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Molecular dynamics study of defect in amorphous silica; generation and migration

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Abstract: Fused silica is a material of interest due to its increasing number of applications in many different technology fields. In thermonuclear fusion it is one of candidate materials for both final focusing optics of lasers for NIF and future inertial fusion reactors and diagnostics of the Safety and Control Systems of the ITER machine as well as DEMO magnetic fusion reactors. In operation, these materials will be exposed to high neutron irradiation fluxes and it can result in point defect. Materials properties of interest (optical absorption, radioluminescence, mechanical properties,…) are closely related to the presence of defects. These defects can be generated directly by irradiation or by the presence of impurities in the material.

We present molecular dynamic simulation of displacement cascade due to energetic recoils in amorphous silica without hydrogen atoms and with 1\% of hydrogen atoms trying to identify defects formation and we present molecular dynamics simulations to study the effects of hydrogen atoms in this material and their interaction with defects; The diffusion coefficients and mechanisms of H mobility in fused silica has been calculated and compared with those existing in the literature. The interatomic potential developed by Feuston and Garofallini has been used in both studies.

1. Introduction

Vitreous silica is a material of great importance, both from the point of view of the fundamental physics, due to its use as model structure, and from the point of view of its great variety of its technological applications. The electrical, dielectric and optical properties, characteristics of vitreous silica, together its chemical inertness and thermal stability, are the main factors that determine the great versatility of applications of this material. In addition, Fused silica is a candidate material for optical and radio-frequency diagnostic systems in magnetic confinement fusion reactors and as final optics in inertial confinement fusion reactors [1,2,3,4,5]. In both cases this material will be exposed to high energy neutron (14 MeV) and gamma irradiation. Radiation induces optical absorption, creating point defects that can act as colour centers [6]. Its defects, that determine in last instance the properties of the material, may be detected, among other methods, by studying the optical absorption bands or its fotoluminescence. At the present time there is abundant information on the different defects and their optical properties [7].

In recent years the development fused quartz with a high radiation resistance (from the optic point of view), at least to ionising radiation has been possible [8]. However the cause of this resistance is not
sufficiently clear. One of the possible parameters that justify this phenomenon is the presence of impurities in the material [9].

The molecular dynamics (MD) is a computer simulation technique that allows one to obtain information on the atomic structure of the system and on its physical properties. Molecular dynamics simulations have proved to be useful in the description of defects produced by irradiation at keV energies in metals and semiconductors [10, 11], however the number of studies in oxides and in particular, in amorphous systems, is more reduced. The objective of this work is the study through molecular dynamics of the defects that are formed in fused silica produced in irradiation conditions and their diffusion.

2. Simulation conditions

The interatomic potential developed by Feuston and Garofalini [12] specifically for fused silica has been used in these calculations. The same authors also developed a version of this potential for the H-Si-O system that has been used for the calculations of radiation effects in the presence of H [13]. This potential is in good agreement with neutron and X ray scattering experimental data. In addition, this potential can reproduce a great number of atoms since it has 5.5 Å cutoff radius. This potential has been fitted for short range interactions to the Universal potential.

The MD simulation computer code used for the MD simulations presented in this work was constructed to study specifically the radiation effects in materials. This program is based on the MOLDY code developed by Finnis et al, and was later modified to run in parallel machines [14]. The initial amorphous structure is generated by melting a beta-cristobalite lattice of SiO$_2$ and subsequently quenching down to 300K. For the calculations presented here two systems, the first with a simulation box with a total of 192000 atoms, that is approximately 15x15x15 nm$^3$, and second simulation box with a 1% of Hydrogen atom in order to study the influence of these atoms in the displacement cascades. This simulation box has been prepared in a similar form. And, we have compared the defects produced by irradiation that appear in both simulation box.

In addition, we have calculated the mean square displacement (MSD) to different temperature to study the diffusion of the H atoms in the bulk. With this information we computed the diffusion coefficient and activation energy with Einstein relations.

