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Molecular dynamics simulation of defect production in irradiated β-SiC

L. Malerba [a,*], J.M. Perlado [a], A. Sánchez-Rubio [a], I. Pastor [a], L. Colombo [b,1], T. Diaz de la Rubia [c]

[a] *Instituto de Fusión Nuclear (DENIM), Universidad Politécnica de Madrid, C/José Gutiérrez Abascal, 2, Madrid 28006, Spain*

[b] *Dip. Scienza dei Materiali – Università degli Studi di Milano 'Bicocca', Via R. Cozzi, 53-20125 Milan, Italy*

[c] *Chemistry and Materials Division, Lawrence Livermore National Laboratory, L-268, P.O. Box 808, Livermore, CA 94550, USA*

Abstract

We used the molecular dynamics (MD) code MDCASK, in which the Tersoff potential is implemented, to study the mechanisms of defect production due to Si- and C-recoils of different energies in β-SiC. In this paper, we highlight some of our most significant results to date. The threshold displacement energies (TDEs) for Si- and C-atoms have been accurately determined along four main crystallographic directions. The difficulty of defining a single TDE in this material, also because of the effect of temperature, is discussed. High-energy (various keV) recoil-induced displacement cascades have been simulated and it was found that defects on the C-sublattice always outnumber defects on the Si-sublattice, temperature scarcely affecting the number of Frenkel pairs produced within the cascade. Finally, our MD model seems to have proved satisfactory in reproducing the experimentally observed process of irradiation-induced amorphization of SiC at cryogenic temperature. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

It is well known in the nuclear fusion scientific community that silicon carbide (SiC), thanks to its intrinsic low activation after neutron irradiation, is one of the few candidate structural materials for future fusion reactors [1]. It is therefore of crucial importance to evaluate carefully the effects of intense irradiation on this material. Atomistic computational simulations are nowadays a powerful tool to study, from the microscopic standpoint, the different phases of irradiation damage production and evolution [2]. In particular, it has been long demonstrated that the classical, empirical interatomic potential proposed by Tersoff enables the performance of reliable molecular dynamics (MD) simulations of the microscopic behaviour of covalent materials like SiC [3–8].

We used the MD code MDCASK, in which the Tersoff potential is implemented [3,4], to study irradiation damage production in β-SiC at different levels. Firstly, we analysed the basic mechanisms of defect production and determined the atomic threshold displacement energies (TDEs), i.e. the lowest atomic kinetic energy required to create a stable Frenkel pair in the crystal, regardless of the displaced atom being the primary recoil or a secondary one [9–11]. Secondly, we considered the production of damage by high-energy (various keV) recoil-atoms, analysing number and type of defects as a function of recoil type and energy, as well as temperature [10,11]. Thirdly, we tried to simulate within the temporal limits of MD (i.e. with no diffusion effects included), the consequence of accumulating damage in the material [9]. In particular, we made an attempt to reproduce computationally the process of electron-irradiation-induced amorphisation that has been experimentally observed in SiC after accumulating ~1 dpa at cryogenic temperatures, i.e. when practically no diffusion mechanism is active [14,15,19,20].

* Corresponding author. Tel.: +34-91 336 3108; fax: +34-91 336 3002.

E-mail address: malerba@denim.upm.es (L. Malerba).

[1] Present address: Dipartimento di Fisica, Università degli Studi di Caglieri, Cittadella Universitaria, S.P. Monserrato–Sestu km 0,700, 09042 Monserrato (CA), Italy.

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We already illustrated elsewhere the details of our methodology, as well as part of our preliminary results [9–11]. This paper is intended as a rapid communication of some of our most recent findings. Therefore, only a few strictly necessary hints about the computational method used will be given. The results reported will emphasise the role of temperature in the process of defect production.

