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Heavy Ion Induced Damages in Ti_3SiC_2: Study of Swelling

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Abstract

For Generation IV reactors, and more particularly, the Gas Fast Reactor, ternary carbide Ti_3SiC_2 is an interesting candidate for the application as a fuel coating; actually, it has the advantage to combine some properties of metals with those generally attributed to ceramics. Unfortunately, few data are available on its behaviour under irradiation. In this study, we attempted to measure and to understand the origin of swelling induced by nuclear collisions. Thus, it seems that Ti_3SiC_2 irradiated at room temperature swells less than silicon carbide and that critical amorphization temperature is lower than room temperature.

Keywords: Ti_3SiC_2, irradiation, heavy ions, nuclear interactions, swelling, AFM

Introduction

Gas-cooled Fast Reactor (GFR) is one of the Generation IV International Forum systems studied by CEA (France). The considered coolant is helium pressurized at 7 MPa and the nominal temperature of use is about 1300 K. Because of the swelling of the fuel induced by the neutron irradiation, the cladding materials must meet certain requirements: refractory materials, toughness, mechanical resistance to prevent the release of the fission products, thermal conductivity higher than 10 W m^{-1} K^{-1} and good resistance to neutron damage. The materials currently considered are refractory carbides, and among these materials, ternary Ti_3SiC_2 may be distinguished for it combines the properties of ceramics with those of metals [1-8].

However, except one paper that we published some months ago [9], no information is available about its behaviour under irradiation. In this paper we have tried to estimate the swelling induced by irradiation thanks to atomic force microscopy, and to determine its origin.

Experimental

Sample preparation

The material studied is commercial polycrystalline Ti_3SiC_2 sintered under neutral atmosphere; it was provided by 3-ONE-2 (Voorhees, NJ, USA). With XRD, we estimated its composition: 80% of Ti_3SiC_2, 15% of TiC and 5% of $TiSi_2$. As-received specimen were polished with diamond suspensions down to 1 µm, and then subsequently irradiated with heavy ions.

Simulation with heavy ions

In the framework of this study we irradiated the specimens with 4 MeV Au ions provided by ARAMIS accelerator of the CSNSM of Orsay (France). These "low energy" ions are used to simulate the impact of alpha particles, and of both alpha and neutron recoils. The irradiations were carried out at room temperature (RT), 773 K and 1223 K for fluences of 10^{12}, 10^{13}, 10^{14} and 10^{15} cm^{-2}; fluence is the number of ions that impacts the target per unit area.

4 MeV Au ions induce essentially nuclear collisions, and so collision cascades, inside the material along the first micrometers. In order to compare the different irradiations in literature, the authors generally mention the number of displacements per atom (dpa) of the target: this ion-energy couple leads to an average of $4.3 \cdot 10^{-15}$ dpa per fluence unit, viz. 4.3 dpa for the highest fluence. The determination of the number of dpa was performed with TRIM-2008 code [10].

Characterization technique

To characterize the swelling, some specimens were partly irradiated: a mask covered a part of the samples in order not to damage it. Thus, with atomic force microscopy (AFM) we can measure the step height between the irradiated and the virgin parts and then estimate the swelling.

Estimation of the swelling

To estimate the swelling of a material induced by neutron irradiations, the authors used to compare the density after irradiation with the one before irradiation. This is due to the fact that neutrons irradiate the whole specimen and that swelling is induced by each place where the specimen is irradiated.

On the other hand, ions irradiate only a thin thickness of the material and the swelling induced by ion irradiation is usually considered as unidirectional, along the ion beam direction [11-13]. Thus to estimate such a swelling, the authors usually measure the step height induced by irradiation and compare it with the depth irradiated that induces the swelling. In this study, step height is measured with the atomic force microscope (AFM).

The authors evaluate this depth either by the average projected range [14], which is the mean of the distribution of the ions implanted in the material, or by the full depth spoiled [15]. In the case of 4 MeV Au ion irradiation in Ti_3SiC_2, these values are 525 nm and 1 µm, respectively, leading to a ratio of two between both of them. Our idea is to consider that all the damages lower than 5 % of the maximum of dpa are not significant enough to induce a measurable swelling and so to neglect the end of the depth spoiled. Thus, the depth inducing measurable swelling is of 760 nm for this irradiation in Ti_3SiC_2.

