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Microstructural changes induced by low energy heavy ion irradiation in titanium silicon carbide

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Abstract

Low energy ion irradiation was used to investigate the microstructural modifications induced in Ti_3SiC_2 by nuclear collisions. Characterization of the microstructure of the pristine sample by electron back-scatter diffraction (EBSD) shows a strong texturing of $TiSi_2$, which is a common secondary phase present in Ti_3SiC_2. A methodology based on atomic force microscopy (AFM) was developed to measure the volume swelling induced by ion irradiation, and it was validated on irradiated silicon carbide. The swelling of Ti_3SiC_2 was estimated to $2.2 \pm 0.8\%$ for an irradiation dose of 4.3 dpa at room temperature. Results obtained by both EBSD and AFM analyses showed that nuclear collisions induce an anisotropic swelling in Ti_3SiC_2.

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1. Introduction

The Gas-cooled Fast Reactor (GFR) is one of the six new systems studied in the framework of the Generation IV International Forum (GIF). These systems are characterized by an increased security level, a better economic competitiveness, and the ability to recycle all the fuel in order to upgrade it to a fissionable material and to minimize long-lived waste production by transmutation.[1] The GFR is designed to work under helium-pressure and at high-temperature (1100–1300 K). Due to these working conditions, non-oxide refractory ceramics are required as fuel cladding. Thus, carbides turn out to be excellent candidates due to their remarkable mechanical and thermal properties. However, their behavior under irradiation has to be investigated.

Among potential carbides, ternary Ti_3SiC_2 presents some interesting properties. In 1972 Nickl et al.[2] remarked that this material is abnormally soft for a carbide, so that its hardness decreases as the applied load increases. For this reason Goto and Hirai[3] qualified Ti_3SiC_2 as a “ductile ceramic”. Furthermore, Ti_3SiC_2 combines the properties of metals with those usually attributed to ceramics.[4–7] Thus, this material is not only soft but also stiff and tough, it behaves as a good electrical and thermal conductor, and it can be easily machined with the tools generally used for steels.

The interesting mechanical properties of Ti_3SiC_2 suggest that this compound could be used as fuel cladding material. Its damage tolerance to mechanical stresses might indicate a high resistance to irradiation. Nevertheless, apart from few recently published articles related to Ti_3SiC_2,[8–10] and $Ti_3(Si,Al)C_2$,[11–14] few information is available about its behavior under irradiation.

Previously,[15,16] we showed that an irradiation performed at room temperature with 4 MeV Au ions to a fluence of 10^{19} m^{-2}

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Table 1
Irradiation conditions.

Temperature	Room temperature	773 K	1223 K
Fluence (m^{-2})	10^{16}, 10^{17}, 10^{18}, 10^{19}	10^{16}, 10^{17}, 10^{18}, 10^{19}	10^{19}

induces both an erosion of the Ti_3SiC_2 grain boundaries, as observed by scanning electron microscopy, and a revealing of the grain structure, as evidenced by atomic force microscopy. We attributed the former phenomenon to a preferential sputtering due to lower threshold displacement energy of the atoms located in grain boundaries. For the latter result, we were led to consider the occurrence of preferential sputtering as a function of the crystallite orientation. In this work, complementary irradiation experiments suggest another explanation.

2. Experimental

The polycrystalline samples were provided by the 3-ONE-2 company (Vorhees, NJ, USA). They consist of about 74% Ti_3SiC_2, 19% $TiC_{0.92}$, and 7% $TiSi_2$ (as estimated by X-ray diffraction). As-received samples were polished with diamond paste of a size down to 1 μm.

The interactions occurring in reactors are essentially elastic (or nuclear) collisions due to primary knock-on atoms from neutrons, and recoil atoms arising from alpha-decays. In order to simulate these interactions, low energy ion irradiations are usually performed. Thus, the polished face of the samples was irradiated with 4 MeV Au ions provided by the ARAMIS accelerator (CSNSM-Orsay, France). Table 1 summarizes the irradiation conditions.