2. Results and discussion

It has already been observed that, in SiC, it is difficult to define a single TDE because: (1) the TDE depends strongly on the crystallographic direction for both Si- and C-atoms and (2) Si-atoms' TDEs are generally higher than C-atoms' [9–12]. Moreover, we already suggested the existence of recombination barriers, whose presence allows the formation of metastable atomic displacements even when the kinetic energy gained by the atom hit (primary knock-on atom, PKA) lies *below* the actual threshold [9–11]. In Fig. 1, the simplest possible picture of this situation is synthesised, according to a thorough revision of our results. Along each main crystallographic direction (with only one exception) there exists an *uncertainty band*, within which an atomic displacement may or may not be produced. When the atom struck receives a kinetic energy within the uncertainty band, the only defect that may be created is metastable. A small perturbation (e.g. due to thermal agitation at high temperature or to interaction with a secondary recoil at low temperature) is sufficient to enable the displaced atom to overcome the recombination barrier, thereby causing the prompt disappearance of the defect, or even preventing its formation. Only kinetic energies above the *upper threshold* always lead to the production of a stable defect. As already discussed elsewhere [9–11], the TDEs reported in other work [7,8,12] coincide mostly with our lower thresholds, discrepancies being attributable to different criteria in defining the TDE [9].

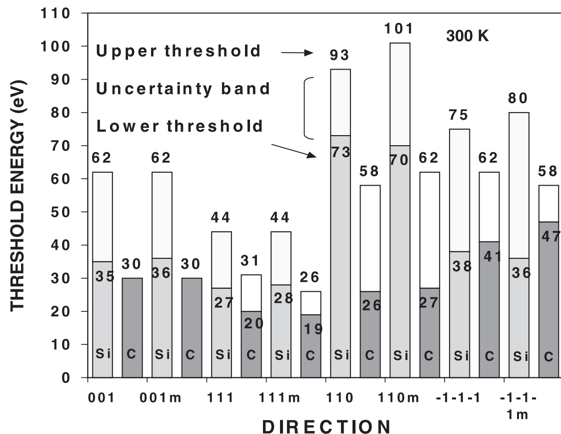

Fig. 1. Upper and lower TDEs and uncertainty bands for Si and C atoms in β-SiC at 300 K along the four crystallographic directions indicated. For each direction, both geometrical and slightly modified angles (*m*) were considered.

The metastability of the defects created within the uncertainty band has been definitively established by computing the lifetime of the Frenkel pair formed when knocking on a Si-atom along direction [0 0 1]. The lifetime dependence on temperature is plotted in Fig. 2. The exponential fit of the data points, obtained by observing for how long the defect survived at different temperatures between 1200 and 2000 K, also permitted us to assess the height of the recombination barrier, ~1.16 eV. In addition, it was observed that both upper and lower thresholds diminish while the temperature of the simulation increases up to 2000 K, the decrease being more pronounced for the upper one (50 eV at 2000 K for Si atom along direction [0 0 1]), so that the uncertainty band also shrinks. These effects are ascribable to thermal agitation. Within the uncertainty band the production of displacement becomes, as expected, less and less probable as the temperature is raised, so that at high temperature the upper threshold should be the only experimentally observable TDE along the relevant direction.

Temperature also affects the number of atomic displacements and replacements per 3-keV Si–PKA cascade, as shown in Fig. 3 (the influence of the type of recoil and of its energy was already presented elsewhere [10,11]). The number of displacements, both at the peak and end of the cascade, grows while the temperature

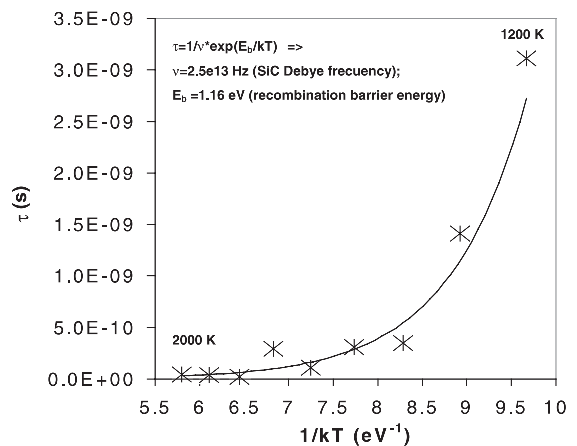

Fig. 2. I_{Si}–Si ⟨0 0 1⟩ dumbell + Si vacancy Frenkel pair (distance $I_{Si}-V_{Si} = 0.775a_0 = 3.38$ Å) lifetime versus temperature (1/*kT*) in the 1200–2000 K range. This defect is created in Si-atoms' [0 0 1]-direction uncertainty band. The symbols represent the computed data; the line gives their best exponential fit, whose mathematical expression is also given in the figure.

