Radiation Damage Modeling of Fused Silica in Fusion Systems

F. Mota 1), M.J. Caturla 2), J.M. Perlado 1), A. Ibarra 3), M. León 3), J.Mollá 3)

1) Instituto de Fusion Nuclear (DENIM) / ETSII / Universidad Politecnica Madrid, Spain
2) Departamento Fisica Aplicada, Universidad de Alicante, Spain
3) Departamento Materiales para Fusion, CIEMAT, Spain
mota@denim.upm.es

Abstract

Amorphous Silica 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 and vary the optical absorption, that is, degradation of the optical properties. In this paper 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. We have made a statistics of the different kind of defects at different energy of primary knock-on atoms (PKA). The range of studied PKA energies are from 400 eV to 3.5 keV and it is made to both component of this material Silicon and Oxygen.

1. Introduction

Fused silica has a wide range of applications. The interest on the effects of radiation in vitreous silica has increased recently due to its possible application as optical transmission component in fusion reactors. In this case, this material will be exposed to high neutron irradiation during operation [1,2]. As a result point defects will be created that can lead to obscuration of the material; that is, degradation of the optical properties of silica. Among the defects observed under neutron irradiation are the Oxygen Deficient Centre (ODC), which is related to a silicon atom with coordination three instead of four, and the Non-Bridging Oxygen Hole Centre (NBOHC), which is related to an oxygen with coordination one [3].

In this work we study the production of point defects due to atomic displacements, such as those produced by neutron irradiation. We centre this study in characterizing the types of defects produced during irradiation based on the coordination and potential energy of all the atoms in the simulation box after each collision event. In terms of the applications of interest, namely fusion reactor materials, the most important defect that has been identified experimentally is a colour center in the 248 nm wavelength , which appears under neutron irradiation and makes the material opaque [1, 2], the so-called oxygen deficient center (ODC). Under gamma irradiation this defect is converted in E' centers with a 214 nm wavelength according to some studies[3]. However, other types of defects are also produced during irradiation, some of which have not been identified yet. Our previous calculations have focused primarily on ODCs [4, 5]. In this study we also identify other possible defects produced during irradiation and compare to experimentally observed ones.

2. Simulation modelling

We use molecular dynamics simulations to study the defects produced in fused silica. Fused silica is an amorphous system, formed by silicon atoms tetrahedrally bonded to oxygen

atoms. The interatomic potential used for our calculations is the one developed by Feuston and Garofalini [6], potential that was fitted to reproduce the structure factor of this amorphous system as obtained experimentally through x-ray diffraction and neutron scattering data. In order to generate the amorphous system of the fused silica, we start with a crystal lattice of beta-cristobalite for SiO_2 because its structure is very similar in its short range order (Tetrahedrons structure SiO_4^{-4}) and this structure is melted (at 7000 K) and quenched through a series of steps until a final temperature of 300 K is achieved. Simulations have been performed using a parallel molecular dynamics code MDCASK [7]. Indicators of the amorphous structure such as the structure factor, the bond angle distribution, coordination or ring statistics are computed in order to check the final structure of the computational box **[6,8].** The simulation box size used had 192000 atoms, that is approximately 15x15x15 nm^3.

In addition, we have used one 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.

Once the initial amorphous structure is constructed and carried to room temperature (300K), we identified those defects produced by energetic atoms. Periodic boundary conditions are used with a bath control to keep the final temperature at 300 K through scaling the velocities of those atoms close to the border of the simulation box. The energetic silicon atoms is chosen in the center of the simulation box for low PKA energy and in the top of the box when the PKA energy is higher.

3. Results

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 1.30 Å because the binding is shorter.

In addition, we are cataloguing the different kind of defects by its potential energy. Each defect has a characteristic energy. We have determined the potential energy of each atom with different coordination. For obtaining it, we have started with the total spectrum of all atoms and we diagnosis the defects that reproduce the different energy level of spectrum, representing the energy of the atoms with different coordination and looking the site in the energy spectrum they occupy. We perform MD simulations with cell boxes size of 192000 atoms and temperatures of 300K without suffer displacement cascades.

