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HEAVY IONS INDUCED DAMAGES IN Ti₃SiC₂: EFFECT OF IRRADIATION TEMPERATURE

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ABSTRACT

Thanks to their refractoriness, carbides are sensed as fuel coating for the IVth generation of reactors. Among those studied, the Ti₃SiC₂ ternary compound can be distinguished for its noteworthy mechanical properties: the nanolamellar structure imparts to this material some softness as well as better toughness than other classical carbides such as SiC or TiC. However, under irradiation, its behaviour is still unknown. In order to understand this behaviour, specimens were irradiated with heavy ions of different energies, then characterised. The choice of energies used allowed separation of the effects of nuclear interactions from those of electronic ones. Thus, AFM, SEM and XRD techniques allowed to note an important spoiling due to nuclear collision whereas electronic interactions would induce the formation of hills and the expansion of the unit cell. Irradiations at higher temperatures allowed to study the effect of temperature on these results.

Key words: Ti₃SiC₂, irradiation, heavy ions, AFM, SEM, XRD

INTRODUCTION

As part of Generation IV International Forum (GIF), new systems are studied, from the point of view of the reactor as well as the fuel cycle. These systems are characterised by an increased security level, better economic competitiveness, and an ability to recycle all the fuel in order to upgrade to a fissionable material and minimize long-lived waste production by transmutation. Among the six systems considered by the GIF, the Gas Fast Reactor (GFR) is studied in France; it is designed to work under helium-pressure and high-temperature (1100-1300 K).

These working conditions led to the selection of non-oxide refractory ceramics as fuel coating. Thus, carbides turn out to be great candidates thanks to their remarkable mechanical and thermal properties. However, their behaviour under irradiation has to be studied in more details.

Among the studied carbides, the ternary Ti₃SiC₂ presents some interesting prospects. Indeed, in 1972 Nickl *et al.*¹ remarked that this material, which was synthesized for the first time by a Viennese group in 1967², is abnormally soft for a carbide. Actually, Pampuch *et al.*^{3,4} and Lis *et al.*⁵⁻⁷ demonstrated that this material is both stiff (Young's modulus of 325 GPa) and soft (Vickers hardness of 6 GPa). Moreover, like Goto *et al.*⁸ some years before, they noted that the hardness of this carbide decreases as the applied load increases; this property led them to qualify Ti₃SiC₂ as a “ductile ceramic”.

These interesting mechanical properties led to think about Ti₃SiC₂ as fuel coating material; actually, its ability to be damage tolerant against mechanical solicitations was expected to be true against irradiations. Nevertheless, besides one paper to be published⁹, no information is available about its behaviour under irradiation.

Therefore, the aim of this study is a better knowledge of the behaviour under irradiation of Ti_3SiC_2 , and more particularly the effect of the irradiation temperature.

EXPERIMENTAL

The studied material is a polycrystalline commercial compound provided by 3-ONE-2 (Voorhees, NJ, USA). Pure bulk of Ti_3SiC_2 is quite difficult to synthesize: the as-received material consists of about 80 % Ti_3SiC_2 , 15 % TiC , and 5 % TiSi_2 (estimation by X-Ray Diffraction). Figure 1 shows the surface state of the as-polished specimens through Atomic Force Microscopy (AFM); the lower hardness of Ti_3SiC_2 compared with the TiC and TiSi_2 phases can be noted.

