Damage characterization of displacement cascades in (U,PU)O_2 fuels by MD

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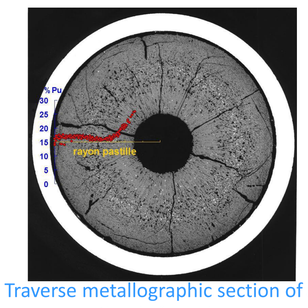
Introduction

Nuclear fuel based on uranium-plutonium oxides (MOX) undergoes under significant structural changes during its lifetime inside a nuclear reactor. For instance, the concentration of Pu varies within the fuel pellet, which affects its thermomechanical behaviour. However, the exact nature of these microstructural changes and their origin are still not fully understood. Moreover, safeness and effectiveness must be assured during processes involving MOX fuel such as fabrication, operation and recycling. Experiments of MOX under irradiation involve high temperatures (~3000 K) and pressures (1-10 GPa), thus they are difficult to carry out. Therefore, Molecular Dynamics simulations were the preferred option.

Firstly, it was carried out an assessment of interatomic potentials based on their resulting thermo-mechanical properties, such as; cracking propagation, elastic constants, stress-strain curves, etc. This was done over the whole range of plutonium content and from 300 K to the melting point. Secondly, it was investigated the primary damage created by an irradiation event.



Traverse metallographic section of a Phenix fuel pin.

Methodology

Empirical potential.

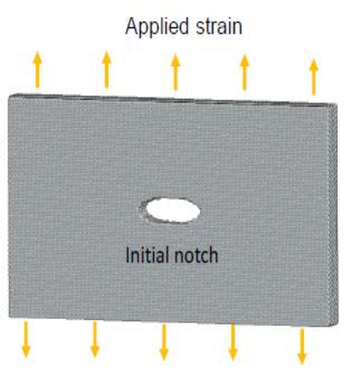
The difference between the form of the empirical potentials is shown below

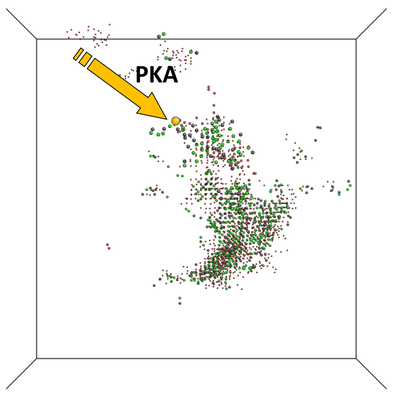
	Coulomb $\frac{q_i \cdot q_j}{4\epsilon_0 r_{ij}}$	Buckingham $A_{ij}e^{-(r_{ij}/\rho_{ij})} - \frac{C_{ij}}{r_{ij}^6}$	Morse $D_{ij}e^{-2\gamma_{ij}(r_{ij}-r_0)} - 2D_{ij}e^{-\gamma_{ij}(r_{ij}-r_0)}$	EAM
Cooper	✦	✦	✦	✦
Potashnikov	✦	✦		
Arima	✦	✦		

- Interatomic potentials: Cooper (M. Cooper et al. J Nucl Mater, 461, 206 (2015)), Potashnikov (S. Potashnikov et al., J Nucl Mater, 419, 217 (2011)), Arima (T. Arima et al., J Alloys Compd, 400, 43 (2005)).
- LAMMPS code was used for the MD simulations.

Stress-strain curves.

- Impose a uniaxial deformation on the box with a constant strain rate until the complete cracking of the system.
- The stress component corresponding to the direction of deformation is recorded while relaxing under NσT the other components of the stress tensor.