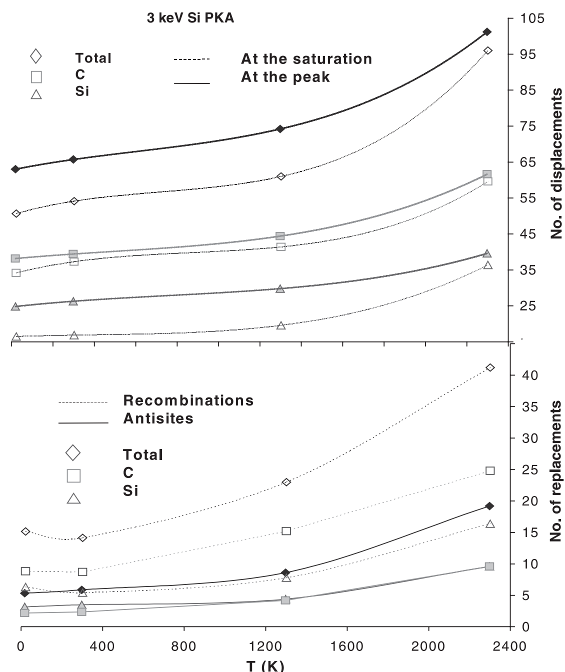

Fig. 3. Displacement (both at the peak and end of the cascade – upper graph) and replacement (antisites and recombinations at the end of the cascade – lower graph) number versus temperature in 3 keV Si–PKA cascades. Each point is the result of averaging five or more cascades. Error bars are not shown for better legibility.

increases, in accordance with the decrease of the TDEs. Also the number of replacements (included in the number of displacements), both 'right' (recombinations) and 'wrong' (antisites) grows, clearly thanks to the enhanced mobility of the atoms and to the fast disappearance of metastable Frenkel pairs. The net effect of this double growth with increasing temperature is that the number of interstitials and vacancies produced within the cascade (not shown in figure) remains practically constant for all temperatures, the larger number of atomic displacements at high temperature compensating for the disappearance of metastable Frenkel pairs. It is also useful to note, in Fig. 3, that displaced C-atoms greatly outnumber displaced Si-atoms, particularly at the end of the cascade. As it is, according to our simulations the defects produced on the C-sublattice always outnumber the defects on the Si-sublattice, the fraction of recombined atoms (number-of-recombined-atoms/ number-of-displaced-atoms) being normally higher for Si-atoms than for C-atoms [9–11]. Analogous findings were reported by other authors [13].

It is difficult to find experimental results comparable with our simulations. A possibility was offered by the process of irradiation-induced amorphisation, studied in the last decade by various authors bombarding β-SiC at low temperatures with both electrons [14,15] and ions [16–22]. In particular, using 2-MeV-electrons Inui et al. found that crystalline-to-amorphous transition in β-SiC could be induced at temperatures below ∼340 K [14]. Similar results had been obtained before by Matsunaga et al. [15]. The dose required is essentially constant below 250 K ($\sim 1.6\times 10^{26}$ e/m^2, or ∼1 dpa, as calculated by Weber for Inui's results [19] and by Zinkle for Matsunaga's [20]), but then increases, until no amorphisation can be induced any more above a critical temperature. Prior to the transition Inui could detect a 10% antisite concentration and estimated a long-range-order (LRO) parameter of ∼0.9. Moreover, according to a review by Wendler et al. [21], the critical energy density for amorphisation of SiC, obtained by averaging results from different authors, is ∼20 eV/atom at liquid-nitrogen temperature (77 K).

We tried to simulate the electron-irradiation process by introducing low-energy (100 eV) Si- and C-recoils in a 512-atom sample contained in a constant volume simulation box at constant temperature (20 K). Previously, we had computationally produced and analysed a reference amorphous-SiC (a-SiC) sample, by simply heating and then carefully cooling down at constant volume the 512-atom box. After accumulating a dose of 0.94 dpa we compared the pair distribution function, $g(r)$, at the end of the simulated irradiation process, with the same function for the reference a-SiC (Fig. 4). The close resemblance between the two curves allows us to say that the damage accumulated by irradiating did produce something very close to, if not coincident with, an amorphous state. This statement is further supported by Fig. 5, where the evolution of the absolute short-range-order (SRO) parameter versus dpa is shown. Though the shape of the curve still needs appropriate interpretation, the SRO parameter does tend to get closer and closer to the value encountered for the reference a-SiC.