Results and Discussion

Swelling measurement

After irradiations, the interface between irradiated and virgin parts is only notable for the highest fluence of the irradiation performed at RT. Thus, the swelling increases when the fluence increases and when the temperature decreases.

On this sample, we measured several steps on grains partly irradiated by AFM and the result is an average step height of 16.8 ± 6.3 nm corresponding to a swelling of 2.2 ± 0.8 %.

In order to know if our estimation is relevant, we also irradiated silicon carbide specimens in the same conditions, leading to an average dpa of 3.1 along 800 nm. Thus, we measured an average step of 131 ± 10 nm, viz. 16.4 ± 1.3 % of swelling. This value is of the same order of magnitude as those in the literature; actually, depending on the authors, the swelling of SiC irradiated with at least 0.5 dpa at RT would be in the 10-20 % range [15-20]. Therefore, we can conclude that this method allows us to have good estimation of the swelling induced by ion irradiation.

Origin of swelling

To grasp the origin of the swelling of Ti_3SiC_2 under irradiation, we compared it to those of SiC and AlN.

Snead *et al.* summarized the variation of the swelling of SiC with its origin as a function of irradiation temperature (see Fig. 1 [21]).

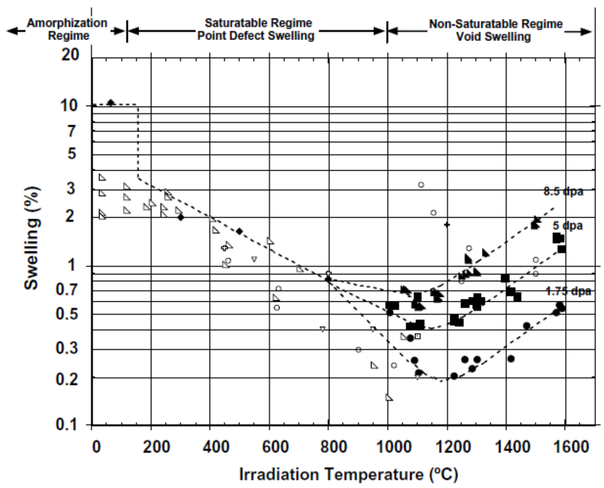

Fig. 1 Irradiation-induced swelling of SiC to high irradiation temperature.

Thus, he defined 3 regimes. First, for irradiation temperatures below the critical amorphization temperature T_c, from which amorphization is not reachable anymore even for high fluences (T_c = 400-650 K for SiC [20, 22-27]), silicon carbide strongly swell (10-20 %, see above) because of the formation of amorphous areas that grow up to the surface: this is the amorphization regime. Second, for the irradiation temperature higher than Tc, amorphization does not occur anymore, and the sole creation of point defects induces the swelling, which is so less important (some percents): this is the point defect regime. Third, when the irradiation temperature is high enough (> 1200-1300 K [21, 28-30]), the defects are mobile enough to create clusters, which can lead to void formation. These extended defects constitute the main contribution to the swelling which is more important, and increases when temperature and/or fluence increase: this is the void regime.

On the other hand, Yano *et al.* showed that aluminum nitride swelling induced by irradiation is directly related to the swelling of the unit cell volume [31,32]. Moreover, they demonstrated that the swelling of its wurtzite structure is anisotropic: unit volume increases when fluence increases. This anisotropic swelling, also noted in other materials that have anisotropic structures [33-37], leads to stress in the irradiated area, inducing, in polycrystalline material, fractures or microcracks at the grain boundaries (see Fig. 2 [38]).

