of σ_s was evaluated by Raman piezo-spectroscopy under irradiation with 2 MeV Au^{2+} ions to a dose of up to 1×10^{16} ions/cm^2 at room temperature (RT), resulting in a high displacement damage. The experiments were conducted at RT because both SiC [6,20] and Si [21–23] were reported to exhibit the largest swelling near RT (<150 °C) in an amorphous state. The effect of the ion irradiation was characterized by transmission electron microscopy (TEM), scanning electron microscopy (SEM), and nanoindentation tests. Finally, the contribution of the differential residual stresses σ_T and σ_s are discussed for a wide operating temperature range from RT to 1200 °C.

2. Material and methods

2.1. Sample preparation

The preforms consisting of β-SiC and C were prepared by mixing β-SiC powders with two different particle sizes (0.5 and 10 μm with a mass ratio of 1:3, 99.9% purity, Beijing HWRK Chemical Co., China) with phenol formaldehyde resin (Xi'an resin factory, China) by ball milling in a sealed jam. The β-SiC to resin mass ratio was selected to be 2:1. The mixture was then cold-pressed by applying a pressure of 100 MPa for 1 min. After that, the curing and carbonization were carried out in a tube furnace under argon atmosphere by increasing the temperature from RT to 900 °C, using a heating rate of 2 °C/min. Finally, the porous SiC/C preforms were infiltrated with liquid Si at 1600 °C for 30 min in vacuum. The bulk density and open porosity of both the preform and the final product were measured by adopting the Archimedes method, and the results are shown in Table 1. The C/Si atom ratio of the final product was determined to be 1:1 using an IR carbon-sulfur analyzer. The volume fraction of residual Si in the final RB-SiC samples was determined by acid etching at 40 °C for 24 h using a mixture of 70 wt.% HF and 30 wt.% HNO_3 according to the following reactions:

$$3\ Si + 4HNO_3 \rightarrow 3\ SiO_2 + 4NO \uparrow + 2H_2O \quad (2)$$

$$SiO_2 + 4HF \rightarrow SiF_4 + 2H_2O \quad (3)$$

and was calculated to be 17.1 vol.%.

The surface of the samples was polished to a surface roughness of 1 μm prior to the irradiation and subsequent sample characterization using a diamond lapping film.

2.2. Ion irradiation and sample characterization

The ion irradiation was performed at normal incidence using the 2×1.7 MV tandem accelerator at the State Key Laboratory of Nuclear Physics and Technology, Peking University. Since our goal was to study the maximum σ_s in the amorphous state generated by the excess agglomeration of radiation-induced point-defects, a local damage dose much higher than 0.25 dpa (i.e., the critical dose D_c for the full amorphization of pure SiC at RT [24]) and a thick amorphous layer were preferred. According to the full damage cascade simulations performed with the stopping and range of ions in matter (SRIM) computer program [25] under the assumptions of threshold displacement energies of 35 and 20 eV for the Si and C sublattices, respectively [26] (Fig. 1), for an Au ion energy of 2 MeV and a dose of 1×10^{16} ions/cm^2 at RT, the resulting fully-amorphous layer extends from the surface to a depth of approx. 620 nm.

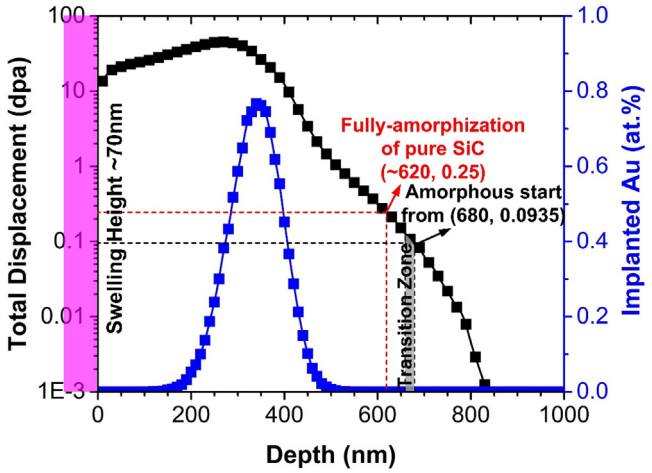

Fig. 1. Depth profile of the displacement damage as predicted by the SRIM simulations and Au^{2+} concentration in RB-SiC after irradiation with 2 MeV Au^{2+} ions to a dose of 1×10^{16} ions/cm^2.

The irradiation-induced crystalline-to-amorphous transition was probed by high-resolution transmission electron microscopy (HR-TEM), also employing the selected-area electron diffraction (SAED) technique, using a Tecnai G2 F20 transmission electron microscope operated at 200 kV. The potential phase transformation which may produce the excess stress σ_{ex} was investigated by grazing-angle incidence X-ray diffraction (GIXRD) using a Philip X'Pert diffractometer equipped with a Cu $K\alpha$ source (λ = 1.54178 Å, 40 kV, 40 mA). During the measurements, the incidence angle was fixed at 1°, and the scanning range was from 20 to 80°.

The determination of the stresses by Raman spectroscopy is based on the piezo-spectroscopic effect, i.e., the spectral shift of the Raman peaks under stress [27]. The Raman spectroscopy measurements were performed on a Reinshaw inVia Raman microscope at RT using an Ar laser with an excitation wavelength of 514.5 nm (30 mW). The incident and scattered beams were focused with a 20 × objective lens, which resulted in a laser spot diameter of ~5 μm.

The surface morphology was studied by SEM using a Hitachi S-4700 field-emission scanning electron microscope. The nanoindentation tests were performed using a Berkovich diamond hard tip, and the indentation depth was kept constant at ~1 μm. The hardness (H) and Young's modulus (E) were extracted from the load-displacement curve using the Oliver-Pharr method [28]. A Netzsch DIL 402E dilatometer was used for measuring the CTE of each sample. The CTEs were measured under helium atmosphere over the temperature range from RT to 1200 °C using a heating rate of 5 °C/min.

3. Results

3.1. Evaluation of the irradiation-induced amorphization

For the irradiation with 2 MeV Au^{2+} ions at a dose of up to 1×10^{16} ions/cm^2 at RT, Fig. 1 reveals that the displacement damage in RB-SiC predicted by the SRIM software is ~13.543 dpa at the surface and reaches its peak value of 45.181 dpa at a depth of 270 nm. The corresponding peak Au^{2+} concentration was 0.7668 at.% at a depth of 340 nm. Fig. 2a depicts the corresponding bright-field cross-sectional TEM image with the SAED pattern shown on the

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