Cracking propagation

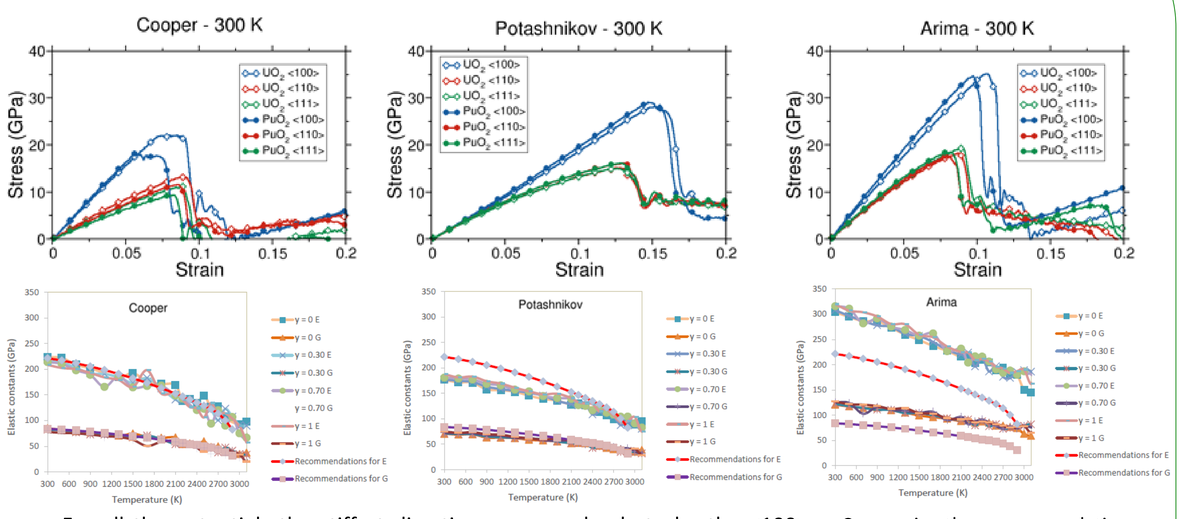
- A constant strain (10^8/s) is applied perpendicularly to the initial crack.
- The system included 4.4×106 atoms with box size equal to 240×70×4 nm

Cascade method

The system is first relaxed until thermal equilibrium is reached under NPT. When the cascade start the system is relaxed under NVE. Periodic boundary conditions are used. System size: 39×39×39 nm

- PKA energy: 75 keV
- PKA: U, Pu
- Plutonium content: 25% and 50%
- For statistics: 5 cascades for each plutonium content with random PKA orientations
- Interatomic potentials: Cooper and Potashnikov

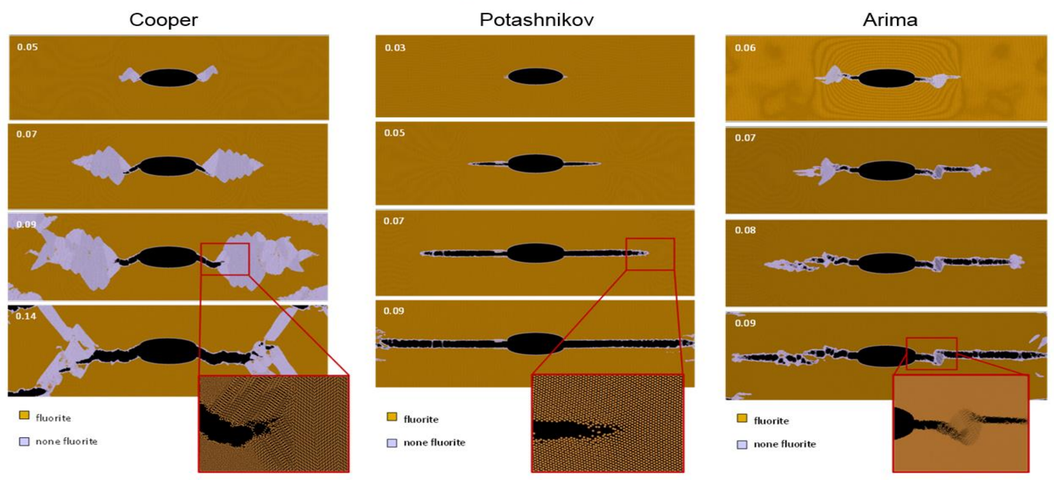
Stress-strain curves



- For all the potentials the stiffest direction appears clearly to be the <100> direction.
- For Cooper's potential, the <111> direction seems to be the weakest direction, which is in agreement with theoretical and experimental results.
- No important difference between plutonium and uranium dioxide was found.
- Concerning the recommendations for the elastic constants Cooper>Potashnikov>Arima J. Fink, J. Nucl Mater, 279, 1 (2000)

