Calculation of Threshold Displacement Energy of Xe-inserted UO_2 Using Atomistic Simulation

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1. Introduction

Uranium Dioxide (UO_2) has been utilized as a major fuel material for Light Water Reactors (LWR). By undergoing nuclear fission reactions, fission products accumulate inside the UO_2 structure. The accumulation of various fission product can cause radiation-induced degradation of the UO_2 fuel structure.

Known as the richest fission gas element, Xenon (Xe) plays the most important role affecting the fuel swelling, which is significantly related to Pellet-Cladding Mechanical Interaction (PCMI) [1]. High burnup fuels contain a relatively large amount of Xe atoms. Xe gas atoms can affect the UO_2 microstructure by forming bubbles on the intragranular bulk area or segregating on the grain boundaries. [2] Moreover, the change of atoms consisting of the lattice would affect the radiation resistance, including the swelling behavior.

Threshold displacement energy (TDE, E_d) is one of the most essential properties to provide the information about the radiation resistance of certain material, as well as the defect formation energy or migration energy of self-interstitial atom (SIA). The basic definition of TDE is the minimum Primary Knock-on Atom (PKA) energy given for PKA to get off its original position in the lattice and form defect. [3][4][5][6] Since this definition can be interpreted in several statistical ways, some research groups have specified the definition of TDE.[4][7]

There was a study by Bauer and Sosin which measured TDE via experimental TEM methods.[8] However, the study pointed out that the uncontrollable parameters such as lattice imperfections, sample thickness, and experimental sensitivity lead to inefficiency and high deviation of the measurement results.

As an alternative method, computational techniques such as Molecular Dynamics (MD) simulations are commonly being used to calculate the TDE of various materials not only in single-element metals [4] but also in multi-element lattices [5], including high entropy alloys. [9]

The TDE of single crystal UO_2 fluorite lattice is also calculated by MD techniques in a few studies. [7][10] However, these studies did not considered fission gases such as Xenon or Krypton in spent UO_2 fuel.

In this study, a MD repetitive simulation is applied in order to calculate and compare the TDE of Uranium atom in the perfect UO_2 fluorite lattice and Xe-containing UO_2 lattice, respectively.

2. Methods

MD simulations on this work were performed via Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) developed by Sandia National Laboratory. Details and methodologies are described in this section.

2.1 Interatomic Potential

LAMMPS stores every atom's coordination data of each timestep. In order to calculate the coordination of atoms at the next timestep, an interatomic potential function made by Cooper et al. is applied. [11] Being one of the most accessible potential formalism successfully representing the interatomic force between UO_2 ceramic fuel and fission gases (Xe and Kr), Cooper et al's potential energy E_i of each atoms in the simulation box can be expressed in two terms: first is the pair potential term and second is the Embedded Atom Method (EAM) term.

2.2 Simulation Detail

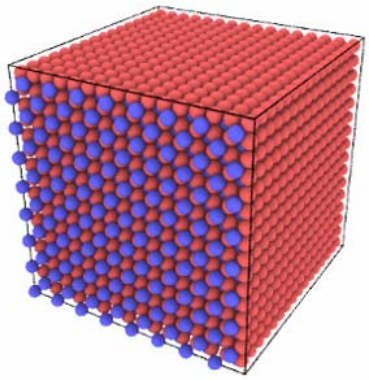

Fig. 1. Initially built $8 \times 8 \times 8$ UO_2 fluorite supercell. Red – Oxygen / Blue – Uranium

The initial cell of 4 Uranium atoms and 8 Oxygen atoms was produced with lattice constant of 5.468Å and underwent a total energy minimization process. Then the initial single cell is replicated 8, 8, 8 times respectively in 3 dimensions to form a supercell of total 6144 atoms. After forming the basic supercell, the system is equilibrated in the Nose-Hoover style isothermal-isobaric ensemble (NPT). The equilibration temperature condition was 600K and 1200K, which is known for coolant temperature and calculated fuel centerline temperature of high-burnup UO_2 fuel respectively. [12] In this stoichiometric UO_2 supercell, a Uranium atom

locating around the center of the supercell is selected as PKA.

For the Xe-containing UO_2 lattice, central 3 atoms including one Uranium atom and two Oxygen atoms (which are positioned linearly) are replaced with 3 neutral Xenon atoms. Then, three Uranium atoms around the linear tri-Xe are selected as PKA, which is visualized in Fig. 2. and the influence of the relative position of each PKA Uranium is considered.