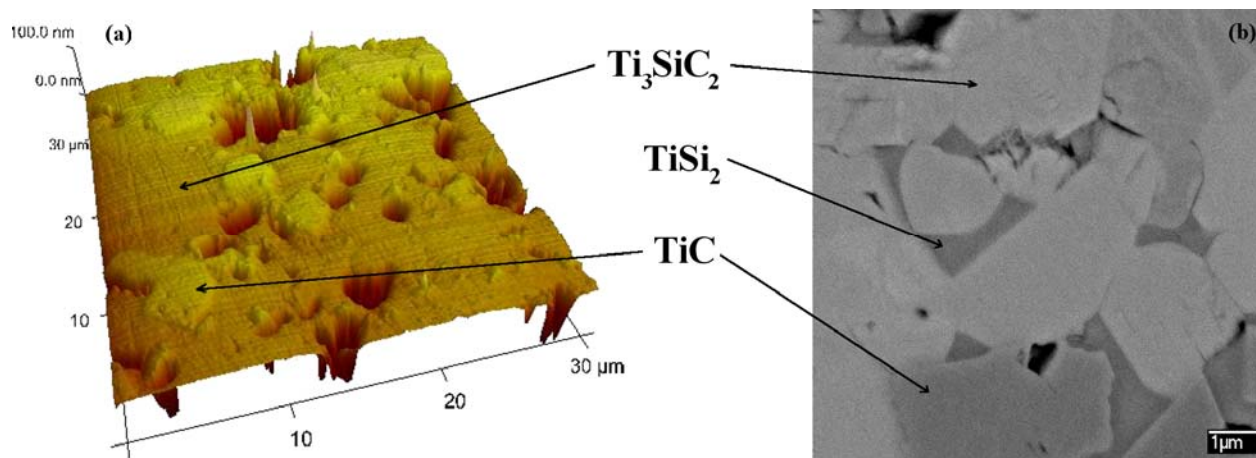


Figure 1. Surface of as-polished Ti_3SiC_2 by (a) AFM and (b) back-scattered electrons FEG-SEM.

The as-received specimens were polished with diamond suspensions down to 1 μm . They were subsequently irradiated with heavy ions. This kind of irradiation aims to simulate the interactions present in reactors (impacts of fission products, recoil atoms of alpha-decays, and neutrons) without the drawback of activating the specimens.

When heavy ions penetrate through a solid, they get decelerated *via* two different processes, namely, elastic collisions with target nuclei (with nuclear stopping power S_n) and inelastic collisions with target electrons (with electronic stopping power S_e). In order to understand the effects of both nuclear and electronic interactions on Ti_3SiC_2 , two kinds of irradiation were performed.

The first one, named “high energy irradiation”, was made with ions delivered by the IRRSUD accelerator at the GANIL (Caen, France). The energy of these ions reaches several dozen of MeV; for this range of energy, ion path consists in a long zone of electronic interactions (some microns) at the end of which nuclear collisions occur. Thus, studying only the first microns, effects of electronic interactions can be understood. In order to study the effect of electronic stopping power, two different ions were used at the GANIL: 74 MeV Kr and 92 MeV Xe ions, which respectively lead to a maximum electronic stopping power of 15.6 and 21.3 keV nm^{-1} . Irradiations with Kr were carried out at room temperature (RT) and 773 K whereas the ones with Xe were at RT and 573 K.

The second one, called “low energy irradiation”, was achieved with the 4 MeV Au ions of the ARAMIS accelerator at the CSNSM (Orsay, France). This kind of irradiation essentially leads to nuclear collisions. This irradiation was carried out at RT, 773 K and 1223 K.

Each kind of irradiation was carried out for 4 ion fluences: 10^{12} , 10^{13} , 10^{14} , and 10^{15} cm^{-2} ; the fluence is the number of ion by surface unit. The Table I gives some irradiation parameters determined by the TRIM code¹⁰.

Table I. Irradiation parameters

Ion	Au	Kr	Xe
Mass (amu)	197	86	129
Energy (MeV)	4	74	92
Projected range (μm)	0.52	7.81	7.60
Nuclear stopping power (keV/nm)	3.29	0.07	0.16
Electronic stopping power (keV/nm)	4.0	15.6	21.3

To characterise surface modifications to the samples, two microscopic methods were used: Field Emission Gun Scanning Electron Microscopy (FEG-SEM) and AFM. The structure of the irradiated zone was analysed by Low Incidence X-Ray Diffraction (LI-XRD) technique.

RESULTS

Low Energy Irradiations

The irradiations performed with 4 MeV Au ions essentially lead to nuclear shocks, which generally cause an important disorder in the materials. As noted elsewhere⁹, this disorder is visible on diffractogrammes from a fluence of 10^{14} cm^{-2} for irradiations performed at RT, whatever the phase.

The samples irradiated at high temperatures (773 K and 1223 K) have not yet been characterized by LI-XRD. Nevertheless, we hope to observe a decrease of disorder due to a greater mobility of atoms making up the samples and to an annealing of the defects by the temperature. This decrease would be a good result if we consider the nominal operating temperatures of the GFR.

In terms of surface conditions, no changes were found before a fluence of 10^{15} cm^{-2} . At room temperature, we noticed a microstructure that we attributed to a sputtering depending on the orientation of crystallites⁹. It is interesting to note that this microstructure is less pronounced during irradiation carried out at 773 K, regaining a microstructure similar to the as-polished specimen for irradiation at 1223 K (cf. Figure 2).