Cracking propagation

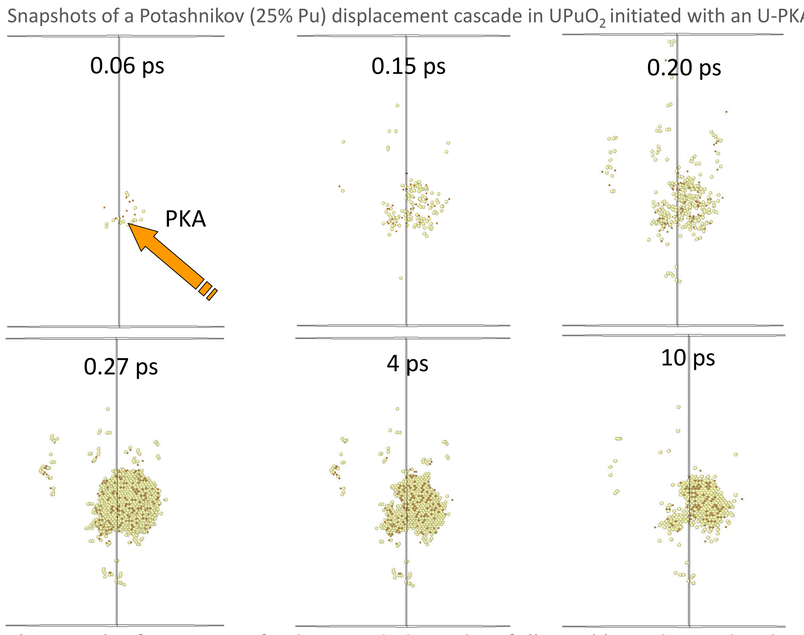
15% Pu, <111>, 300 K



- Crack propagation is different for each potential as can be seen in the snapshots
- A plastic zone can be found for Cooper, whereas for Potashnikov is inexistent.
- The yield strength is the highest for Arima's potential at 10 GPa followed by Cooper at 8 GPa, and Potashnikov at 5 GPa.

Displacement Cascades

Snapshots of a Potashnikov (25% Pu) displacement cascade in $UPuO_2$ initiated with an U-PKA

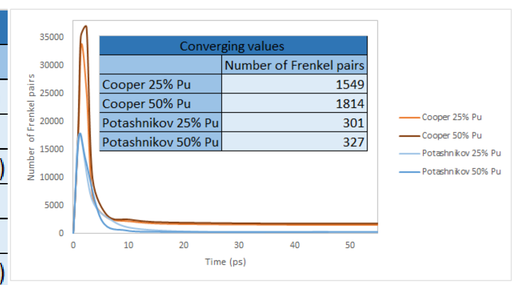


We observe the formation of sub-cascade branches followed by a thermal spike and finally recrystallization.
At the end of the cascade, no amorphization is observed, and only a few point defects are created.

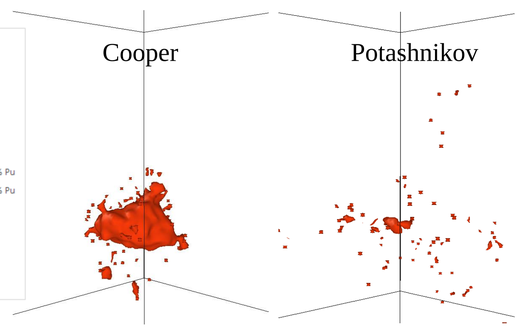
Number of displaced atoms after 10 ps averaged over 5 independent cascades

Number of displaced atoms		Cooper	Potashnikov
25 % Pu	U	2889 (15.4 %)	2440 (11.0 %)
	Pu	991 (5.2 %)	834 (3.7 %)
	O	14881 (79.3 %)	18739 (85.1 %)
50 % Pu	U	1934 (9.9 %)	1735 (7.4 %)
	Pu	1994 (10.2 %)	1749 (7.4 %)
	O	15443 (79.7 %)	19941 (85.1 %)

Number of Frenkel pairs averaged over 5 independent cascades



Defect analysis snapshots for Cooper and Potashnikov potentials based cascades at the end of the relaxation (~50 ps)



- The number of Frenkel pairs is higher for Cooper's potential that for Potashnikov's.
- At the end of the cascade, defected material is found in the main cascade body for Cooper potential, whereas for Potashnikov potential only a few point defects are created.
- The number of displaced atoms tended to follow stoichiometry.

Conclusions

- The results show clearly that the elastic stiffness constants are best reproduced with Cooper's potential, which has been fitted on mechanical properties determined at 0 K with DFT calculations. Potashnikov potential gives fairly good agreement while Arima's potential overestimates largely the elastic stiffness constants.
- Analysis of stress-strain curves obtained with uniaxial loading shows for the three potentials that the <111> crystallographic direction of the fluorite structure is the weakest, as expected.
- For all these mechanical properties, the three potentials show little dependence on the plutonium content (< 5%) as the experimental data show.
- The behaviour during crack propagation simulations is also very different between the three potentials. For Cooper and Arima potentials crack propagates through secondary phase that appears ahead of the crack tip leading to an unexpected plastic-like behaviour in the stress-strain curves. Conversely, for Potashnikov potential crack propagates by cleavage, which is typical of a brittle-like behaviour.
- Concerning the displacement cascades, the number of Frenkel pairs is higher for Cooper's potential that for Potashnikov's. This can be explained by the defected zone found in the main cascade body using Cooper potential. The number of displaced atoms tended to follow stoichiometry.
- As future work, a more deep analysis will be carried out on cascades. In the case where phases can be found, the phase field method may be applied.