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The threshold energy for defect production in SiC: a molecular dynamics study *

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Abstract

We discuss the results of molecular dynamics computer simulation studies of the threshold energy for defect production in β-SiC. The simulations are performed with the Tersoff potential for SiC which provides accurate values of many of its defect properties. In addition, we show that it properly describes the melting behavior of SiC. Simulations were carried out for Si and C recoils in three-dimensional cubic computational cells with periodic boundary conditions and up to 4096 atoms. The results show anisotropy in the threshold for Si and C recoils as well as for the recoil direction. The lowest threshold is 25 eV for C recoils along [111] and the highest is 85 eV for Si recoils along [110]. Details of the defect configurations obtained will be discussed.

1. Introduction

In the last few years, SiC has been proposed as a structural component for the first wall of fusion reactors [1]. The most recent magnetic [2] and inertial [3] fusion reactor studies all incorporate it in their designs, either in the form of monolithic β-SiC or as SiC/SiC ceramic-matrix composites. A critical property for the increasing interest in SiC-based materials for fusion reactors is their low activation under neutron irradiation [4]. Low activation materials must conform to severe standards regarding their behavior under neutron irradiation. In particular, waste management, accident safety, and maintenance considerations are most important. SiC provides a most encouraging solution, showing a significant reduction in the end-of-life radioactive inventory [4]. Also, their high temperature resistance, low density and coefficient of expansion, together with a good thermal conductivity and corrosion behavior, make these materials an excellent prospect for future fusion reactors [5].

Despite the promise of SiC-based materials as low activation components in fusion reactors, little is known about their response to high energy neutron irradiation. Recent work [6–8] has begun to assess the radiation damage-induced property changes in these materials. Of concern is the lattice swelling that takes place between 60 and 1000°C [9]. This results from volume changes arising from the introduction of defects and chemical disorder. Above 1000°C, SiC exhibits void swelling [9,10], similar to metals but at a considerable lower peak rate of $\Delta V/V \approx 0.2\%/\text{dpa}$. Also important for reactor considerations is the observed differential amorphization rate and modulus change of the matrix and the fiber in the composites [11].

In inertial fusion reactors, sub-μs fluence bursts of $\approx 10^{13}$ neutrons/cm^2 are expected to reach the first wall as frequently as every 0.1 s, after being moderated by blanket materials [12]. Therefore, detailed knowledge of the primary state of damage and the diffusion properties of the point and extended defects present is

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needed to predict the effect of the pulsed nature of the irradiation.

The basic displacement processes together with a complete understanding of the structure and properties of point and extended defects in SiC is key to predicting its evolution and that of SiC-based composites under neutron irradiation. A fundamental quantity critical to the development of a better understanding of radiation effects in multicomponent materials is the average value of the threshold energy for defect production, E_d. E_d is used in calculations of the defect production efficiency and therefore of the standard unit of damage, i.e., the number of displacements per atom (dpa) [13].

Molecular dynamics (MD) computer simulation is an appropriate tool that can help develop this fundamental understanding. In an MD simulation, the classical equations of motion of an ensemble of particles are solved [14]. These particles are assumed to interact through an interatomic potential chosen to represent and describe the physical and chemical properties of the system of interest. El-Azab and Ghoniem [15] have used MD simulation to study the threshold energy for defect production in β-SiC. They employed the so-called Pearson potential [16]. In this paper, we employ the Tersoff interatomic potential [17] to study the threshold energy for defect production, the point defect geometry, and the melting properties of SiC. Our results indicate that while a large difference in E_d exists for recoils in the Si and C sublattices along the [110] direction, E_d has very similar values for recoils in the other low index crystallographic directions, namely the [001] and [111]. In addition we present results on the nature and geometry of the defects produced by these recoils and discuss the melting properties of this model of SiC. We show that Tersoff's potential accurately reproduces the melting behavior of SiC. This gives us confidence that this model is adequate for radiation damage studies of this material.

2. Model

The molecular dynamics simulations described in this paper were performed with the computer code MOLDYCASK [18]. The crystals employed are three-dimensional β-SiC boxes with periodic boundary conditions. Crystal sizes ranged from 1728 to 4096 atoms. All simulations were carried out in the microcanonical ensemble, i.e., at constant energy, volume and number of particles. For the studies of melting, the simulation boxes contained 284 atoms with free surfaces in the $+Z$ and $-Z$ directions and periodic boundaries elsewhere.

