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Role of pre-existing point defects on primary damage production and amorphization in silicon carbide (β-SiC)

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ABSTRACT

Molecular dynamics simulations of displacement cascades were conducted to study the effect of point defects on the primary damage production in β-SiC. Although all types of point defects and Frenkel pairs were considered, Si interstitials and Si Frenkel pairs were unstable and hence excluded from the cascade studies. Si (C) vacancies had the maximum influence, enhancing C (Si) antisites and suppressing C interstitial production, when compared to the sample without any defects. The intracascade recombination mechanisms, in the presence of pre-existing defects, is explored by examining the evolution of point defects during the cascade. To ascertain the role of the unstable Si defects on amorphization, simulations involving explicit displacements of Si atoms were conducted. The dose to amorphization with only Si displacements was much lower than what was observed with only C displacements. The release of elastic energy accumulated due to Si defects, is found to be the amorphizing mechanism.

1. Introduction

The cubic polytype of silicon carbide (3C-SiC or β-SiC) is a promising candidate for use as a structural and cladding material for next generation nuclear reactors due to its high strength, hardness, elastic modulus, low neutron capture cross-section [1] and excellent chemical inertness [2]. One of the other requirements a material must satisfy in order to be suitable for nuclear applications is that it must resist radiation damage. Point defects and defect clusters generated from radiation damage can cause Radiation Induced Amorphization (RIA) [3] of SiC leading to inferior mechanical [4] and thermal properties [5].

Modelling of radiation damage in β-SiC has received significant attention at both the atomistic and continuum levels. Molecular dynamics (MD) has been used to determine threshold displacement energies [6], understand primary damage in single crystal[7–9], bi-crystal [10,11] and nano-crystalline SiC [12–14]. Rate theory based formulations model the evolution of defects over longer timescales to study RIA [3,15–17], role of sinks [18] and irradiation hardening [19], for example. Most of the mentioned rate theory models assume that the rate of defect production is independent of pre-existing point defects. While it is known that point defects play an important role in causing RIA in SiC, neither the amorphization mechanism nor the type of defects which contribute to it is clearly known. Electron irradiation studies suggest that mere creation of Frenkel pairs (FP) are sufficient to amorphize SiC [20]. Computational studies on the other hand point out that both FP and antisites may play a role in RIA of SiC [21]. Experimental evidence also points to cascade overlaps induced effects as a possible driving force for amorphization [22]. Thus, cascade overlaps, where regions rich in point defects interact, seem to be an important factor in the amorphization mechanism. The interactions of cascades with other types of defects like stacking faults seem to be beneficial since they increase the threshold displacement energy [23] of C and Si atoms and also lower the barrier for the migration of silicon interstitials [24] potentially enhancing the rate of recovery. More recently, it has been shown that increased chemical disorder (increasing alloying elements or the concentration of the alloying element) resulted in reduced damage accumulations in Ni base alloys [25,26]. Thus both computational and experimental studies suggest that pre-existing faults, defects or alloying elements in materials affects the material's radiation response. In this context, it seems important to understand how the primary defect production is affected by pre-existing damage (point defects) in the sample.

Several simulations, either based on MD [14,21,27–30] (or) *ab initio*-MD [31] have been conducted on samples with pre-existing damage. MD simulations in Ref. [21,27,29], model displacement cascades by repeatedly conducting displacement cascades on the same simulation cell, at specific time intervals. These studies conclude that at low doses individual point defects and small clusters are produced. It is then

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proposed that these defect clusters coalesce to form amorphous domains leading to RIA. Other simulations conducted in Ref. [14,28,30,31] irradiated their samples by repeatedly displacing atoms to create FP within the simulation cell. These simulations study the overall amorphization behavior of the material. Continuum models have also been developed, where the effect of pre-existing damage has been accounted while predicting the evolution of amorphous phase with dose [32,33]. These models are however not based on rate theory and hence do not explicitly model the evolution of point defects. Current rate theory models assume that the production of a certain defect is independent of other defects which exist in the system due to prior bombardments. While existing theoretical models and MD studies do provide insights on possible amorphization mechanisms in β-SiC, it is still not known how a certain defect can affect the production of other defects. Therefore, the present work seeks to quantify how individual point defects affects the primary damage production in β-SiC.

