Effects of Thermal Diffusivity Analysis after Irradiation

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Abstract. The diffusion calculation gives a vivid understanding as to what happens in the SiC-cladded material. Molecular Dynamics (MD) and Molecular Statics are being employed to study the diffusion coefficient phenomena. The MD simulations in this study are been built on the ZBL potential. In this work we initially applied the MD simulation for minimization within the temperature range of 1000–3000 K. Then the MOX fuel is then used to perform assessment of radiation damage by ions at burnup temperatures as well. Various chemical states are developed depending on the condition of the fuel. Within the fuel lattice the O atoms break bonds with the U-Pu atoms at higher temperature. The very short diffusion lengths mechanisms results were obtained measured for uranium atom over the course of this 300ps simulation.

1. Introduction

All Nuclear fuel raw materials come from the earth like the fossil fuels. These major materials generate its radioactive by-products from the extraction, enrichment, fabrication, and consumption stages of the process [1]. When a spent fuel is taken out of the reactor it produces a lot of heat as well as radiation. Prior to previous studies metallurgy been used to enhance mechanical properties which have shown that adding a duration quantities with several oxides to ceramic bodies will have large effects on the mechanical performance and microstructure of the ceramic [2-4]. During the inclusion of minor actinides like CmO2, NpO2 and AmO2 with UO2 or ThO2 is desirable so that these species can undergo transmutation in a reactor [5]. Computational models can be used to analyze the nuclear materials so that we can have a better understanding of their behaviors, which can help to increase their efficiency and stability.

Silicon carbide (SiC) has an excellent thermo-mechanical property that makes it a subject of interest for high-temperature mechanical applications. There are numerous phases with high thermal conductivity, including the silicon carbide nanoparticles SiC [6][7] and nanotubes [8], which when added to UO2 pellets makes thermal conductivity improved. However, there are two main conflicting effects as to when introducing the SiC into UO2. Initially, with a high thermal conductivity substance, SiC will enhance the thermal conductivity of UO2. Nevertheless, as a strong-covalent-bond compound, SiC increases the sintering difficulty, reduces the density and consequently weakens the thermal conductivity. Therefore, there shall be a critical sintering condition at which the enhancing effect counteracts the weakening effect.
In this present work, additional calculations and MD simulations were employed to completely evaluate the impact diffusion of (SiC, U-Pu) O2 microstructural-based system. This is very significant towards understanding results of non-uniform lattice change of the mixed oxides from no to high burnup. This affirms the diffusion mechanisms exclusively have a dependent factor on point defects. Emphasis is place on comparing excessive oxygen diffusion coefficient through the comparison in with the mixed oxide fuel. This was ideally made so that predictions can begin to be made for actinide oxides.

2. Interatomic Potential Model

Molecular dynamics simulation provides a unique insight with the atomic-scale processes applicable to solve problems in material science. We performed our simulation by using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code from Sandia National Laboratories [9]. Molecular dynamics simulation uses a force computed in empirical potentials, usually known as classical molecular dynamics.

The interatomic potential energy of a system $V(r)$ of $n$ atoms with coordinates $r_i, r_j, \ldots, r_n$ can be written in terms of one, two, as well as many-body terms as:

$$V(r) = \sum_{i=1}^{n} V_1(r_i) + \sum_{i=1}^{n} \sum_{j>i}^{n} V_2(\eta_{ij}) + \sum_{i=1}^{n} \sum_{j>i}^{n} \sum_{k>j}^{n} V_3(\eta_{ij}, \eta_{jk})$$

(1)

Furthermore repulsive potential described in the short interatomic distances is accustomed to collision cascade simulations as a result of high kinetic energy within atoms. This geometry optimization was carried out in LAMMPS using the empirical potential of nuclear fuel model Ziegler–Biersack–Littmark (ZBL) [10].

$$V(r) \approx \sum_{i=1}^{n} V_1(r_i) + \sum_{i=1}^{n} \sum_{j>i}^{n} V_2^{\text{eff}}(\eta_{ij}) \quad \text{with} \quad \eta_{ij} = |r_i - r_j|$$

(2)

This effective potential $V_2^{\text{eff}}(\eta_{ij})$ perhaps is disintegrated into the long-range Coulombic interaction of ionic particles and the short-range portion.