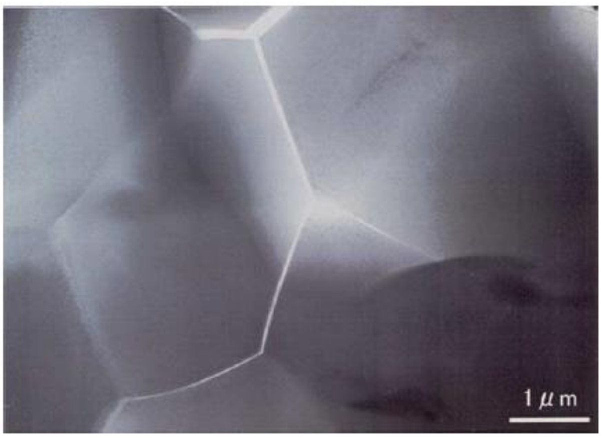

Fig. 2 Transmission electron micrograph of as-irradiated AlN (fast neutrons, E > 0.1 MeV, 17 dpa).

In the previous paper [9], we noticed neither amorphization nor change in the Ti_3SiC_2 lattice parameters by low-incidence X-ray diffraction. Moreover, the first observations of cross-sections by transmission electron microscopy allowed us to confirm the absence of amorphous phase, but also to note the formation of many black dots (aggregates of point defects). Adding to this the fact that the swelling is low compared to the one of SiC irradiated under the same conditions, the swelling of Ti_3SiC_2 is certainly due to the creation of these black dots and so occurs in the point defect regime. Thus, the critical amorphization temperature would be lower than room temperature. Eventually, in the same paper we mentioned the presence of cracks along the grain boundaries (see Fig. 3 [9]). Ti_3SiC_2 having a hexagonal crystalline structure, anisotropic swelling like for AlN would explain this crack formation.

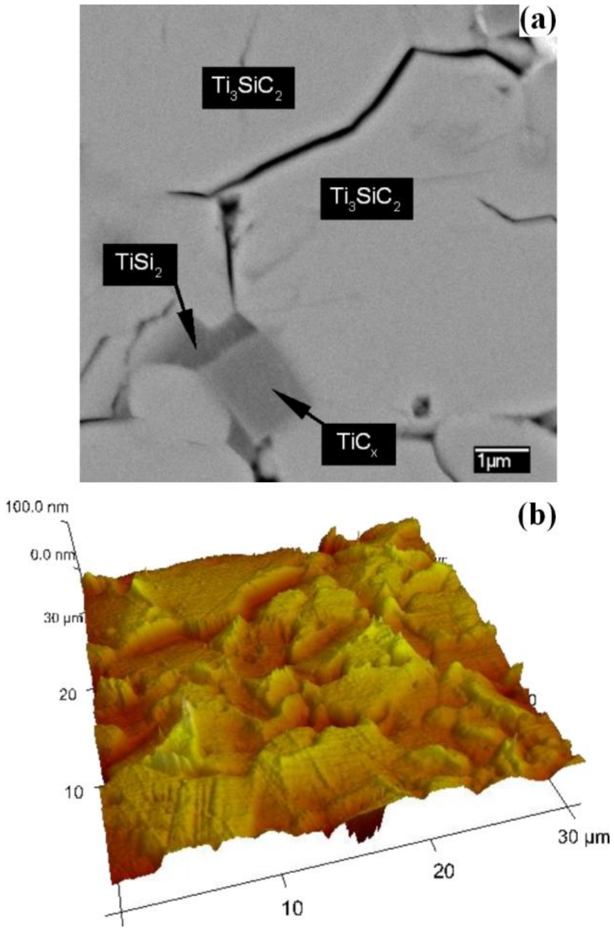

Fig. 3 Surface of Ti_3SiC_2 after 4 MeV Au irradiation at room temperature for a fluence of 1015 cm^{-2} by SEM (a) and AFM (b).

Conclusions

In this study, we set up a protocol to estimate the swelling induced by ion irradiations. This protocol seems to provide swelling in agreement with the results available in the literature obtained for neutron irradiations.

Thus, we showed that the swelling of Ti_3SiC_2 increases when fluence and/or temperature increases. That is why we could only measure the swelling on the specimen irradiated with 10^{15} cm^{-2} of 4 MeV Au ions at room temperature: 2.2 ± 0.8 %.

Eventually, comparing the different results obtained on our specimen with other techniques [9] with those obtained for SiC and AlN in literature, we concluded that at room temperature Ti_3SiC_2 is in the point defect regime ($T_c < RT$) and that its swelling is anisotropic.

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