Mechanical property evolution with radiation damage in (U,Pu)O_2 a MD simulation study

L. van Brutzel, H. Balboa, A. Chartier

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Mechanical Property Evolution with Radiation Damage in $(U,Pu)O_2$:

a Molecular Dynamics Simulation Study

L. Van Brutzel, H. Balboa, A. Chartier

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The Nuclear Materials Conference

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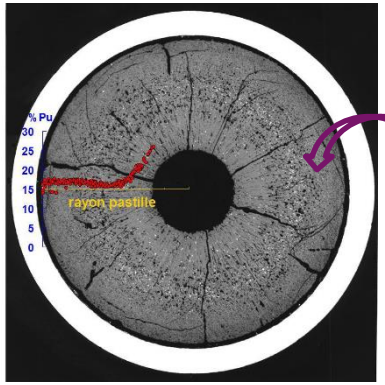
materialstoday

Connecting the materials community

MOX FUEL DESIGN, AN ENGINEERING CHALLENGE

Transverse metallographic section of a Phénix fuel pin



In reactors

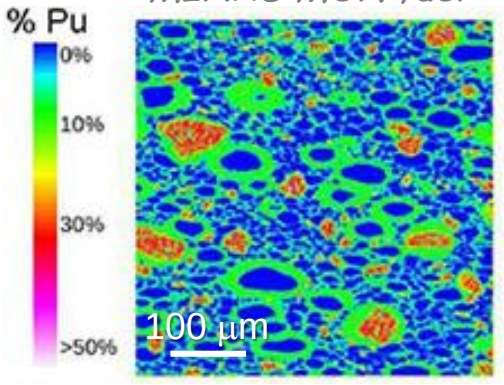
- Temperature, ∇T
- Stress, $\nabla \sigma$
- Burn-up

Microstructure heterogeneity

- High Pu zone

↗ radiation damage

EPMA images of MINAS MOX fuel

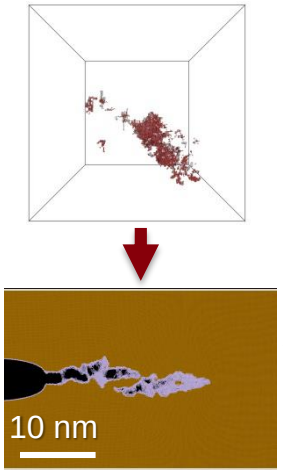


(Oudinet JNM 375 (2008) 86)

Origin of microstructure evolution?

Mechanical properties?

- **Method**: atomistic simulations → nanoscale $(U,Pu)O_2$ Molecular Dynamics
- **Goal**:
  - Understanding phenomena
  - Input parameters for mesoscale models



radiation

mechanical properties

OUTLINE

I. Properties of non-irradiated $(U,Pu)O_2$

II. Structure evolution with radiation damage

III. Mechanical behavior for irradiated $(U,Pu)O_2$

IV. Summary

EMPIRICAL POTENTIALS

- Rigid ion potentials (no charge transfer)

$$\phi(r) = \underbrace{\mathrm{E}_{\mathrm{Coulomb}}(r^{-1}) + \mathrm{E}_{\mathrm{rep}}(e^{-r}) + \mathrm{E}_{\mathrm{vdW}}(r^{-6})}_{\text{2-body}} + \underbrace{\mathrm{E}_{\mathrm{EAM}}(r,\rho)}_{\text{many-body}}$$

- Two potentials have been assessed systematically:
  - **MOX-07**: 2-body (Potashnikov JNM 419 (2011) 217)
  - **Cooper**: 2-body + EAM (Cooper JNM 461 (2015) 206)

    E_{EAM} ⇨ reproduces Cauchy violation ($C_{12} \neq C_{44}$)

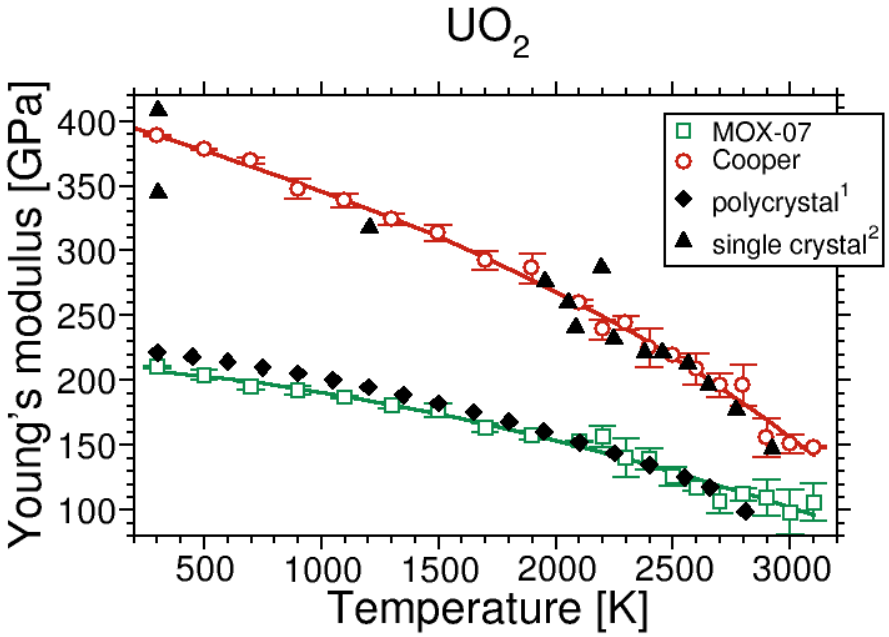
- For both potentials thermodynamic properties fit Fink's recommendation (Fink JNM 279 (2000) 1)
  - Lattice parameter, thermal expansion → 2100 K
  - Vegard's law verified → 2100 K ⇨ $(U_yPu_{1-y})O_2$ solid solution
  - Specific heat: $C_p = \frac{1}{n}\left(\frac{\partial H}{\partial T}\right)_P$ → λ-peak = superionic transition at T ≈ 0,8 T_m

