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A thermal modelling of displacement cascades in uranium dioxide

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ABSTRACT

The space and time dependent temperature distribution was studied in uranium dioxide during displacement cascades simulated by classical molecular dynamics (MD). The energy for each simulated radiation event ranged between 0.2 keV and 20 keV in cells at initial temperatures of 700 K or 1400 K. Spheres into which atomic velocities were rescaled (thermal spikes) have also been simulated by MD to simulate the thermal excitation induced by displacement cascades. Equipartition of energy was shown to occur in displacement cascades, half of the kinetic energy of the primary knock-on atom being converted after a few tenths of picoseconds into potential energy. The kinetic and potential parts of the system energy are however subjected to little variations during dedicated thermal spike simulations. This is probably due to the velocity rescaling process, which impacts a large number of atoms in this case and would drive the system away from a dynamical equilibrium. This result makes questionable MD simulations of thermal spikes carried out up to now (early 2014). The thermal history of cascades was compared to the heat equation solution of a punctual thermal excitation in UO_2. The maximum volume brought to a temperature above the melting temperature during the simulated cascade events is well reproduced by this simple model. This volume eventually constitutes a relevant estimate of the volume affected by a displacement cascade in UO_2. This definition of the cascade volume could also make sense in other materials, like iron.

1. Introduction

In pile the nuclear fuel is submitted to high temperatures and irradiation doses which lead to its structural and chemical evolution. In this process, irradiation-induced defects play a major role. In uranium dioxide, which is the fuel the most used worldwide, the mechanisms of defects production under irradiation have been investigated experimentally as theoretically. First of all the electronic energy loss of a nucleus is likely to induce a thermal excitation of the surrounding matter, which is reputed to induce fission gas resolution in the uranium dioxide lattice for instance [1]. Such an excitation is usually referred to as a thermal spike, often described by the two temperatures model (TTM) [2]. The formation of ion tracks induced by swift heavy ions was also studied experimentally in UO_2, namely by transmission electron microscopy [3].

Displacement cascades resulting from atomic collisions under a nuclear energy loss regime were early described by binary collision approximation (BCA) models such as the one proposed by Kinchin and Pease [4]. Radiation damage estimates were derived from such simple descriptions of radiation events, the most famous probably being the Norgett, Robinson and Torrens (NRT) law [5] which considers that 80% of atomic displacements lead to defect formation. A more precise description of displacement cascades in materials was later provided by molecular dynamics (MD) simulations [6]. In uranium dioxide, most of defects induced by displacement events were shown to recombine [7]. The number of created defects in UO_2 was however found to lie within a factor of two relatively to the NRT law. The displacement threshold energy usually considered for this law does not account for the displacements during the simulation of an energetic cascade event, and its overestimation in this former case appeared to partially compensate for the underestimation of the recombined defect fraction [8].

BCA concepts such as the displacement threshold energy E_d fail indeed in describing high energy cascades which induce a collective motion of atoms. At high energy in UO_2, after balistic collisions occurs a stage which involves the formation of a highly disordered volume, appearing as a melt of atoms in the solid. The high temperature gradient inside the disordered volume was shown to have an effect on the defect formation, for instance via the direct formation of vacancy clusters in uranium dioxide [9] through the vacancy sweeping mechanism [10]. Dislocation loops can also directly be formed under irradiation which corroborates transmission electron microscopy observations of irradiated fuel samples (see for

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instance [11]). The displacement threshold energy is one order of magnitude lower in this disordered volume relatively to values derived from binary collisions [12].

This paper focuses on the thermal history of a displacement cascade. This was carried out by comparing classical molecular dynamics simulations of full displacement cascades and thermal spikes with the solution of the heat equation solution of an initial point-shaped thermal excitation.

2. Modelling

Displacement cascades were simulated using the program CP2K [13,14] and the calculation conditions applied in [12], involving an equilibration step of UO_2 single crystals before a uranium primary knock-on atom (PKA) is accelerated. The Morelon potentials were used [15] to model the uranium dioxide lattice and those of Ziegler, Biersack and Littmark (ZBL) [16] for ballistic collisions. In addition to previous calculations, atom velocities were regularly stored to provide an estimate of the atomic agitation inside the simulation cell.

Spherical thermal spikes were also simulated for further comparison with displacement cascades. The kinetic energy was distributed amongst atoms contained inside a sphere. It was either distributed equally between all atoms or adjusted to follow the radial distribution given by the heat equation solution (Eq. 1 below). The direction of each rescaled atom was either random or kept unchanged after the end of the cell equilibration step. In all cases the maximum amount of energy given to an atom inside the sphere was kept below 20 eV, the minimum displacement threshold energy within the UO_2 lattice [17,18]. These calculations were performed in a NVE ensemble with in most cases a thermostat (velocity rescaling) applied to the first raws at the edges ot the simulation box, this having no visible effect on subsequent results since the amount of energy dissipated at boundaries remained relatively low. The energy E_0 of PKA and thermal excitations were comprised for this study between 0.2 keV and 20 keV, and the temperature T_{eq} of the equilibrated simulation cells was initially either 700 K or 1400 K. Three similar calculations were each time carried out to provide a raw estimate of the dispersion of results. For sake of clarity are only presented in this paper the thermal spikes initiated according to a velocity distribution in agreement with the heat equation solution (Eq. 1), along with vector directions corresponding to those of the equilibrated simulation box. Results indeed appeared to be quantitatively similar whatever the initial conditions applied, notably when they were similar to those applied in [19].