3. Result and conclusion

Identification of defects in an amorphous system is quite complex and unlike in perfect crystals is not unique, therefore, a definition of point defect must be described. For each atom in our lattice, we determine its coordination, considering a cutoff between first and second nearest neighbours distance of 2.15 Å (this distance is the minimum between first a second nearest neighbours distance in the pair correlation function). But when the nearest neighbour is a hydrogen atom we have used the distance of 1.30 Å since the bond length is shorter in this case.

Defects identified in silica are: 1.- ODC (oxygen deficient centers), silicon with 3 oxygen as nearest neighbors that is supposed as affecting as E’ center because our defects relax of similar way (in a plane [15]) as it was describe in a experimental model [16,7]. 2.- Si5: Silicon with coordination 5, silicon with 5 oxygen as nearest neighbors. This defect is difficult to compare with experiment because it does not exist in the literature but a preliminary conclusion of this type of defect is that it corresponds with environment with oxygen excess, and in these zones different kinds of defects are formed as Peroxy Radical (POR) and Peroxy Bridge. [7]. 3.- O1: Oxygen with coordination 1, one silicon as nearest neighbor, similar to Non-Bridging Oxygen (NBOs) defect [7]. 4.- O3: Oxygen with coordination 3, oxygen with 3 silicon as nearest neighbors. This defect is also difficult to compare with experiment but just as in the previous case of Si5, the O3 corresponds with a environment with oxygen deficiency; that is, it is possible that our potential [12,13] transform the structures as Si3 in other structures more probably as the O3. It is clear that this structure is a Si3, that in its relaxation, the
center silicon has been linked with other oxygen that it was already linked to other two silicon. 5. - **Si4**: Silicon with coordination 4 (perfect coordination). And 6. - **O2**: Oxygen with coordination 2 (perfect coordination). In the reference [7] a description of the atomic configurations of these defects can be found. The potential energy range, of each kind of different coordination number, is easy to obtain [arti culo [17]. Figure 1 shows the results of the average of number of defects generated (ODC, Si5, O1, O3) versus Prin may Knock-on Atoms (PKA) energy, followed for a total time of 4.5 ps, using a time step of 0.1 fs. Before the number of defects is computed, we use a tool to remove the thermal fluctuations so that we can determine the stable defects. This process verifies if the atom is going towards equilibrium or away from it. If the atom moves away from equilibrium its velocity is set to zero. It is clearly observed in Figure 2 that the probability of creating stable defects, in general increases with the initial energy of the recoil atom.

![Figure 1: Average of number of generated defects (Si3, Si5, O1, and O3) versus PKA energy, that is, The final defects (after displacement cascades) menus initial defect (before displacement cascade).](image)

To analyze the defects in presence of H atoms we generated the simulation box with a 1 % of H atoms. These H atoms were implanted in the materials of random form, whenever they do not coincide with other atoms of the lattice. Next, the annealing process leads to H atoms to its more stable positions in the bulk of the simulation box. The defects in presence of H atoms are basically of two types, H atoms binding with an oxygen atom with coordination 1 [18, 19], that is, the H atom is connected to O1 defect and the scheme representation is H-O-Si. The other possible form is H atoms in free state. In Figure 2, it is possible to see the production of defects as a function of the PKA energy. But, in order to better follow of these defects we have identified these two types of defect for four structures. The H-O-Si=, and O-H are two different form to represent the H atom binding with a oxygen atom, but, in first with compute the coordination of the oxygen atom and in the second we compute the coordination of the H atom. And the H free and H-O=2Si correspond with the H atoms free but in the second form the H atoms is nearest from the oxygen with coordination two (with two Si).
In addition, we have analyzed the mobility of these impurities (H atoms). In order to make it, we have computed an average of the MSD of the 100 H atoms. This average improves the result of the behaviour of the atom. In the figure 3 we represent the behavior of the MSD to different temperature. It is easy to see that the mobility of the H atoms grow with the temperature. We have observed two slopes in diffusion coefficient versus 1/kT that it corresponds to two different activation energies [20]. These activation energies are $E_1 = 0.45$ eV and $E_2 = 2.54$ eV. $E_1$ corresponds with the energy necessary to move when the H atom is free [21], and the $E_2$ correspond with the energy necessary to break the bond H-O.
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