Defects identified in silica are (Figure 1): 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 [9]) as it was describe in a experimental model [10,11]. This kind of defects has a range of potential energy between [-25.05 eV: -24.55 eV] and before the cascade collision while after 6 fs for one cascade of 1.5 keV the range of energy grow significantly [-26.65eV:-16.85 eV] . 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. [11]. The range of potential energy is [-30.25 eV, -27.95 eV] before irradiation and after irradiation [-30.45 eV,-24.90eV]: .- **O1**: Oxygen with coordination 1, one silicon as nearest neighbor, similar to Non-Bridging Oxygen (NBOs) defect [11]. Its range of potential energy is [-7.85 eV:-4.2 eV] before irradiation and after irradiation [-14.10 eV:-9.30 eV]. 4.- **O3**: Oxygen with coordination 3, oxygen with 3 silicon as

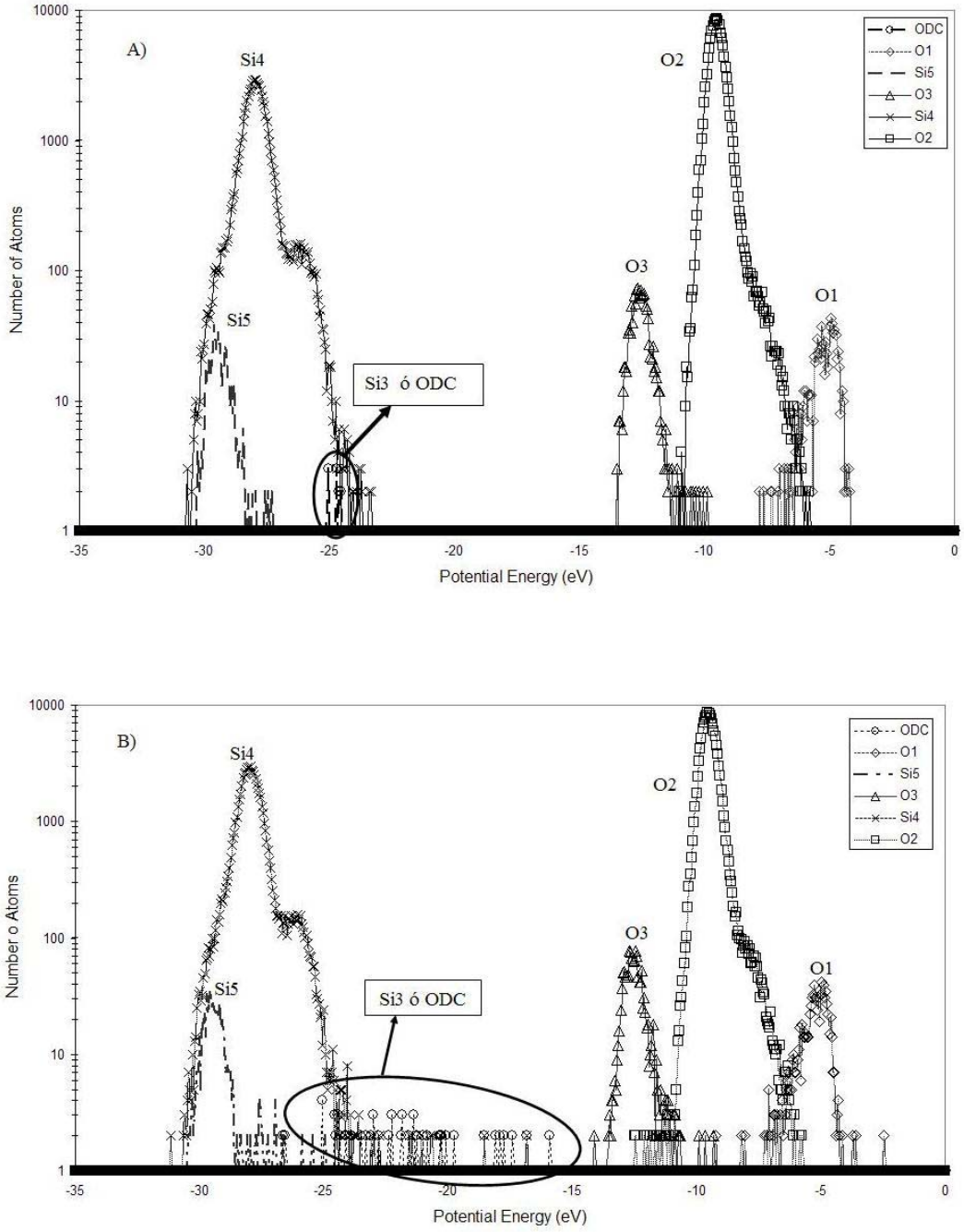

Figure1: Potential energy spectrum of atoms with different coordination. Figure 1a) is before the displacement cascade and Figure 1b) is during the displacement cascade 6 fs after the initial collision

