Atomic scale insights on the microstructure evolution of urania under irradiation

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Introduction

Urania is commonly used as a fuel in nuclear industry. Urania is heavily irradiated during its in-reactor stay, and faces drastic microstructural modifications, including few percents swelling and increase of dislocation density. Dislocations are identified as perfect dislocations loops and transform with increasing fluence into lines at sufficiently high temperature – i.e. at 600 °C. However, the early stages of their nucleation are hardly attainable experimentally. One commonly infers that their nucleation is related to the aggregation of point defects or defects clusters into dislocations.
In the present paper, we clarify the first steps of the effect of irradiation on urania by combining molecular dynamics simulations and experimental investigations [1].

Methodology

Molecular dynamics simulation
- Morelon potentials
- NPT ensemble

Irradiation
- Frenkel pairs creation every 2 ps → dose rate K_0 = 1.6 10^9 dpU/s
  → recombination regime

Analysis
- Dislocations with DXA algorithm in Ovito [2]
- Point defects with Voronoï
- Swelling extracted by indexing XRD peaks

Comparison with experiments:

Experimental dose rate $K_0 = 10^{-3}$ dpa/s

Characteristic distance after time τ = 1s of irradiation → $(V_{at}/(K_0 \times \tau))^{1/3}$ = 34 Å
Characteristic distance for self-diffusion → $(D \times \tau)^{1/2}$

D(U à 900K) = 10^{-31} m²/s [3]→ 1.18 10^{14} s for 34 Å length diffusion
→ recombination regime in experiments

Dislocation density ρ in recombination regime: $\rho \propto K_0^{1/6}$ [4]

→ Rescaling of dislocation densities by a factor of ~ 10^{-2}

Nucleation and growth of dislocations

Stage 1 (up to 0.07 dpU marked with grey dashed line):

- Increase of point defects concentrations, with equal concentration of vacancies and interstitials (bottom of Figure) → as predicted by rate equations.
- Recombination lifetime of uranium Frenkel pairs = 1.6 ps according to the relation $\tau_{FP} = 1/K_0\tau_1^2$ → spontaneous recombination.

Stage 2 (between 0.07 dpU and 0.25 dpU (marked with red dashed line):

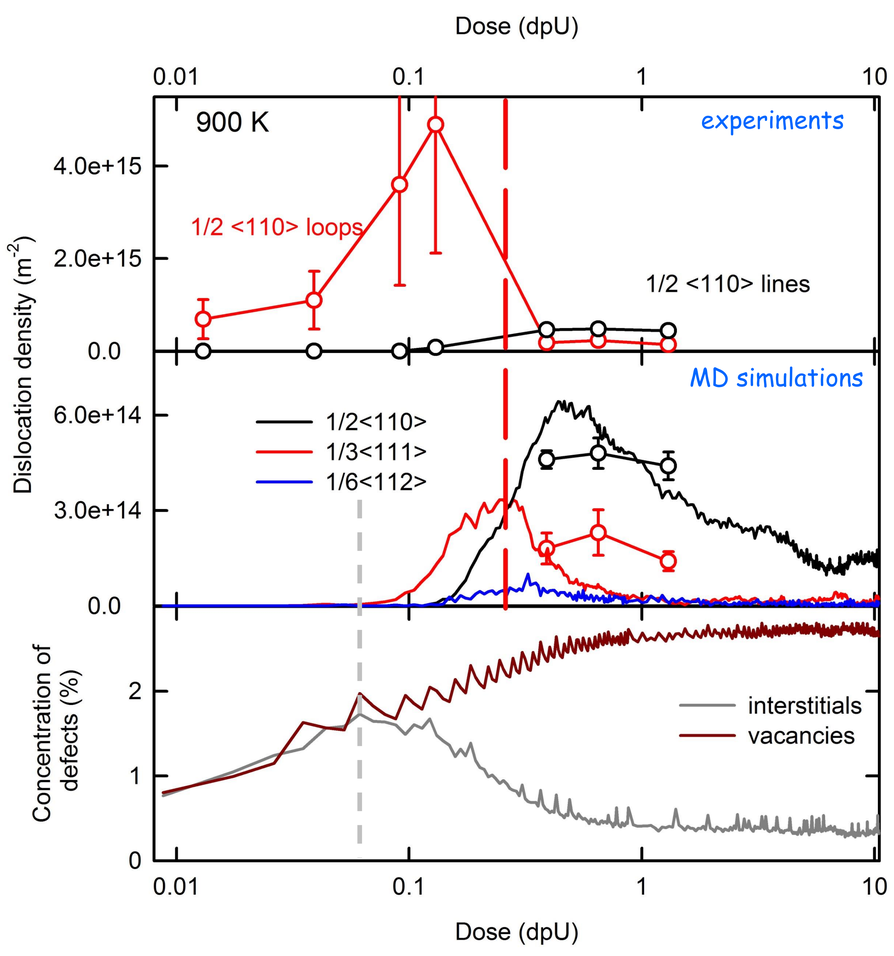
- Frank loops nucleate (1/3<111> middle of Figure) from point defect clustering.
- Interstitials decrease (bottom of Figure), indicating a dislocation bias for them.
  → Frank loops of interstitial character.