In order to compare the results of these irradiations with previous data (especially those using neutron irradiations), it is usual to deal with the number of displacements per atoms (dpa) of the target, induced by the irradiation as a function of the depth within the irradiated material. The fluence scale has been converted into a dpa scale on the basis of TRIM-2008 calculations,[17] by considering the number of vacancies produced with 4 MeV Au ions as a function of depth in Ti_3SiC_2. The displacement energies were: 25 eV for Ti, 15 eV for Si, and 28 eV for C. Fig. 1

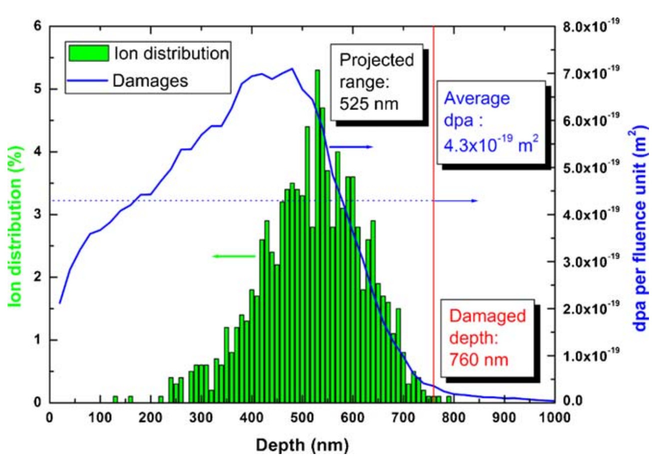

Fig. 1. Depth distribution of implanted ions and number of displacements per atom (per fluence unit) for Ti_3SiC_2 irradiated with 4 MeV Au ions.

Table 2
Wyckoff positions of the atoms for the three phases present in the studied material.

Phase	Ti_3SiC_2			TiC			$TiSi_2$	
Space group	P63/mmc(194)			Fm-3m(225)			Fddd(70)	
Atoms	Ti_I	Ti_{II}	Si	C	Ti	C	Ti	Si
Wyckoff positions	2a	4f	2b	4f	4a	4b	8a	16e

shows the variation of the damage level (in dpa per fluence unit) and of the ion distribution (also estimated with the TRIM code) as a function of the depth in the target material. This figure shows that the damaged thickness may be estimated to 760 nm. In this layer, irradiation induces an average dpa per fluence unit of 4.3×10^{-19} m^2, viz. 4.3 dpa for an irradiation to 10^{19} m^{-2}.

Different techniques were used to characterize the surface modifications induced by ion irradiation. Atomic force microscopy (AFM) aims both to analyse the surface topography modifications and to measure the swelling. Field emission gun scanning electron microscopy (FEG-SEM) was used to underline differences between Ti_3SiC_2 and the other phases by imaging the surface of samples with back-scattered electrons. Coupled to FEG-SEM, electron back-scatter diffraction (EBSD) was used to characterize the crystallites before irradiation. EBSD is a powerful technique for the quantification of both the microtexture and the microstructure of polyphased crystalline materials.

For EBSD, as-received samples were also polished with diamond suspensions down to 1 μm. Then, they were polished with ¼ μm colloidal silica suspension for 3 h. EBSD analyses were carried out using an HKL Technology (now Oxford Instruments) system installed on a Zeiss Supra 55 VP FEG-SEM operating at 17–20 kV and a probe-current of about 20 nA. EBSD analyses were not possible on irradiated samples because of the loss of crystallinity induced by nuclear collisions.[8,15]

Crystal structure data were created using the Twist add-on with the data shown in Table 2. In this table, Ti_I atoms correspond to the atoms of the basal planes linking the CTi_6 octahedrons, and Ti_{II} atoms to those bordering the silicon basal planes. Representations of the Ti_3SiC_2 lattice can be found elsewhere.[4–6] The complexity of the Ti_3SiC_2 diffraction patterns leads to the failure of the automatic indexation algorithm for some particular orientations. More precisely, the band recognition process using the standard Hough transform fails when Kikuchi bands are closely spaced and nearly parallel, which is the case of the Ti_3SiC_2 diffraction pattern. Nevertheless, the number of non-indexed patterns is relatively small, leading to a reliable microstructure analysis.

3. Results and discussion

3.1. Characterization of pristine samples

The characterization of as-prepared (or pristine) samples was partly reported previously.[15,16] Briefly, we first noticed a difference in the contrast between the three phases using back-scattered electrons in the FEG-SEM. Then, by AFM we were