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Simulation of radiation driven fission gas diffusion in UO_2, ThO_2 and PuO_2

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ABSTRACT

Below 1000 K it is thought that fission gas diffusion in nuclear fuel during irradiation occurs through atomic mixing due to radiation damage. Here we present a molecular dynamics (MD) study of Xe, Kr, Th, U, Pu and O diffusion due to irradiation. It is concluded that the ballistic phase does not sufficiently account for the experimentally observed diffusion. Thermal spike simulations are used to confirm that electronic stopping remedies the discrepancy with experiment and the predicted diffusivities lie within the scatter of the experimental data. Our results predict that the diffusion coefficients are ordered such that $D^*_O > D^*_{Kr} > D^*_{Xe} > D^*_U$. For all species >98.5% of diffusivity is accounted for by electronic stopping. Fission gas diffusivity was not predicted to vary significantly between ThO_2, UO_2 and PuO_2, indicating that this process would not change greatly for mixed oxide fuels.

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1. Introduction

Being the dominant material used as nuclear fuel, the behaviour of UO_2 is of particular importance for the safe and efficient operation of nuclear reactors. Its ability to maintain many of its properties despite extreme irradiation, temperature gradients and chemical changes have made it suitable in this role [1]. However, for the safety of regular burnup fuel and to push for higher burnups, a sound understanding must be achieved of how fission products affect fuel behaviour, such as thermal conductivity [2,3], microstructure [4] or swelling [5,6]. A particular problem for nuclear fuel is the release of the fission gases Xe and Kr from the pellet and into the helium-filled clad-pellet gap. This has the combined effect of reducing thermal conductivity and increasing the pressure of the plenum or cladding [7]. Other than causing an increased risk of centerline pellet melting, the high temperatures arising due to poor gap thermal conductivity also give rise to higher mobility and additional release of fission gases [8–10], further exacerbating the problem. A mitigating factor could be an increase in pellet thermal conductivity as the concentration of fission gas in solution in UO_2 is reduced [11]. Nonetheless, an eventual consequence of continued fission gas release is the rupturing of the cladding material and this represents a significant risk for nuclear reactor operation. As discussed by Turnbull et al. [12] there are three regimes for gas diffusion: i) higher temperature (>1500 K) intrinsic diffusion, ii) intermediate temperature radiation enhanced diffusion and iii) the low temperature (<1000 K) irradiation induced athermal (temperature independent) contribution. In the latter case, which is commensurate with temperatures in the periphery of the fuel pellet, it is thought that the atomic mixing, which occurs during damage, drives gas mobility. Höh and Matzke [13] made similar observations regarding U self-diffusion in UO_2 and UC. There are two stages of the interaction between radiation and a material: electronic and ballistic stopping [14]. Initially fission fragments interact most strongly with the electrons and deposit energy into the material by raising the temperature of the surrounding lattice it passes through. As the fission fragment slows it begins to interact ballistically with atoms in the lattice causing primary knock on atoms to create cascades. This represents two distinct behaviours that must be treated separately during atomistic modeling by using thermal spike and cascade simulations respectively [14–19]. Although this process has been described as temperature independent previously, modeling results indicate that this may not strictly be true due to higher atomic mixing at higher temperatures [14,19].

The underlying mechanisms for fission gas behaviour, as discussed, are complex and inter-related, whereby the temperature and radiation flux in the pellet have important effects [12]. There

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has been a strong focus on investigating the impact of fission gas on thermal conductivity [11], fission gas mobility in the bulk lattice or at extended defects [8–10,20,21] and the behaviour of fission gas bubbles [22–24]. By understanding and predicting these processes over a range of conditions, in particular a large temperature range, a greater understanding of fuel behaviour can be achieved. However, the accuracy of atomic scale simulations is underpinned by the models and theories upon which they are based. In particular, MD simulations are highly dependent upon the ability of a parameter set to accurately describe the properties of both the host UO_2 (or MOX) and the interaction of fission gas with the host lattice. Previously, Cooper, Rushton and Grimes (CRG) developed a many-body potential for pure actinide oxides and their mixed oxides [25,27,28], which is capable of accurately predicting a large number of the thermophysical properties of these systems from 300 K to 3000 K. In particular, this is the first instance of an empirical potential being able to reproduce the bulk modulus of UO_2 from 300 K to 3000 K. Recently, Cooper et al. [29] have developed complementary potentials designed for modeling Xe and Kr behaviour in CeO_2, ThO_2, UO_2 and PuO_2 (or MOX) over a wide range of temperatures, including liquids. Therefore, this potential set is suitable for the investigation of the low temperature irradiation induced contribution to fission gas mobility, whereby the lattice exhibits huge variations in local temperature from the background temperature (600 K–1500 K) to well above the melting temperature at the center of the radiation damage. The CRG model provides a good description for UO_2 of the melting point and volume change during melting [15], the thermal expansion [25] and the classical phonon transport [26]. As modeling high energy irradiation events would be sensitive to these properties, the CRG model is suitable for this study. For example, if a potential model over estimated the melting point or the thermal conductivity, the proportion of time spent in the liquid phase for a thermal spike would be underestimated.