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 [6] 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. Its range of potential energy is [-13.5 eV:-11,15 eV] and after irradiation [-14.10 eV:-9.30 eV]. 5.- **Si4:** Silicon with coordination 4 (perfect coordination) In the case a) the range of potential energy is [-30,7eV:-23,35eV] and in the case a) [-31,15eV:-22,75eV] and 6.- **O2**: Oxygen with coordination 2 (perfect coordination). In the case a) the range of potential energy is [-11eV:-5,8eV] and in the case b) [-12,5eV:-5,75eV]. In the reference [11] a description of the atomic configurations of these defects can be found.

We have analysed range of energies studying in detail the spectrum of PKA energy between 40 to 3500 eV. In this range of energy, we have made several cascades for each one, since it is necessary to conclude from an average. The production of a defect in an amorphous material will depend on the location of the energetic atom chosen, since not all positions are equivalent. Therefore the calculations have been repeated for a large number of cases, we have made from 24 cases to low energies, since the box is smaller and it is less expensive computationally, until 5 cases to the higher energies, changing both the incidence direction and initial atom.

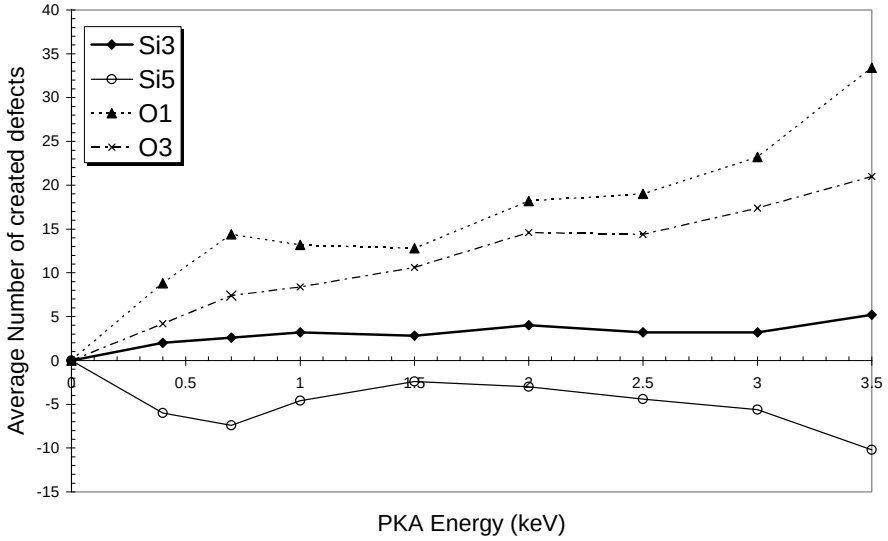

Figure 2: Average of number of generated defects (Si3, Si5, O1, and O3) versus PKA energy.

The Figure 2 shows the results of the average of number of defects generate (ODC, Si5, O1, O3) versus PKA energy followed for a total time of 4.5 ps to higher energies, using time step of 0.1 fs. After of simulation we use a tool to remove the thermal fluctuation and to can determinate which of the defects are stables. This process verify if the atom go forward to the equilibrium state of move away. If the atom move away of the equilibrium fix its velocity as zero but if the atoms go forward to the equilibrium do not change its velocity. This process accommodate the entire atoms in the site more probable of minimum energy to ensure that the defects are not as consequence of thermal vibration. It is clearly observed the probability of creating stable defects, in general, increases with the initial energy of the recoil atoms.