Atomic interactions were described with the Tersoff potentials for Si and C [17]. These potentials are short range and extend only to within the first and second neighbor shells. The potentials are described by a two-body interaction which includes a three-body angular dependent part. This contribution is critical for the modeling of covalent systems such as SiC.

The Tersoff potential properly models many of the point defect and dynamical properties of SiC [17]. The melting point of a material, in particular, is an important quantity in the determination of its radiation damage response [19]. We have examined in detail the melting behavior of Tersoff's SiC model and found that it provided very reasonable agreement with experiment. We observe a broad melting transition between 2500 and 3000 K. This is shown in Fig. 1 where the total energy of the system is plotted as a function of temperature. The latent heat of the transition is ~ 0.5 eV/atom. Because the Si and C sublattices disorder at different temperatures, the exact melting temperature is difficult to define in the simulations. Nevertheless, the experimental melting point of SiC is 2818 K [20], which gives us confidence that Tersoff's model of SiC is appropriate for radiation effects simulations.

The current version of MOLDYCASK takes advantage of the message passing architecture of multiple instruction multiple data (MIMD) massively parallel architectures. On one processor of a Cray YMP C90 at LLNL, the code requires 4×10^{-4} s/atom to complete a full MD step with the Tersoff potential.

The simulations were performed by first equilibrating a crystal of the desired size at 10 K for several hundred time steps. After equilibration, an atom was given the kinetic energy corresponding to the recoil under study. The direction of the recoil was chosen along one of the low index crystallographic directions, namely [100], [110], [111], or [−1, −1, −1]. Simulations were performed for both Si and C recoils. Along each of these directions, a series of recoil events at several

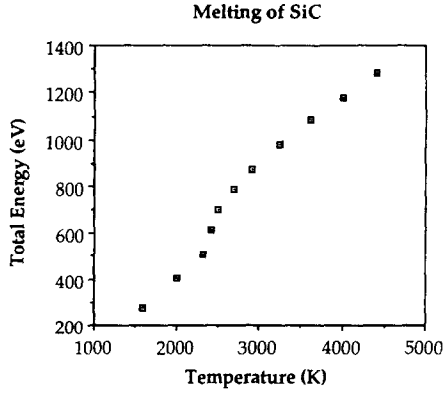

Fig. 1. Total energy of a SiC crystal as a function of temperature. The discontinuity indicates the temperature regime in which the phase transition occurs.

energies were run. The simulations were performed for times up to 3 ps with a minimum time step of 0.25 fs. This time step was allowed to vary as the simulation progressed up to a maximum time step of 2.5 fs. After each of these runs the crystal was examined for the presence of vacancy–interstitial pairs (Frenkel pairs). If none were found the recoil energy was increased and the simulation run again. In this manner, we were able to determine the threshold displacement energy of SiC to within 5 eV for each recoil direction. A few of the defect configurations were further investigated by annealing the crystal at 500 K for a short period and following this with a quench of the kinetic energy of all the atoms in the cell. The quench was performed by setting to 0 the velocities of those atoms for which the product of their velocity and acceleration was negative. In this manner, only those atoms pointing towards their equilibrium position are allowed to move with full dynamics. The system is therefore driven towards a low energy configuration. Typical quenching rates achieved in this manner are $\approx 10^{14}$ K/s.

3. Results

The results of the threshold displacement energy calculations are summarized in Table 1. For Si primary knockon atoms (PKA) along the [001] direction, the threshold is found between 30 and 35 eV. Fig. 2a shows the x, y, and z displacement of the 30 eV Si recoil as a function of time. As can be clearly seen, the atom moves along [001] but falls back into its original position after ≈ 0.2 ps. For the rest of the simulation, the PKA simply fluctuates around its lattice site. In Fig. 2b the result for a 35 eV Si PKA is displayed.

Table 1
Summary of the threshold energy for defect production on the Si and C sublattices of β-SiC along the low index crystallographic directions

Defect threshold energies		
Direction	PKA type	PKA energy (eV)
[001]	Si	35
	C	40
[111]	Si	35
	C	25
[110]	Si	85
	C	30
[$\bar{1}\bar{1}\bar{1}$]	Si	40
	C	50

Here, the z-coordinate of the PKA increases as the atom moves along [001]. In contrast to the previous case however, the PKA remains stable at a distance $\sim 0.8a_0$ from its original position. Detailed analysis of the data after 2 ps, shows that the PKA forms a [001] split interstitial with the atom originally in the replacement position. A vacancy is left at the PKA site. The distance between the atoms in the dumbbell is $0.5a_0$ Å.