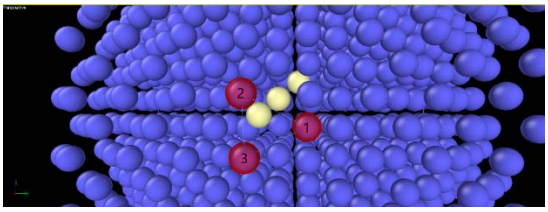

Fig. 2. Selected PKA Uranium for Xe-added cell
Yellow – Xenon / Blue – Uranium / Red – PKA U
Oxygen atoms are excluded in this visualization

For TDE calculation simulation, 10, 15, 20, … 70 eV of PKA Energy is given to the selected PKA, respectively, in form of kinetic energy. The selection of PKA direction is described in the next section, 2.3.

It is sufficiently discovered that even with the same PKA energy, the number of stable Frenkel pairs depends on the direction of PKA.[3][4][7] Along with this idea, Chen et al.[4] classified the definition of TDE in several classes. i) Direction-specific TDE considers whether a stable defect has formed or not in 'certain direction chosen by cubic subdivision sampling'. ii) Angle-averaged TDE is acquired naturally by calculating the average of the all direction's direction-specific TDE. iii) Production probability TDE is determined as the energy which makes the probability of stable defect formation one-half, regarding all repeated cases in every direction's trial. iv) Defect count TDE takes the energy value when the average number of formed stable Frenkel defects in every repeats meets 0.5. Another reference written by Dacus et al.[7] also classifies the TDE definitions, which specified the 'atom displaced its original position in the lattice' to 'stable Frenkel defect formation'. Hence, this work has evaluated whether the TDE condition is achieved or not by the condition of stable Frenkel defect formation and its average success rate, P_{form}.

In this work, all the TDE values in the results are considered by the definition of production-probability TDE.

2.3 Selection of PKA Direction Vectors

Regarding the directions of PKA irradiation events in the crystalline lattice structure, ideally the neutron collisions would have angularly isotropic distribution on the unit sphere surface. Robinson et al.[3] states that the crystal-symmetry based direction sampling method would be extremely harder as the crystal structure gains its complexity, and uniform distribution on the unit sphere will cost very high computing power.

Thompson's Problem, which is introduced as an alternative pseudo-isotropic selection method for direction vectors, is originally a simple mathematical problem to locate N point charges on the unit sphere so that it can get the minimum electric potential energy.

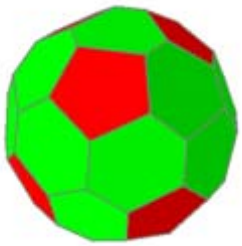

Fig. 3. N=40 Solution vectors of Thompson's Problem [I-1]

Thompson problem solution vectors for N=40 was used for the selection of the Direction vector. For each direction, 20 times of repeats are tried by varying the random seed of the velocity generation algorithm in LAMMPS.

2.4 Evaluation of Stable Frenkel Defect Formation

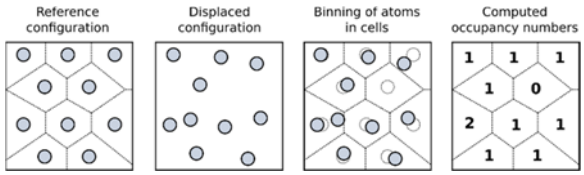

Fig. 4. Voronoi Tessellation [I-2]

The number of formed Frenkel defects is calculated from Voronoi tessellation computation (a.k.a. Wigner-Seitz Analysis) using LAMMPS. In Voronoi tessellation, whole volume of the simulation box is divided into several cells according to the coordination of atoms at the initial timestep. At certain timestep, the coordination of the atoms are analyzed based on which Voronoi cell they are currently in.

5picoseconds consisting of 5000 timesteps (1fs for each timestep) is executed for every PKA simulations.

3. Results and Discussion

3.1 Stoichiometric UO_2 supercell

Table 1: TDE of Uranium and Oxygen in UO_2, Reference and this work

Source	Evaluation Method	TDE (eV)
Dacus et al.	MD with 4 different interatomic potentials	U : 60~65 O : 125~200
Meis et al.	Sudden Approximation Calculation	U : 50 O : 20

Martin et al.	MD with cascade simulation + NRT Law	U : 40~50 O : 20
This work	MD	U : 60~70

Several studies have evaluated the value of TDE of UO_2 in various ways, which is listed in Table 1 [7][10][13].