Stage 3 (0.25 dpU up to 0.35 dpU):

- Frank loops transform into perfect loops (1/2<110> middle of Figure)
- Mechanism suggested by Baufeld [5]: $\frac{a}{6}\langle 11\bar{2}\rangle + \frac{a}{3}\langle 111\rangle \rightarrow \frac{a}{2}\langle 110\rangle$
  see Shockley partials (1/6<112>) and their connections with Frank and perfect loops (inset vertical Figure).
- Frank loops not seen experimentally due to their sub-nanometer size.
- Perfect loops observed in experiments and calculations.

Stage 4 (above 0.35 dpU):

- Perfect loops convert into lines.
- Forest dislocations stabilize above 1 dpU, dislocation densities of 5x10^{14} m^{-2}.
- Defect concentrations stabilize with vacancies / interstitials = 6
  3-4 estimated by Nakae *et al* [6] (bottom of Figure).



Swelling

Comparison with experiments:

- Lattice expansion close to experiments [7,8,9]
- Lattice expansion – contraction peak observed experimentally in nuclear fuel [10]

Swelling deconvolution:

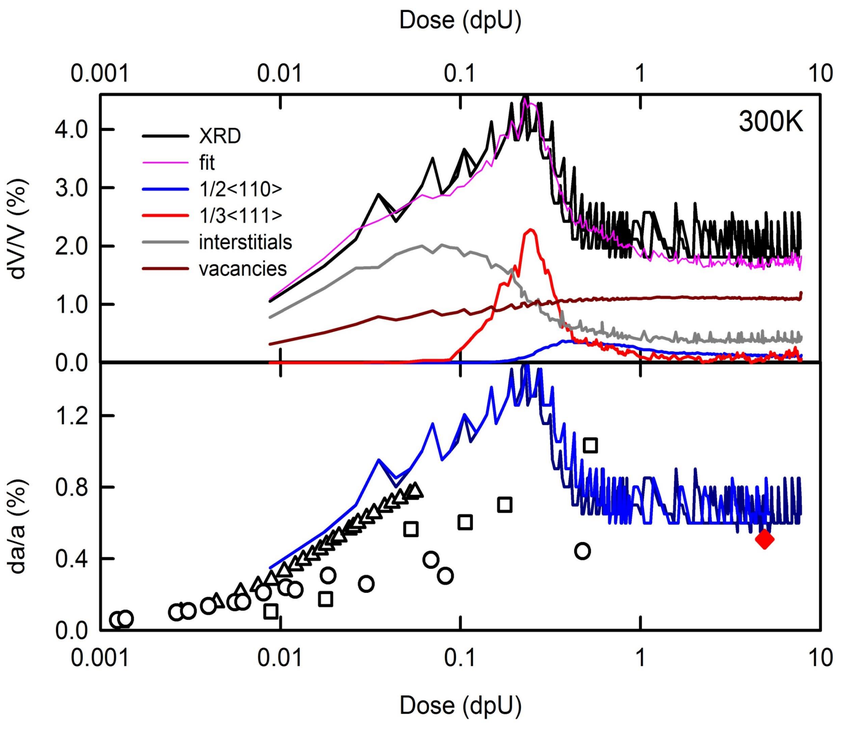
$$\frac{\Delta V}{V} = V_I^f \times \rho_{line} + \frac{\overline{R_{loop}} \times b}{2} \times \rho_{loop} + \frac{V_{rel}^I}{\Omega} \times C_I + \frac{V_{rel}^V}{\Omega} \times C_V$$

V_{rel}^V and V_{rel}^I	relaxation volumes of vacancies and interstitials
Ω	atomic volume (Ω)
½ $\overline{R_{loop}} \times b$	formation volumes of dislocation loops
b	Burgers vector
$\overline{R_{loop}}$	average loop radius
V_I^f	formation volume of lines
ρ_{line} and ρ_{loop}	line and loop dislocation densities
C_I and C_v	interstitial and vacancy concentrations

Swelling stage
- 0.0 to 0.1 dpU → point defects
- 0.1 to 0.2 dpU → enhanced by Frank loops

Contraction stage
- 0.2 to 1.0 dpU → transformation of Frank loops into unfaulted loops dislocations.

Residual swelling at steady state is mainly related to vacancies and to interstitials.



Conclusion

Starting from a defectless urania, we observe the nucleation and growth of dislocations under Frenkel pairs accumulation.

Detailed analysis shows a four stages evolution :
(i) an increase of point defects
(ii) then the nucleation of Frank loops 1/3 <111> from the aggregation of point defects
(iii) the transformation of Frank loops into perfect loops 1/2 <110>
(iv) and finally their stabilization as lines.

Our simulations also show a swelling up to 3.2% when point defects are present, this swelling suddenly decreases to 1.5%, as soon as dislocations nucleate.

Nucleation and growth of dislocations in UO2 operate in the spontaneous recombination regime.

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