The heat equation $\dot{T} - D_T \Delta T = 0$ was finally used to simulate the thermal dissipation which follows a punctual energy deposition, T being the local temperature function and D_T the thermal diffusivity of the considered medium. The mean kinetic energy e_c of each atom was here assumed to be given by $e_c = 1.5\, k_B T$ although the studied systems are far from thermodynamic equilibrium (with k_B the Boltzmann constant). The Eq. 1 derived from the heat equation for an excitation of energy E_i shows that the kinetic energy of atoms follows a peaked radial distribution which flattens as the time t goes. The density ρ of UO_2 was calculated from [20] at the equilibrium temperature T_{eq} of the material before the thermal excitation occurs (at $t = 0$).

$$e_c(r,t) = \frac{E_i}{\rho \times (4\pi D_T\, t)^{3/2}} \exp\left(\frac{-r^2}{4 D_T\, t}\right) \tag{1}$$

The volume of material above the melting temperature is assumed to constitute an estimate of the volume affected by a displacement cascade. In UO_2, the melting temperature T_m is near 3120 K [20]. This volume V, called here disordered volume, was calculated from MD results by decomposing the simulation box into $1.5a \times 1.5a \times 1.5a$ cubes, some of them containing atoms of mean kinetic energy above $1.5\, k_B T_m$. a is the dimension of an elementary UO_2 cell of 12 atoms. Local temperature and density functions were defined considering little cubes of similar dimension.

V can also be calculated from the heat equation solution described above. It is in this case given by the Eq. 2, with D_T the thermal diffusivity in the solid near T_m (5.76×10^{-7} $m^2\, s^{-1}$ [20]). The differences between the experimentally measured UO_2 properties and their equivalent in the MD modelled material (see [21]) were here assumed to be negligible.

$$V = \frac{32\pi}{3}\left[D_T t \ln\left(\frac{E_i}{1.5\, k_B (T_m - T_{eq})\rho\,(4\pi D_T t)^{3/2}}\right)\right]^{3/2} \tag{2}$$

3. Disordered volume

The Fig. 1 shows the evolution of the volume V, into which the values of the local temperature function (as defined in the previous section) are above T_m. Displacement cascades initiated with a uranium PKA of 20 keV are compared to spherical thermal spikes initiated at same energy E_0. The maximum value of the disordered volume is approximately the same for cascades and spherical thermal spikes. However the lifetime of this volume is almost 10 ps for the thermal spike whereas it barely reaches 3 ps during the full cascade simulation: it behaves as if it was excessively isolated from the surrounding material.

The evolution of V from the heat equation according to Eq. 2 is also reported for a thermal excitation of energy E_i equal to 10 keV. The maximum disordered volume is indeed the same as in 20 keV cascade and thermal spike simulations when $E_i = 10$ keV in this case. This can be explained by the equipartition theorem. Indeed the energy increment E_0 should distribute equally into a kinetic and a potential contribution when the simulation cell is been driving to its thermodynamic equilibrium. In Fig. 2a 10 keV of kinetic energy is converted into potential energy after 0.35 ps 20 keV cascades were initiated (results averaged over 3 cascades). Half of the energy E_0 converts into potential energy after around 0.35 ps for cascade simulations.

Similar curves were obtained for various T_{eq} and E_0. Besides for all initial conditions (temperature, cascade energy and initial pK_a position and direction), the volume V appeared to be maximum at approximately the same time, between 0.3 and 0.4 ps (around 0.32 ps for 20 keV cascades in a simulation cell at 700K as shown

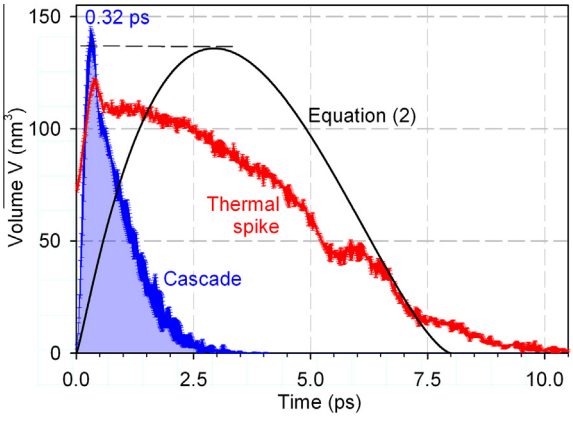

Fig. 1. Volume V at $T \geqslant T_m$ during a 20 keV cascade simulation (blue), a thermal spike simulation of same energy E_0 (red) and calculated from Eq. 2 with $E_i = 10$ keV (black), in a UO_2 medium at T_{eq} = 700 K.