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Atomistic simulations of threshold displacement energies in SiO_2

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Abstract

Silica is one of the candidate materials for final focusing mirrors in inertial fusion reactors. This material will be exposed to high neutron irradiation fluxes during operation. Radiation damage results in point defects that can lead to obscuration of this material; that is, degradation of the optical properties of silica. In this paper, we present molecular dynamic simulations of defect production in silica glass. Results on the threshold displacement energies due to oxygen primary knock-on atoms (PKA) are reported concluding that a range of energies (20–40 eV) exists in which the defects have a probability to be created. In addition, we determine a range of distances for a PKA to become a stable defect from its original position. Our present analysis is focused on the formation of oxygen deficient centers (ODC).

1. Introduction

The development of fusion reactors requires understanding of materials performance under the extreme conditions that will take place in such an environment. Fused silica is one of the materials considered as the final optics in inertial confinement fusion reactors. These lenses will be exposed to high energy neutrons (14 MeV) and ions. The radiation could affect the optical properties of the material, creating point defects that can act as color centers and therefore produce obscuration of these optics [1]. Indeed, experiments have shown that neutron irradiation induces the production of oxygen deficient centers (ODC) [2,3] and their conversion to E′ centers after gamma irradiation [3]. However, significant reduction of the absorption coefficient of these irradiated samples is also observed experimentally through high temperature annealing (>350 °C) pointing to a process of diffusion and recovery of the damage of the sample at these temperatures [3,4].

Unlike metals where the production and annihilation of defects using molecular dynamics simulations has been widely studied [5], there are very few calculations of radiation effects in oxides using these techniques. Several issues could be responsible for this, from the reliability of the interatomic potentials used to the difficultly of identifying defects in compound and amorphous materials such as fused silica. However, the success of such models to interpret the processes occurring during irradiation of metals and semiconductors lead us to follow similar studies in this material. We present calculations of the minimum energy to produce a stable defect of the type 3-fold coordinated silicon, which is related to the oxygen deficient centers that can be observed experimentally. The study is focused on the defects produced by an initial energetic oxygen atom and a fixed temperature, 300 K.

2. Simulation model

Molecular dynamics simulations are used to study the defects produced in fused silica by energetic atoms.

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Fused silica is an amorphous system, formed by silicon atoms tetrahedrally bounded to oxygen atoms. Although there is no long range order in this material, there is a middle range order that can be characterized by features such as the ring size distribution, or the pair correlation function. The interatomic potential used for our calculations is the one developed by Feuston and Garofalini [6]. This potential was fitted to reproduce the structure factor measured experimentally by neutron diffraction. It includes a screened Coulomb potential, a pair potential, Born–Mayer type, and a three-body term, of the same type as the one used by Stillinger and Weber [7] for their interatomic potential for Silicon.

The initial fused silica is generated by a melting and quenching procedure, starting with a β-cristobalite structure of SiO_2. This lattice is melted to a very high temperature (7000 K) through 50 000 steps of 0.5 fs each; then all reminiscence of order is gone. At that stage, the temperature is reduced by scaling the velocities of the atoms in incremental steps of 1000 K, from 7000 to 1000 K, relaxing through 50 000 steps of 0.5 fs each. Finally, the temperature is reduced from 1000 to 300 K and it is relaxed with the procedure described before. At this point we calculate the structure factor, the bond angle distribution, coordination and ring statistics. We have compared this procedure of fused silica formation with results of other work [6,8], and we note that our results are in good agreement with experimental measurements.

Using the configuration of fused silica glass at 300 K, we have studied the minimum energy necessary to create a stable defect in this material. Simulations have been performed using a parallel molecular dynamics code MDCASK [9], with boxes of several sizes, 1536, 12 288 and 1 92 000 atoms depending on the PKA energy. In the following sections, we will present threshold displacement energies to produce defects when the initial energetic atom is an oxygen in the amorphous silica lattice.

3. Results

The energetic recoil is selected as one of the oxygen atoms in the center of the simulation box and given random initial direction. We have studied recoils of energies from 20 to 200 eV in steps of 10 eV; however, the range between 20 and 40 eV has been analyzed in shorter steps of 5 eV.