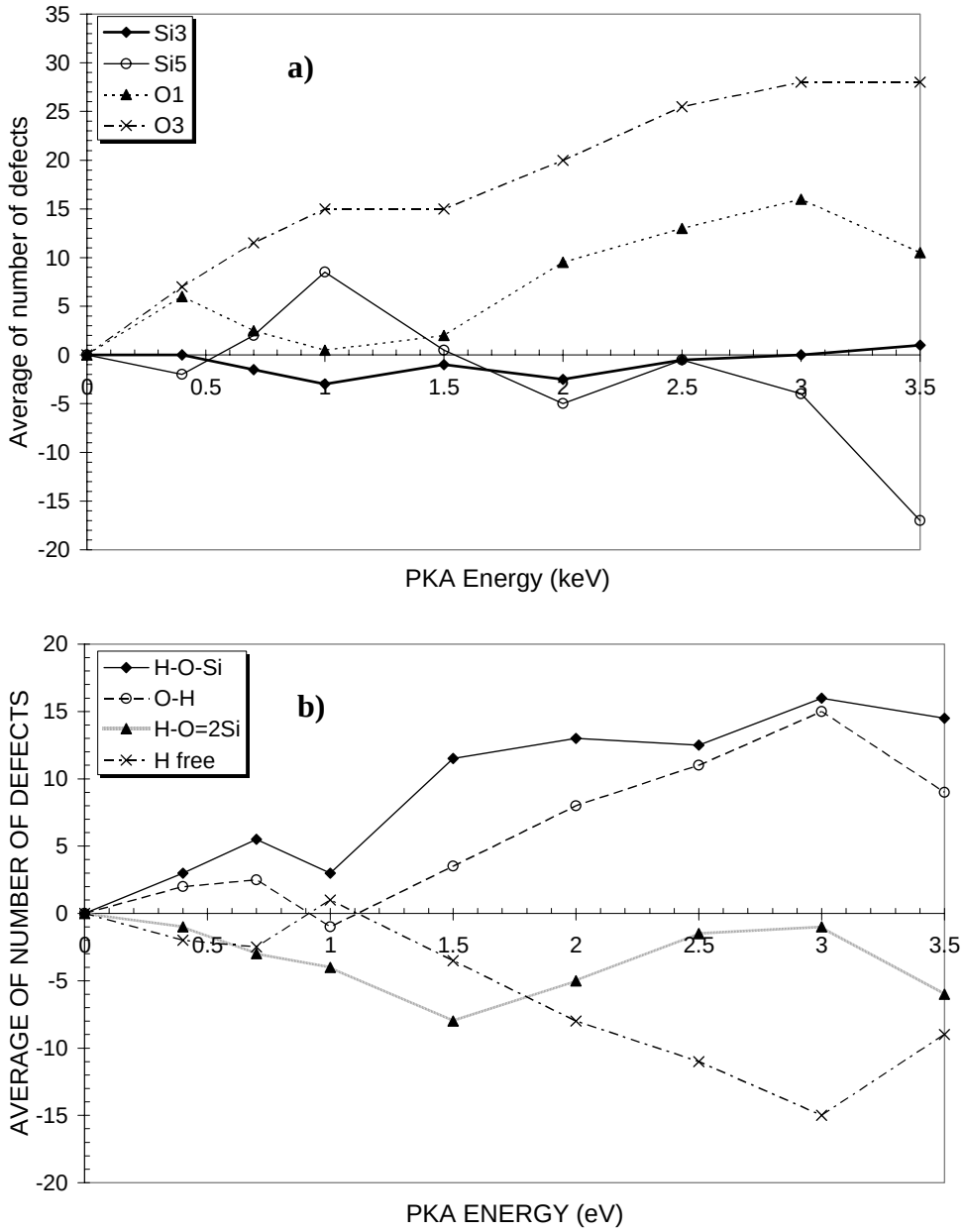

Figure 3: Average number of defects produced by the energetic atoms as a function of recoil energy with 1% H in the sample. (a) Si5, Si3, O3 and O1 and (b) H related defects.

Hydrogen isotopes will be deposited also on the surface of the fused silica components coming from the reaction chamber. On the other hand, some experimental results show that radiation damage can be different depending on hydrogen content, indicating that a detailed knowledge of the hydrogen role in fused silica should be fully understood. We present molecular dynamics simulations to study the effects of hydrogen atom in this material, their interaction with defects and the interaction with the displacement cascade. The interatomic potential developed by Feuston and Garofallini [6] and modified by Webb to introduce the hydrogen interaction [12] will be used in these studies.