For C recoils along [001], the threshold energy for defect production was found to be between 35 and 40 eV. The resulting Frenkel pair was composed of a C–C split [001] dumbbell interstitial with the two atoms separated by $0.3a_0$, and a vacancy at the C PKA site.

Along [111], no defects were produced for Si recoils at $E_{pka} \leq 30$ eV. At $E_{pka} = 35$ eV, a C–C [110] split interstitial was found with a C vacancy at the first neighbor site to the Si PKA. Fig. 3 illustrates this case in more detail. It is interesting to note that this event did not result in any net displacement of silicon atoms

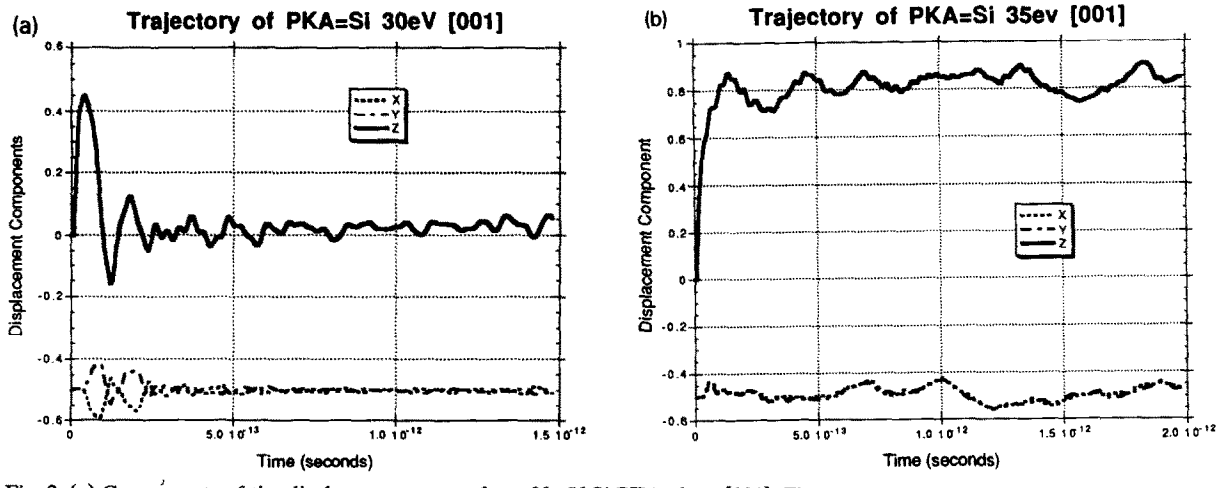

Fig. 2. (a) Components of the displacement vector for a 30 eV Si PKA along [001]. The atom moves in the Z direction but returns to its original position within ~ 0.25 ps. (b) Components of the displacement vector for a 35 eV Si PKA along [001]. In this case the atom does not return to its original position but forms a split dumbbel interstitial (see text for details).

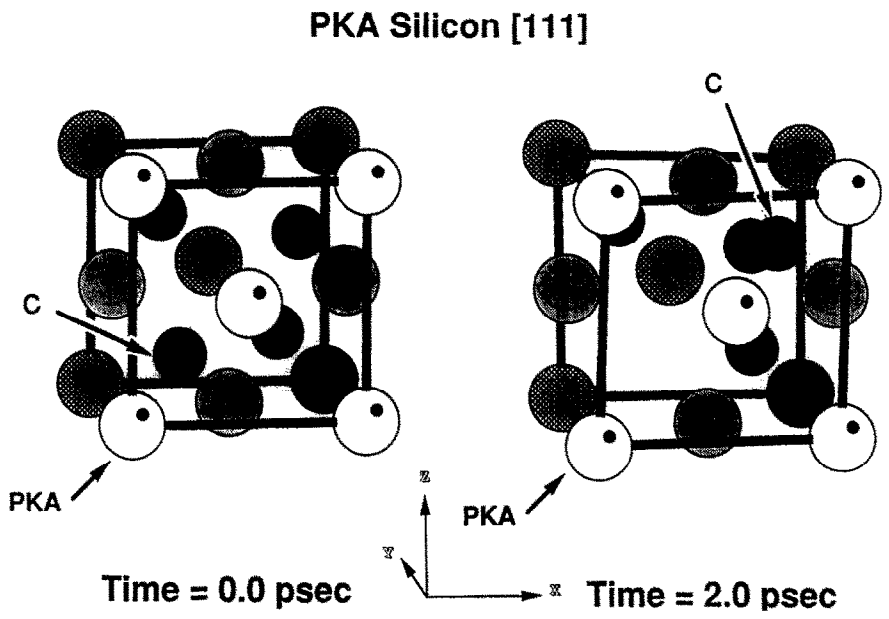

Fig. 3. Rendering of the unit cell in which a 35 eV Si PKA along [111] has been simulated (a) at $t = 0$, and (b) at the end of the event, $t = 2.0$ ps. The Si PKA resulted in a C vacancy and [110]-split C–C interstitial.