$$V_2^{\text{eff}}(\eta_{ij}) = V_{LR}(\eta_{ij}) + V_{SR}(\eta_{ij})$$

(3)

Within the charges $q_i$ and $q_j$ distant from each other by $r_{ij}$, the long-range portion $V_{LR}(r_{ij})$ is:

$$V_{LR}(r_{ij}) = \frac{q_i q_j}{r_{ij}}$$

(4)

The interactions with each other and with U-Pu and O typically involved, produces only short-range repulsion and dispersion interactions [11]. The short-range portion can be written as:

$$V_{SR}(r_{ij}) = A_{ij} e^{-\beta_{ij} r_{ij}} - \frac{C_{ij}}{r_{ij}^6} + D_{ij} \left[ e^{-2\rho_0(r_{ij} - r_0^3)} - 2 e^{-\rho_0(r_{ij} - r_0^3)} \right]$$

(5)
With \( Z_i \) and \( Z_j \) representing atomic numbers and \( r_0 \) the Bohr radius.

\[
V_{ZBL}(x) = 0.1818e^{-3.2x} + 0.5099e^{-0.9423x} + 0.2802e^{-0.4029x} + 0.02817e^{-0.2016x}
\]

(7)

All time step of a molecular dynamics (MD) run has an optimisation of the shells to their zero-force positions for each atomic configuration. Short-range interatomic potential has effect on the shell particles which will then decrease some computational activity within the non-bonded interactions. There is no electrostatic force of interaction that exists between the core as well as its own shell. The shell models give a better representation of the equilibrium properties including the elastic constants in static calculations [12].

The Density functional theory (DFT) data was obtained for the selected MOX property of interest using Molecular statics calculations, such as melting temperature, oxygen vacancy migration energy and diffusion coefficient phenomena. The interaction potentials valid for all values of \( r_{ij} \) have been developed for (SiC, U-Pu) \( \text{O}_2 \) by fitting to a database of energies of different structures calculated using DFT [13]. This was done due to reduce any uncertainty produced by the selected functional form and range of activity within the spline.

3. Results and Discussion

The simulation results revealed several derived important aspects using the diffusion coefficients for large and small (SiC, U-Pu) \( \text{O}_2 \) clusters. The results seems to be trailing off towards lower temperatures, which is clearly coherent, since an error or noise could result in an over estimation of the diffusion coefficient when the number of displacement is small. As can be observed in Fig. 1 the derived diffusion coefficient for the small 324-ion (SiC, U-Pu) \( \text{O}_2 \) cluster. The fitted graphs correspond with 0.53 eV and 0.31 eV of activation energy for diffusion.

In Fig. 2, a representative plot of the diffusion coefficients of oxygen derived from 3129 ion with (SiC, U-Pu) \( \text{O}_2 \) cluster results. Here the surface migration results are clearly invalid. The bulk oxygen results show almost similar behavior as the 324-ion cluster, where the diffusion coefficient seems higher than expected at lower temperatures. Although the diffusion length of uranium is almost certainly too short to perform any reliable analysis derived diffusion coefficients which is presented in Fig. 3. The widening of the distribution graphs due to thermal motion of the ions is what is calculated.
Figure 1. Analyzed oxygen diffusion coefficients derived from the small 324 ions (SiC, U-Pu) O$_2$ cluster.

Figure 2. Analyzed oxygen diffusion coefficients derived from the large 3129 ions (SiC, U-Pu) O$_2$ cluster.
4. Conclusion
In this work, a pair in the simulated structure with the interatomic ZBL potential function was used. With the ZBL potential we plotted a comparison of the LAMMPS calculated diffusion coefficients of the MOX with temperature dependence. These graphs could be seen that lattice predictions for the pre-exponential factor were all in the range of $10^{-12}$ to $10^{-10}$ m$^2$/s. A crystal of 3129 ions of (SiC, U-Pu) O$_2$ cluster was simulated at various temperatures. The crystallite which was used during this simulation was cut along the stable (111) plane. This was done to obtain pure stoichiometric MOX. The influence of SiC is more significant in the MOX having results which further verify the developed formula. This model presented can be incorporated into fuel recycling model to improve calculations of the fuel accident conditions and thermal conductivity as well.

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