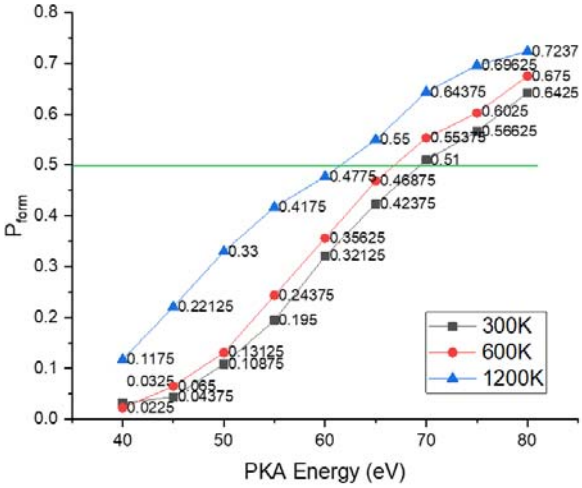

Fig. 5. PKA Energy - P_{form} graph of Uranium PKA in stoichiometric UO_2 supercell

For the perfect UO_2 supercell in this study, P_{form} has reached to 50% in range of 60~70 eV. P_{form} showed its positive relationship with temperature, due to the increase of thermal vibration of the particles.

The TDE value acquired from this work and P_{form} curve matched with the result from Dacus et al.[7] which concluded the Uranium PKA energy in UO_2 as 60~65 eV, since the specified definition for TDE and methodology was almost identical. However, the TDE value by Martin et al.[13] was different because they applied the NRT law to calculate the TDE value of Uranium and Oxygen assuming the proportional characteristic of PKA energy and the number of point defects.

3.2 Xe-added UO_2 supercell

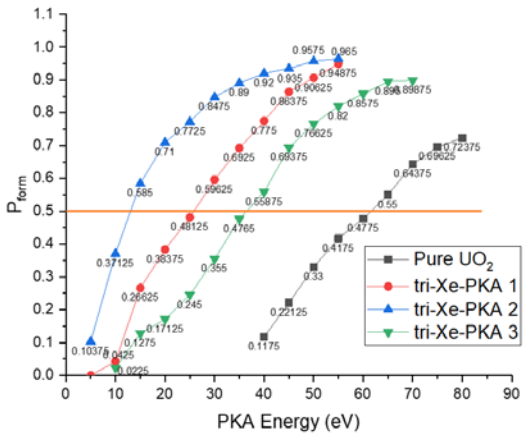

Fig. 6. PKA Energy - P_{form} graph of Uranium PKA in tri-Xe implanted UO_2 supercell at 1200K

For the Xe-implanted UO2 supercell, obvious tendency is observed compared to the original UO_2 lattice. The PKA Energy - P_{form} curve is shifted to the left, which indicates the formation of stable Frenkel defect becomes easy. The value of TDE is reduced down to 25~30eV, 10~15 eV for the PKA1 and PKA2 respectively, which were closer (2~3 Å) to the nearest Xenon atom. Even the PKA3, which was 4.1Å away from the nearest Xe, showed the TDE value of 35~40 eV which was far lower than that of Uranium atom in pure UO_2 lattice.

In the spent UO_2 lattice, cumulative fission yield of Xe isotopes in total is about 14~15% of the decayed fissile Uranium[14]. This can directly mean that roughly Xenon's atomic number density is about 0.6% of that of Uranium in the UO_2 lattice, assuming the initial enrichment of Uranium to 4%. Number density of 0.6% seems not significant for the whole lattice. Nevertheless, for every single Uranium atoms, the distance to the nearest Xe atom (d_{Xe}) can be sufficiently small to meaningfully reduce the TDE, which is deciding factor of microstructure radiation resistance.

4. Conclusions

MD simulation using LAMMPS software is performed to calculate and compare the TDE of Uranium atom in the pure UO_2 and Xe-implanted UO_2 supercell, respectively. The definition of TDE selected for this work was Production-probability TDE, which can consider the varying TDE depends on its direction. The value of Uranium TDE in pure UO_2 supercell was 60~70 eV depending on the system temperature of 300~1200K. The TDE of Uranium in Xenon-implanted UO_2 supercell decreased to 10~40eV depends on the PKA Uranium atom's relative position to tri-Xe atoms.

The decrease of TDE caused by the presence of fission gas atoms might be applied for adjustment of input values for ion irradiation-related computer codes such as SRIM.

ACKNOWLEDGMENTS

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