In the Figure 3 is possible to see the production of defects in function of the PKA energy when in the simulation box there are 1 % of hydrogen atoms. It is clear that three different kinds of new defects appear. The H-O-Si≡, H free, H-O=2Si and O-H. The O-H is the sum of the defects H-O-Si≡ and the defects H-O=2Si, that is, it is the hydrogen atom with coordination 1 with oxygen atom. Then, the nearest neighbours oxygen could have coordination 2 H-O-Si≡ or coordination 3 H-O=2Si. Therefore, the defects H-O-Si≡ are the oxygen atoms with coordination 1 with a silicon atom and coordination 1 with a hydrogen atom. And the H-O=2Si the oxygen atom linked with 2 silicon atoms and 1 hydrogen atom. Finally the last defect is which we call H free. These defects are the H with coordination zero; that is, they have not nearest neighbour atoms. So we have identified the hydrogen defects by two different ways, calculating the coordination of hydrogen atom and calculating the coordination of oxygen atom.

4. Conclusion

The main type of defects produced in fused silica under irradiation due to nuclear collisions has been obtained with molecular dynamics simulations. The main defects observed are oxygens with coordination one, which could be related to the NBOHC. Silicon atoms with coordination three are also observed, related to ODC. In addition, it is possible to identify the defects Si5 and O3 with zones with high density of oxygen (in which there are defects as POL and POR) and zones with low density of oxygen respectively [11].

It is possible to observe that, when increasing the PKA energy, the number of almost all defects generated by the displacement cascade increase, except the Si5. It seems that this structure is not stable for our potential [6], since it doesn't describe the bond O-O, because this interaction is repulsive.

When H is included in the lattice new defects appear, in particular O-H type of defects. The number of these defects increases with recoil energy, while the number of O1 defects is reduced with respect to those measured without H. This seems to indicate that as the recoil generates defects in the sample, free H migrates towards these newly generated defects creating O-H. However, with the potential used [12], the position more stable to the H atoms is when it is bonded with a oxygen atoms, forming HO- structures. Further calculations are being performed where most of the H is initially located at O-H sites. In addition, in both cases, the number of common defects increases, but it is observed in a different form. The O3 increases, when we have H, faster than in the simulation box without H atoms. The number of O1, when H atoms are in simulation box grows slower, because this kind of defects is shared with the new type of hydrogen defects H-O-Si≡ mainly.

5. Acknowledgments

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6. References

[1] E. R. Hogdson, "Challenges for insulating materials in fusion applications", Nucl. Instrum. & Methods B 191 (2002) 744
[2] Jeffery F. Latkowski,* Alison Kubota, Maria J. Caturla, Sham N. Dixit, Joel A. Speth, And Stephen A. Payne, "Fused Silica Final Optics For Inertial Fusion Energy: Radiation Studies and System-Level Analysis, Fusion Science and Technology", V43 N4 (2003) 540-558
[3] C. D. Marshall, J. A. Speth, S. A. Payne, "Induced optical absortion in gamma, neutron and ultaviolet irradiated fused quartz and silica", J. Non-Crist. Solids **212**, 59 (1997)
[4] F.Mota, M.-J.Caturla, J.M.Perlado, E. Dominguez, A. Kubota, "Atomistic simulations of threshold displacement energies in SiO_2", J. Nucl. Mater. 329-333 (2004) 1190-1193
[5] F. Mota, M.J.Caturla, J.M.Perlado, E.Dominguez, A. Kubota, Fusion Engineering and Design, 75-79 (2005) 1027-1030
[6] B. P. Feuston and S. H. Garofalini " Empirical three-body potential for vitreous silica" 89[9] 5818-5824 (1988)
[7] MDCASK http://www.llnl.gov/asci/purple/benchmarks/limited/mdcask
[8] A.Kubota,M.J.Caturla,J.S.Stolken,M.D.Feit. OPTICs EXPRESS, 8(2001)
[9] J. Mollá [1], F. Mota [2], M. León [1], A. Ibarra [1], M.J. Caturla[3], J.M. Perlado. "The role of the fused silica stoichiometry on the intrinsic defects concentration" J. Nuclear Materials. In Press (2006)
[10] F.J. Feigl, W.B.Fowler, and K.L.Yip Oxigen vacancy model for E_1' center in crystalline SiO_2, Solids State Comm. 14 (1974) 225-229
[11] G. Pacchioni, L.Skuja, D.L.Griscom, "Defects in SiO_2. and related dielectrics: science and technology". NATO science series (2000).
[12] E. B. Webb and S. H. Garofalini "Relaxation of silica glass surfaces before and after stress modification in a wet and dry atmosphere: molecular dynamics simulations" J. of Non-Cryst. Solids 226 (1998) 47-57