Due to the amorphous nature of the material identification of point defects is not unique and, therefore, a definition of point defect must be described. For each atom in our lattice, we determine its coordination, considering a cut-off between first and second nearest neighbors distance of 2.35 Å to account for temperature vibrations (1.6 Å is the bond length in silica glass [6,10]) and indicated by the first peak in the calculated pair correlation function. We have focused on the identification of 3-fold coordinated Si atoms. We consider each of these defects as the site for an ODC. We should point out, however, that these under-coordinated silicon atoms have been calculated using the cut-off value quoted above and could be a true oxygen deficient center or a stretched Si–O bond. Therefore, our calculations will always overestimate the values of ODCs obtained experimentally. Besides these 3-fold coordinated Si atoms, other types of defects are also identified, specially at the highest energies studied. However, we have focused only in the production of 3-fold coordinated Si atoms in this study.

We have also calculated how far the atoms have been displaced from their initial position. In the case of the oxygen PKA, calculating the distance to the silicon atoms that it finds in its path together with the calculation of the coordination of the silicon atoms can be used to understand the defects produced during all the collision events.

Using a simulation box of 12,288 atoms ($8a_0 \times 8a_0 \times 8a_0$), we have simulated PKAs with energies of 20, 25, 30, 35, 40, 50, 70, 100, 110, 120, and 200 eV. Various initial PKA directions have been simulated changing their azimuthal angles in steps of 15°. Fig. 1(a) represents the number of ODCs versus time for two different initial energies of the PKA, 25 and 120 eV. Simulations were carried out up to 1.2 ps. As can be seen

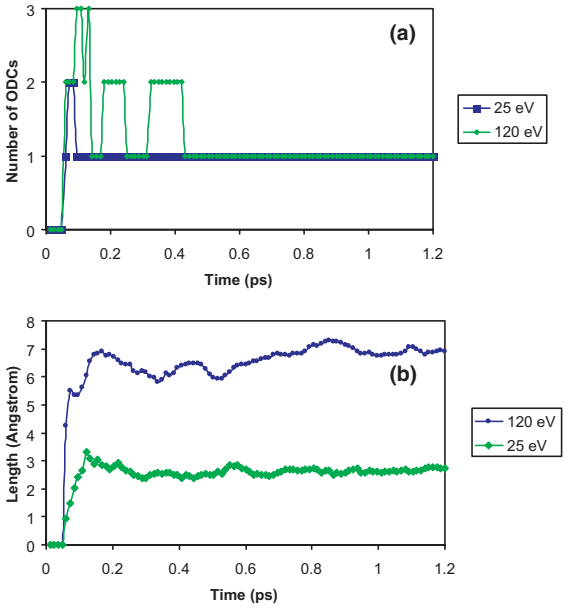

Fig. 1. (a) Number of oxygen deficient centers versus time (ps) for PKAs of 25 and 120 eV and (b) displacement (Å) of oxygen PKA from its original position versus time (ps).

in Fig. 1, no variations of the number of defects are observed after times on the order of 0.5 ps at this temperature (300 K). Fig. 1(b) gives us another view of the same effect, now represented by the distance traveled by the PKA from its original to its final position for two different energies; the time scale is the same as that in Fig. 1(a).

Unlike in the case of a crystalline structure, in an amorphous system the result of a simulation like the one presented above will depend on the location of the energetic atom chosen, since not all positions are equivalent. However, on average every direction should be equivalent. It is therefore necessary to repeat the simulation for different initial oxygen atoms and recoil directions and obtain statistical average of the efficiency for defect production. Using the results of two different oxygen locations and an average over 24 different initial directions, we obtain the probability of producing a defect as a function of the energy of the PKA. This probability is shown in Fig. 2. Clearly, and as expected, the probability of creating a stable defect increases with the initial energy of the recoil. For energies higher than 50 eV every PKA will produce a stable defect, while there is a 50% chance of creating a stable defect for energies on the order of 30 eV.

We also have observed that there is a minimum distance for the PKA to attain its new stable position. The minimum distance that a PKA needs to be displaced from its initial position to produce a unique stable ODC is approximately 3 Å. Moreover, we also have found another characteristic distance to generate two ODCs, which is approximately 4.2 Å. Finally, we define a third distance where three ODCs have a probability to be created, ≈12 Å, as can be seen in Fig. 3.