but produced a Frenkel pair in the C sublattice. For C recoils, the threshold energy for defect production was between 20 and 25 eV, lower than for Si PKAs. This may be understood in terms of the crystal symmetry. Along [111], the symmetry for C recoils is different than for Si. Here, the C PKA moves along the cube diagonal through the tetrahedral site without encountering any atom directly in its path for a distance of $3\sqrt{3}/4a_0$. The situation can be reversed by simulating trajectories along $[-1,-1,-1]$; here a Si recoil moves a distance $3\sqrt{3}/4a_0$ without encountering an atom directly in its path, while a C PKA must displace it nearest neighbor Si atom towards the tetrahedral site in the unit cell. For Si recoils along [111] the threshold energy was found between 35 and 40 eV. For C PKAs, no stable defects were formed up to $E_{pka} = 50$ eV.

Along the [110] direction is where the largest difference between Si and C recoils was found. For Si PKAs, the threshold for defect production was found between 80 and 85 eV. At 80 eV, the defect formed by this recoil was a C–C split interstitial with a C vacancy at the first tetrahedral neighboring site to the Si PKA. The Si PKA fell back to its original position. After a short annealing run at 500 K followed by quenching of the kinetic energy of crystal, this defect was observed to annihilate, indicating its metastable nature. At 85 eV however, the event resulted in a Si–Si split interstitial with a Si vacancy at the original Si PKA site. For C recoils along [110], a C–C split interstitial was found for C PKA energy of 30 eV. No defects were found at $E_{pka} = 25$ eV. The very high value of E_d in the silicon sublattice along [110] is consistent with our findings for pure silicon [21]. For this system, which was simulated with a different interatomic potential, namely the Stillinger–Weber potential [22], $E_d \approx 50$ eV along [110], a much higher value than that for the other low index directions which is ~ 20 eV.

4. Discussion and summary

It is interesting to note that for the cases described above, all collision chains are ~ 1–2 replacements long at most. In fact, only the case of an 85 eV Si PKA resulted in a replacement collision sequence longer that one atomic replacement. In that case, the PKA replaces its neighboring Si which in turn forms a [110] split interstitial with the next Si along the [110] line. It is also interesting to note that Si recoils along [111], the defects produced all reside in the C sublattice. It will be necessary however, to further investigate the role played by the short range nature of the Tersoff interatomic potential in stabilizing Frenkel pair configurations that might otherwise be unstable. For example, in the case of [001] recoils, the vacancy and the self-interstitial are only $1a_0$ apart. A longer range potential in which interactions extend to within the third and fourth nearest neighbor shells might result in the recombination of this Frenkel pair.

Also interesting is to compare our results to the simulation studies of El-Azab and Ghoniem [15]. Their results indicate that the value of the threshold energy

strongly depends on the sublattice on which the recoil takes place, as well as on the recoil direction. They found that for C recoils, the lowest value of E_d is ~ 10 eV along [111] while for Si recoils it is ~ 40 eV along [100]. Although the magnitude of the minimum E_d differs from our results, in particular for C PKAs, both studies agree on the fundamental observation that the lowest value of E_d for C is along [111] and for Si is along [100]. Moreover, both studies have found that the [110] direction is particularly difficult for Si displacements, but not so for C.

Potentially, the fact that the value of the threshold energy depends not only on the recoil direction, but also on the sublattice on which the recoil takes place, has strong implications for radiation damage calculations. The anisotropy based on the sublattice on which the recoil takes place brings into question the concept of dpa for multicomponent materials, since it then becomes invalid to define an average value of the threshold displacement energy of the system. It then becomes critical to extend these and other MD studies to check the validity of the classical force laws employed and to search for quantum mechanical based simulation models that can provide accurate information on defect energetics, stability and geometry.

Unfortunately, the lack of experimental results concerning E_d in SiC precludes any attempt at comparing our results and those of El-Azab and Ghoniem to experiment. Therefore, confirmation of the extent of the validity of either one of these MD studies must await further work.

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