Displacement cascades are conducted on unirradiated (single crystal free of defects) and irradiated samples with specific kinds of point defects, and the results are compared. An important step in successfully conducting cascades on irradiated samples is the creation and maintenance of the required concentration of the specific point defect in the MD simulation cell. Prior *ab initio* [34–36] and MD studies [37] on defect thermodynamics and kinetics in SiC have considered low concentrations of defects in their samples. For example, in *ab initio* calculations, the smallest system size used was 2 × 2 × 2 unit cells of SiC (8 unit cells) with 64 atoms. One interstitial in this simulation cell constitutes a concentration of $\frac{1}{64} \approx 1.6\%$. During radiation, it is possible that concentrations exceed this value and defect rich regions are generated due to cascade overlaps [22,23] leading to amorphization. During the course of the current work, it was found that Si interstitials (Si_i), Si Frenkel pairs(Si_{FP}) and C Frenkel pairs(C_{FP}), in certain concentrations could not be maintained in the simulation cell. These defects either recombined or destabilized the lattice locally producing other defects. One way to understand defect induced instability leading to amorphization is by repeatedly displacing atoms, thus creating FP, and then monitoring the evolution of energy or a disorder parameter with each displacement. In this direction, MD [28,30] or *ab initio*-MD [31] simulations on single crystal and nanocrystalline samples [14] of β-SiC have been conducted to understand amorphization mechanisms. Both, Refs. [14,28] focus on displacing C atoms, arguing that C defects are produced in abundance during the cascade due to its lower threshold displacement energy when compared to Si and is hence more important. Another reason for simulating radiation damage involving only C displacements is that it is also experimentally possible to directly displace C atoms only and amorphize the material, without affecting the Si sublattice [38]. Although fewer in number, Si defects may destabilize the lattice locally and cause amorphization, probably at doses lower than what is seen via C displacements. Explicit Si displacements were considered in Ref. [31], and it was observed there that most Si_{FP} recombined quickly due to low barrier for Si_{FP} recombination. Ref. [30] also displaced both C and Si atoms like what happens in a real material to understand the relationship between swelling, disorder and amorphization in SiC. However, the explicit contribution of Si defects towards the amorphization process was not the focus. Although the mentioned works shed significant insight into several aspects of amorphization in SiC, it is still not clear what role the Si defects play during the amorphization process. Experimentally it is not possible to displace Si atoms only. In reality both Si and C defects coexist. However, the manner in which local Si defects affects lattice stability and what mechanisms might be involved in regions which are rich in Si defects is still not clear. One way to understand the contribution of Si defects towards lattice stability and amorphization is to examine how the dose to amorphization (DTA) with only Si displacements differs from the DTA observed from pure C displacements. Furthermore, chemical disorder is expected during amorphization [39], and the effect of Si defects on the level of disorder induced is also not known.

Thus, in this work two issues pertaining to radiation damage in SiC are addressed. Firstly, the production of point defects in samples with various concentrations of silicon (V_{Si}) and carbon vacancies (V_C), carbon interstitials (C_i), carbon (C_{Si}) and silicon antisites (Si_C), and C_{FP} is discussed. Si_i and Si_{FP} were excluded from cascade studies since they were unstable and could not be maintained in the simulation cell (See Section 2.2.1 for details). Then, amorphization based on the explicit displacement of Si atoms is studied and the results are compared to the that produced from the displacement of only C, and both Si and C atoms.

2. Methods

All MD simulations were conducted using Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [40] software package.

2.1. Interatomic potential

This work employs the hybrid Tersoff/ZBL potential which has been used earlier to model SiC for understanding primary damage in single and nanocrystalline samples [9,10,12], swelling and elastic properties [41,42]. It should be pointed out that a recent work [43] compares two different interatomic potentials with regards to the nature of primary damage in single crystal SiC and it was concluded there that the GW potential [44] is probably better in predicting the cascade dynamics in single crystal SiC. This conclusion was mainly drawn based on the evolution of C_i and V_C during the cascade, where the extent of intracascade recombination of C_i or V_C was much higher for GW than the Tersoff potential. The discrepancy in behavior was attributed primarily to the large recombination barrier for the C_i-V_C pair predicted by the Tersoff potential. The GW potential predicted lower, however, qualitatively a similar barrier to density functional theory (DFT) calculations. The energy landscape for the C_i-V_C barrier was still different for the GW potential when compared to DFT predictions. Furthermore, it is not clear whether conclusions concerning the suitability of a potential, based on defect energetics and kinetics in the dilute regime can be extrapolated to situations when higher concentrations of point defects prevail. Hence, the identification of a perfect potential which can reproduce the cascade dynamics accurately is still an ongoing research question. We therefore believe that the use of Tersoff/ZBL potential is adequate for the goals of the present study to increase qualitative understanding of the processes occurring during radiation damage.

2.2. MD simulation cells

2.2.1. Cascades and the stability of irradiated simulation cell

For cascade simulations, the cell is a rectangular prism with a square base having 30 unit cells of SiC in the x and the y directions and 60 unit cells in z direction. For the unirradiated sample, this simulation cell has 432,000 atoms. Periodic boundary conditions are applied to all six faces to simulate the bulk material. One of the challenges while conducting cascades on samples with point defects is to ensure that the cascade explores regions with the same concentration of point defects everywhere. The direction in which the cascade proceeds and the cascade morphology are both sensitive to the microstate at which the cascade is initiated. It is impossible to ensure that the cascade explores regions with a certain concentration of point defects if they were randomly distributed in the sample. In order to enforce the requirement that the cascade(s) always move through regions with a specific concentration of point defects, the simulation box was constructed using a super-cells approach following the procedure in Ref. [45]. First, a super unit-cell (SUC) consisting of 'N' SiC unit cells is constructed. A certain concentration of a particular point defect is created in the SUC. The SUC, is then repeated in the x, y and z directions to obtain a simulation cell with uniform concentration of the point defect. To this end the