In this work, MD simulations of thermal spikes and damage cascades have been used to investigate the electronic stopping and ballistic phases of irradiation respectively. The relationship between the energy deposited by irradiation and the mean squared displacement (MSD) of Xe, Kr, Th, U, Pu and O is used to predict the diffusion coefficients as a function of fission rate. Comparison is made to previous experiments [12,13] as well as simulations on U mobility during ballistic [19] and electronic stopping [14].

2. Methodology

2.1. Potential model

MD simulations, employing a set of interatomic potentials for ThO_2, UO_2 and PuO_2 derived previously [25,28] [1], are carried out using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [31]. In this model the potential energy, E_i, of an atom i with respect to all other atoms has two components - i) a pair potential description of each system and ii) a many-body embedded atom method (EAM) contribution, using the model of Daw and Baskes [33]:

$$E_i = \frac{1}{2}\sum_j \varphi_{\alpha\beta}(r_{ij}) - G_\alpha\left(\sum_j \sigma_\beta(r_{ij})\right)^{\frac{1}{2}} \quad (1)$$

where the pairwise interaction between two atoms i and j, separated by r_{ij}, is given by $\phi_{\alpha\beta}(r_{ij})$ (equation (2)) and has both long range electrostatic, $\phi_C(r_{ij})$ (equation (3)), and short range contributions. The former are calculated using the Ewald method [34] with the Particle-Particle Particle-Mesh (PPPM) implementation of the method being adopted in order to improve computational efficiency [35]. The short range contributions are described using Morse, $\phi_M(r_{ij})$ (equation (4)), and Buckingham, $\phi_B(r_{ij})$ (equation (5)), potential forms [36,37]. Where α and β are used to label the species of atom i and atom j respectively.

$$\varphi_{\alpha\beta}(r_{ij}) = \varphi_C(r_{ij}) + \varphi_B(r_{ij}) + \varphi_M(r_{ij}) \quad (2)$$

$$\varphi_C(r_{ij}) = \frac{q_\alpha q_\beta}{4\pi\varepsilon_0 r_{ij}} \quad (3)$$

$$\varphi_M(r_{ij}) = D_{\alpha\beta}\left[\exp\left(-2\gamma_{\alpha\beta}\left(r_{ij}-r^0_{\alpha\beta}\right)\right) - 2\exp\left(-\gamma_{\alpha\beta}\left(r_{ij}-r^0_{\alpha\beta}\right)\right)\right] \quad (4)$$

$$\varphi_B(r_{ij}) = A_{\alpha\beta}\exp\left(\frac{-r_{ij}}{\rho_{\alpha\beta}}\right) - \frac{C_{\alpha\beta}}{r_{ij}^6} \quad (5)$$

where $A_{\alpha\beta}$, $\rho_{\alpha\beta}$, $C_{\alpha\beta}$, $D_{\alpha\beta}$, $\gamma_{\alpha\beta}$ and $r^0_{\alpha\beta}$ are empirical parameters that describe the pair interactions between atom i and atom j. In comparison to the original CRG model [25] the updated version of the PuO_2 parameters [28] gives an improved description of the melting point of PuO_2 and is used here.

The second term in equation (1) uses the EAM to introduce a many-body perturbation to the pairwise interactions. The many-body dependence is achieved by summing a set of pairwise interactions, $\sum\sigma_\beta(r_{ij})$, and passing this through a non-linear embedding function: $\sigma_\beta(r_{ij})$ is inversely proportional to the 8th power of the inter-ionic separation (equation (6)) and a square root embedding function is used (equation (1)), where n_β and G_α are the respective constants of proportionality. The derivation of the parameters and a description of the functional terms used in the EAM component are given in Refs. [25,28].

$$\sigma_\beta(r_{ij}) = \left(\frac{n_\beta}{r_{ij}^8}\right)\frac{1}{2}(1+\mathrm{erf}(\kappa(r-1.5))) \quad (6)$$

In order to prevent unrealistic forces occurring at short separations a short range cut-off using an error function is applied at 1.5 Å that reduces the EAM component gradually (see equation (6)). This ensures that there is no discontinuity in the interatomic energy, which would arise from an abrupt cut-off. Buckingham potential parameters for the Xe-Th, Kr-Th, Xe-U, Kr-U, Xe-Pu, Kr-Pu, Xe-O and Kr-O were reported previously [29]. The gas-gas interactions used here were developed previously by Tang-Toennies [38], enabling mixed gas Xe-Kr systems to be studied.

Fig. 1a) shows the variation of the U-O dimer energy (solid lines) and the lattice energy (dotted lines) as a function of U-O separation using the CRG model [25] with κ=5 $Å^{-1}$,10 $Å^{-1}$ or 20 $Å^{-1}$ (see equation (6)). Note that the error function used in the original potential (κ=20 $Å^{-1}$) was too abrupt for the short separations created during damage events, giving a kink in the U-O interaction. It can be seen from Fig. 1a) that κ=5 $Å^{-1}$ was a suitable selection for the error function cut-off ensuring smooth U-O interactions. To prevent unrealistic forces at the short separations accessed due to such high energy events, a ZBL potential was splined to the CRG model. Fig. 1b) shows the U-O dimer energy (solid lines) and the lattice energy (dotted lines) as a function of U-O separation, whereby the CRG model was splined to the short range ZBL

[1] Supplementary material describing the use of this potential for use in GULP [30], LAMMPS [31] and DL-POLY [32] are provided at http://abulafia.mt.ic.ac.uk/potentials/actinides.

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