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Molecular dynamics study of the bulk temperature effect on primary radiation damage in uranium dioxide

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ABSTRACT

The effect of bulk temperature on the primary damage induced by a displacement cascade was investigated in uranium dioxide using classical molecular dynamics simulations. In this study, the Morelon potentials were used to model the middle-range interactions between the atoms that constitute the host matrix during the radiation events. Cascades were initiated by accelerating a uranium primary knock-on atom at 10 keV inside a perfect UO_2 lattice at a temperature between 700 K and 1800 K, a range which comprises in-pile temperatures of oxide fuels in light water reactors in standard operating conditions. Cascade overlap sequences were also simulated at 700 K and 1400 K in order to study the radiation damage accumulation in the oxide fuel. This study reveals the maximum damage level which the material can accommodate for decreases with the temperature. Furthermore the direct formation of vacancy clusters under irradiation is considerably slowed down above 1000 K, notably during cascade overlap sequences.

1. Introduction

Irradiation induced damage plays a major role in the microstructural evolution of uranium dioxide at all stages of the fuel cycle. It is therefore an important subject as this ceramic material is the most widely used nuclear fuel worldwide. In normal operating conditions, the fuel pellets are subjected in light water reactors to high radial thermal gradients. Its typical temperatures at the centre and at the extreme periphery of the pellets are respectively around 1350 K and 750 K.

In this study the effect of bulk temperature on primary radiation damage was investigated. The radiation events are here energetic displacement cascades. Classical molecular dynamics simulations were performed since most of binary collision approximation (BCA) models do not account for any temperature effect. The threshold displacement energy can nonetheless vary with the temperature in the Marlowe code [1]. This may lead to a slight reduction of the calculated number of radiation-induced Frenkel pairs when the temperature rises. The minimum energy transferred to displace a lattice atom should indeed decrease since the strength of its binding to the surrounding lattice drops (to such a level that it can freely move above the melting temperature).

Molecular dynamics simulations were run at different temperatures from 300 K to 1800 K. Energetic displacement cascades were initiated at 10 keV. Cascades were also successively overlapped within a same simulation cell in order to study radiation damage accumulation in the material. The results in terms of nature and number of crystalline defects were then compared. They revealed that the bulk temperature of the oxide impacts the material damage level the material can accommodate for and also defect clustering processes under irradiation, in particular when the radiation damage accumulates [2].

2. Simulation conditions

Middle-range U and O atom interactions were modelled by the set of empirical potentials based on a rigid ion model developed by Morelon et al. [3]. It is in particular fitted to reproduce the experimental defect formation and migration energies [4]. It describes quite satisfactorily and over a large range of temperatures some basic UO_2 properties such as thermal expansion coefficient and heat capacity [5]. During energetic cascades, interatomic distances can be much less than equilibrium distances, therefore the universal Ziegler–Biersack–Littmark potential [6] was used to describe interactions at less than 0.16 nm.

The cascade overlap sequences at 700 K were presented in a previous paper [7]. The calculation conditions of additional simulations are basically the same as those reported in this study. The

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Table 1
Simulation conditions of displacement cascades.

Initial energy of the uranium PKA (keV)	Temperature of the box (K)	Max. number of cascade overlaps
10 [7]	700	22
10	1100	1
10	1400	22
10	1800	1

simulation ensemble is a cubic cell with periodic boundary conditions containing 1.875×10^5 atoms. A UO_2 single crystal is first equilibrated under NPT conditions for 20–30 ps using a Nose–Hoover thermostat [8]. A displacement cascade is then initiated under NVE conditions (energy is not conserved at the cell sides as described below) by accelerating a uranium projectile to a kinetic energy of 10 keV. This energy is wholly transferred to the material through nuclear interactions. The thermostat applied during the cascade event consists in rescaling the velocities of the atoms within the first three planes at the sides of the simulation cell.

Several cascades were simulated in similar conditions (same simulation cell and cascade energy) to estimate the scatter associated to the results (error bars). To this aim accelerated projectiles of various initial position and direction were defined. In each set of conditions nine cascades were simulated in an initially undefective material. Three cascade overlap sequences were also run in the same way as in [7]: a new cascade event is added in the simulation cell each time it returned to its equilibrium temperature (after few tens of ps). The total number of defects was deduced by decomposing the UO_2 lattice into Voronoi polyhedra centered on the different network sites [7]. The simulation conditions of displacement cascades are reported in Table 1.

3. Damage level

The number of defects after a single cascade simulation of 10 keV is reported in Fig. 1 as a function of the bulk temperature of the oxide. The counted defects are here the vacancies and interstitial atoms irrespective of whether they form clusters or not. However in the case of dislocation loops, a specific data treatment was carried out: lattice distortions they induce often result in a drift of the surrounding lattice planes which the Voronoi decomposition process could therefore detect as defective.