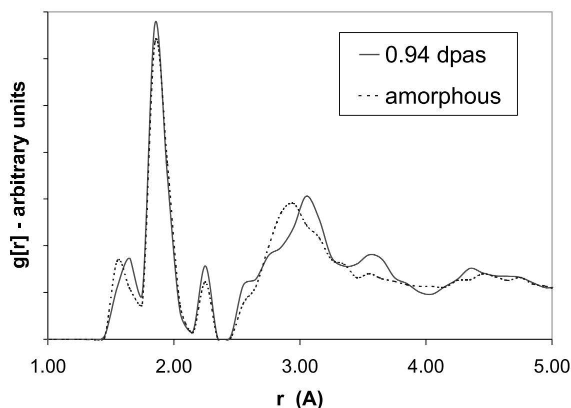

Fig. 4. Pair distribution function in the computational sample of β-SiC after accumulating 0.94 dpa, compared with the same function for the reference a-SiC.

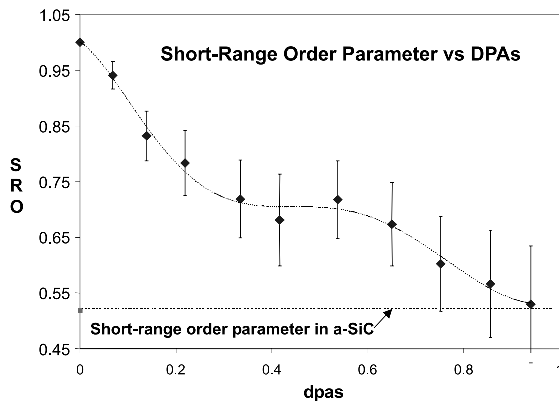

Fig. 5. SRO parameter as a function of accumulated damage in the computational sample of β-SiC. The straight line at 0.52 corresponds to the disorder level in the reference a-SiC. The SRO parameter plotted is an average of four different definitions found in the literature; the error bars correspond to the standard deviation.

To accumulate 0.94 dpa we had to introduce 120 recoils, thereby transferring to the sample 23.4 eV/atom. The total simulated time was 1 ns, so that the (unrealistic) overall dose-rate was $\sim 10^9$ dpa/s. However, at the end of the irradiation process we too found a 10% antisite concentration, corresponding to a LRO parameter of 0.9. The concentration of Frenkel pairs, on the other hand, was as high as 20%. A deeper discussion and more thorough comparison between experimental observations and simulation has to be postponed to a future paper. Nevertheless, we believe that the simulation has proved to be able to reproduce quite satisfactorily experimentally obtained numbers. As it is, the unrealistic dose rate of $\sim 10^9$ dpa/s does not render meaningless the comparison with experiment for two reasons. Firstly, though it has been seen that the dose rate does have an effect on amorphisation dose in *real* SiC at ambient temperature [22], at cryogenic temperature both in *real* and *computational* SiC all diffusive mechanisms are supposedly frozen, so that such effect should be negligible. Secondly, we observed evidence of extremely low mobility of defects (from the MD-timescale standpoint) in *computational* SiC even at temperatures as large as 1300 K [9]. Therefore, we deem that the same result would have been obtained even allowing a much longer time to elapse between PKA events and achieving a much lower simulated dose rate, at unaffordable computation time and cost.

3. Conclusions

We have studied, using MD computer simulations, the mechanisms of irradiation damage production in β-SiC. We have shown that, due to the existence of recombination barriers, the TDEs for Si- and C-atoms in SiC depend not only on the type of recoil and its direction, but also on temperature. We have also shown that, on the contrary, the number of Frenkel pairs produced within a displacement cascade is not much affected by temperature. Finally, our MD model appears to have proved to be satisfactorily reliable in reproducing the experimentally observed process of irradiation-induced amorphisation of SiC at cryogenic temperature. Hence, it is expected that a deeper analysis of the simulation results should help cast some light on the microscopic mechanisms governing this phenomenon in SiC.

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