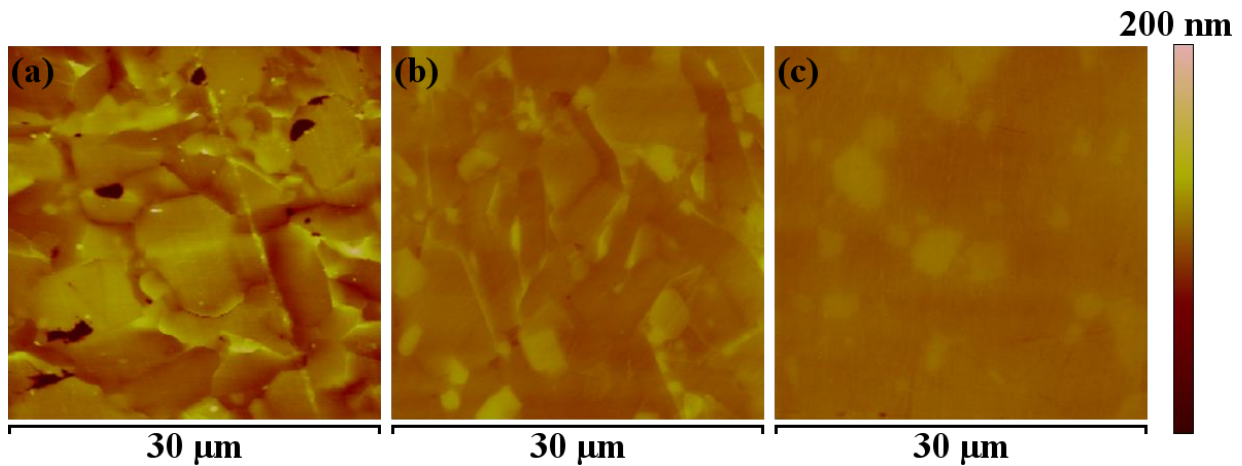


Figure 2. Surface of Ti_3SiC_2 after a 10^{15} cm^{-2} 4 MeV Au irradiation at (a) RT, (b) 773 K, and (c) 1223 K.

High Energy Irradiations

Concerning the high-energy irradiations, we noted by LI-XRD some disorder from 10^{14} cm^{-2} for 92 MeV Xe irradiation at RT, but only for phase Ti_3SiC_2 ; this disorder is noticeable through a rise of the baseline. Moreover, we noticed an expansion of the hexagonal unit cell of Ti_3SiC_2 along c axis without change in the unit volume⁹. This result is also true for 74 MeV Kr irradiation at RT.

The temperature has two effects on XRD results. First, a drop-off of the rise of baseline shows a decrease of the disorder as a function of temperature. Second, the unit cell is less expanded for irradiation with 92 MeV Xe at 573 K to be almost unchanged compared with virgin sample for the irradiation with 74 MeV Kr at 773 K. Note that whatever the irradiation, the change of the unit cell parameters occurs at constant unit volume (cf. Figure 3).

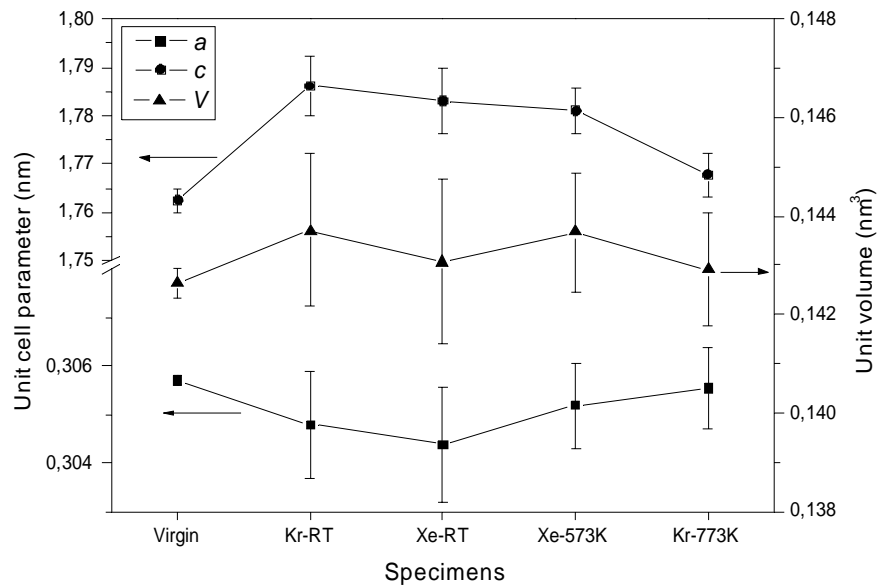


Figure 3. Evolution of Ti_3SiC_2 unit cell parameters as a function of irradiation parameters for high energy irradiation; lines are only to guide the eye.