From 1100 K, the oxygen sublattice becomes mobile at the timescale of displacement cascade simulations (around 20 – 30 ps). The number of temperature induced oxygen Frenkel pairs during the cascade events was estimated by performing molecular dynamics simulations in cascade conditions (see part 2), but without any atom acceleration. The number of O Frenkel pairs resulting from a cascade (red dots in Fig. 1) is then evaluated above 1100 K as excess number relative to thermal O Frenkel pairs.

The number of radiation induced defects seems to be barely dependent on the bulk temperature. The evolution of the number of defects remains low with respect to the scatter of the results (from nine simulated cascade events).

Atoms are however far easier to displace in the volume disordered by the cascade (also referred to as the thermal spike [9]). A previous study [10] indeed showed that the threshold displacement energy is considerably lower when a thermal spike occurs in UO_2. Besides the size of the thermal spike increases with the temperature [11] (it depends on $(T_m - T)^{-1}$, where T_m is the material melting temperature, around 3120 K [12]): this explains why the number of displaced atoms also rises during the simulated cascade events [10].

If more atoms are displaced whereas the number of defects remains stable, there should be more Frenkel pair recombination.

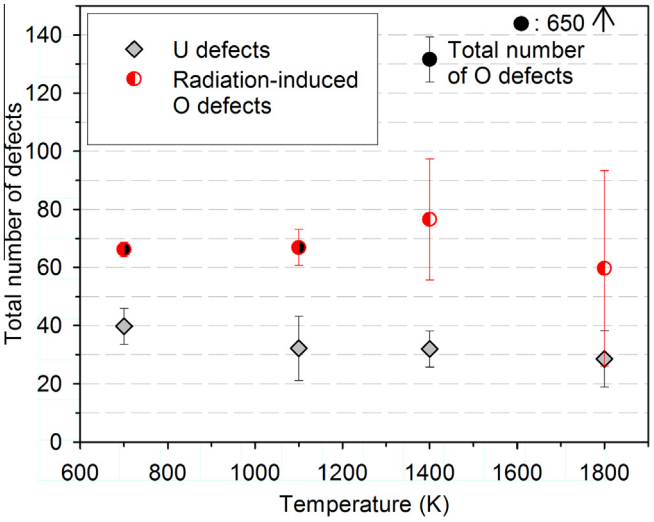

Fig. 1. Total number of uranium (grey dots) and oxygen radiation-induced defects (red dots) as a function of the bulk temperature after a 10 keV displacement cascade. Above 1100 K, the temperature induces the formation of additional oxygen Frenkel pairs (the total number of O defects, in black dots, therefore increases). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

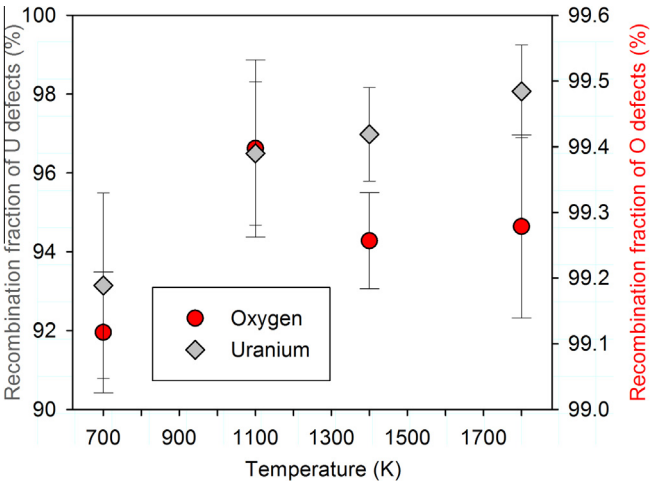

Fig. 2. Recombination fraction R of uranium and oxygen defects as a function of the bulk temperature of the simulated UO_2 immediately after a 10 keV displacement cascade.

The immediate recombination rate R is here defined as the fraction of displaced atoms that resulted in a Frenkel pair of defects at the end of the cascade simulation (as in [13]). Fig. 2 reveals that this parameter is an increasing function of the temperature, in particular for uranium defects. The error bars with respect to oxygen defects are however very large. The scatter of oxygen data (from nine different cascade simulations) makes them hard to comment.

The numbers of uranium and oxygen defects during the 10 keV cascade overlap sequences carried out at 700 K and 1400 K are reported in Fig. 3. Dash lines show how the number of defects would evolve if the damage simply adds indefinitely each time a new cascade event occurs. The number of defects clearly levels off in both U and O sublattices. After 15 cascades, its slight increase is attributed only to the growth of the volume impacted by the successive cascade events. The defect density actually saturates, in particular at 700 K [7]. The maximum level of damage which the material can accommodate for appears however to be very