In Fig. 4, we represent the maximum distances that the PKA is displaced from its initial position for different energies, and different azimuthal angles and for one of the oxygen PKA considered. For this particular case, azimuthal angles of 90° and 210°, for all energies, the PKA has a very small displacement, leading to no formation of an ODC considering that the displacement is smaller than 3 Å. This effect is explained because along those directions there is neighboring silicon atoms.

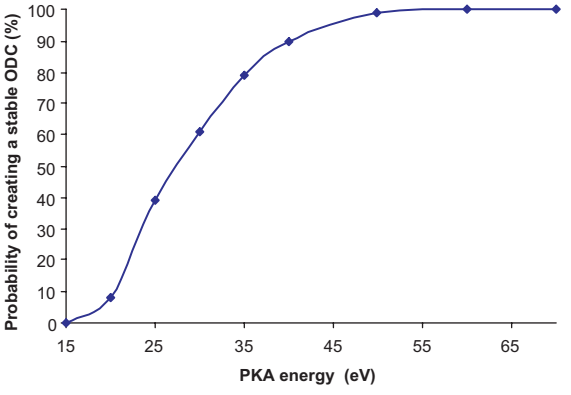

Fig. 2. Probability (%) of creating stable ODC versus oxygen PKA energy (eV).

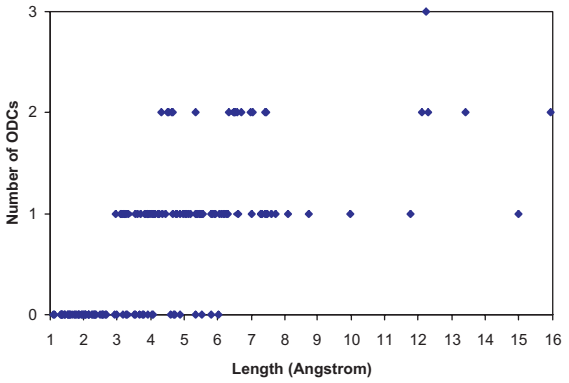

Fig. 3. Number of ODCs versus the maximum distance (Å) the oxygen PKA is displaced from its initial position, for all the simulated angles and energies.

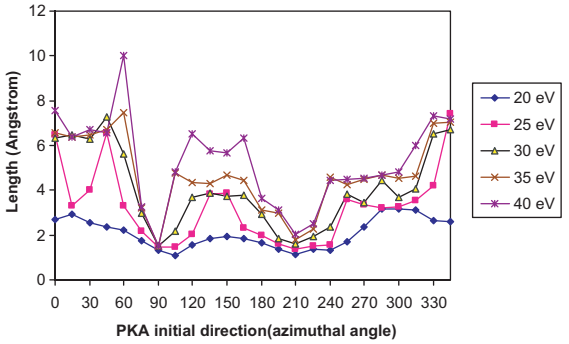

Fig. 4. Maximum distance that oxygen PKA is displaced from its initial position versus its initial direction of movement.

4. Conclusion

We have calculated the threshold displacement energy for the formation of 3-fold coordinated silicon atoms in fused silica when the initial recoil atom is an oxygen. Defects such as 3-fold coordinated silicon atoms are related to oxygen deficient centers (ODC) that can be identified experimentally [3]. The results from our calculations show that there is a range of probabilities to create such defects, with a 50% chance for energies as low as 30 eV. Only for energies higher than 50 eV every recoil will produce a stable defect. Previous estimates based on calculations of displacement cascades at energies of 1 keV and higher resulted in a higher value for

the threshold displacement energy, closer to 100 eV [11]. We should point out, however, that in this case the energy will be deposited not only in oxygen atoms, as in the case of the calculations reported here, but also in silicon atoms. Further studies will include the calculation of threshold displacement energies where the energetic recoil is a silicon atom instead of an oxygen one and will be compared to cascade calculations. Our simulations have also revealed a correlation between the maximum displacement of the initial oxygen PKA and the production of stable defects.

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