With regard to the change in the surface, the first notable point that, unlike irradiation with 92 MeV Xe at RT, the 74 MeV Kr irradiation does not cause changes whatever the fluence or temperature. Concerning the 92 MeV Xe irradiation, we underlined the formation of hills on Ti_3SiC_2 phase and a revelation of the microstructure due to a different behaviour under irradiation of the secondary phases against Ti_3SiC_2 ⁹. The difference that is noted for these two high-energy irradiation suggests the existence of an electronic stopping power threshold beyond which there is modification of the surface of the samples.

At 573 K, the surface state of our samples after 92 MeV Xe irradiation is quite similar to irradiations at RT. However, if the revelation of microstructure is always obvious, we can notice that the shape of the hills slightly varies; actually, as shown in Figure 4, they seem larger than for irradiation at RT. This coalescence phenomenon could be compared with the one noted during sintering, suggesting a transfer of matter by temperature.

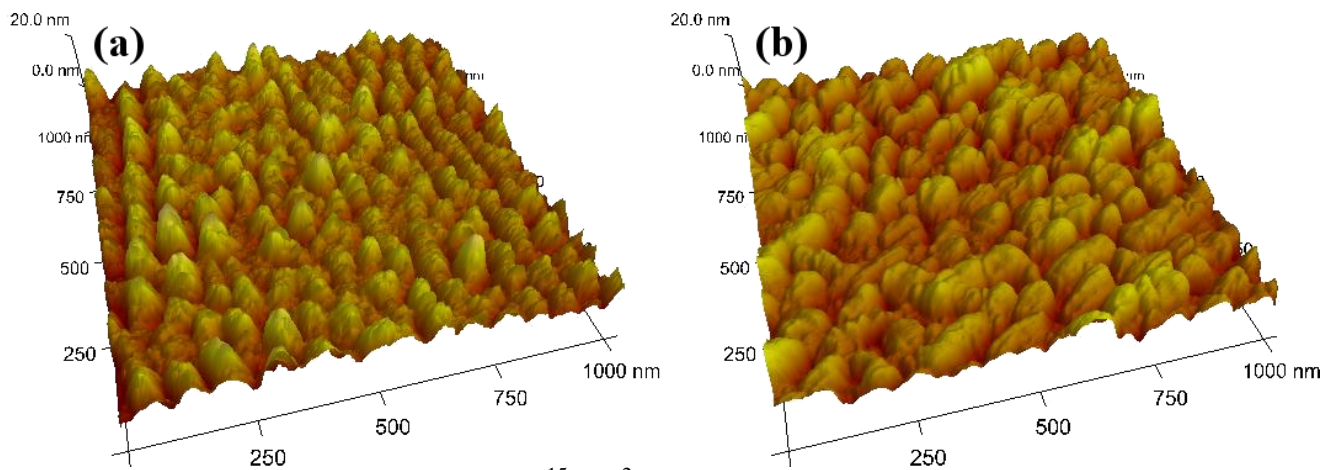


Figure 4. Surface of Ti_3SiC_2 after 10^{15} cm^{-2} 92 MeV Xe irradiation at (a) RT and (b) 573 K.

CONCLUSION

In this study, we demonstrated a different behaviour of Ti_3SiC_2 under low energy and high energy irradiation. Thus, if the nuclear shocks tend to create an important disorder as well as a sputtering of the surface, the electronic interactions essentially cause an expansion of the unit cell along c axis and the formation of hills beyond an electronic stopping power threshold between 15.6 and 21.3 keV nm^{-1} .

Temperature seems to have two different effects. The first one, as it can be noticed in other carbides like SiC^{11-14} , is an annealing effect of the irradiation defects: decrease of disorder whatever the irradiation, of unit cell expansion due to high energy irradiations, and of the surface changes for the 4 MeV Au irradiation. The second one is the enlarging of the hills. We understand this effect as a transfer of matter due to both the temperature and the electronic interactions.

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