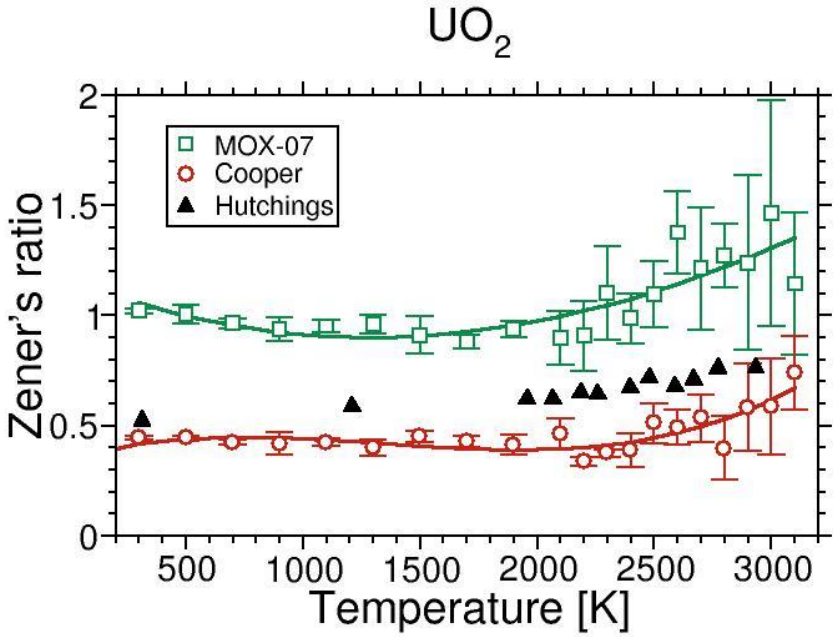
(Balboa JNM 495 (2017) 66)

POTENTIALS: MECHANICAL PROPERTIES I

Elastic stiffness coefficients (C_{11}, C_{12}, C_{44})

- Anisotropy factor Zener's ratio: $Z = \frac{2C_{44}}{C_{11}-C_{12}}$, Z=1 → isotropic (cubic: Z<1 → $E_{111}<E_{100}$)



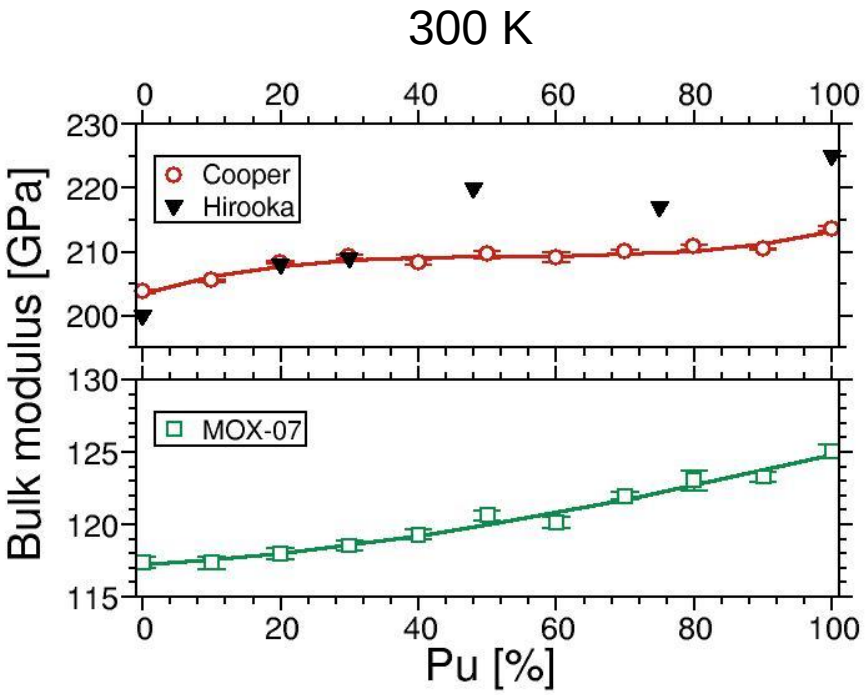


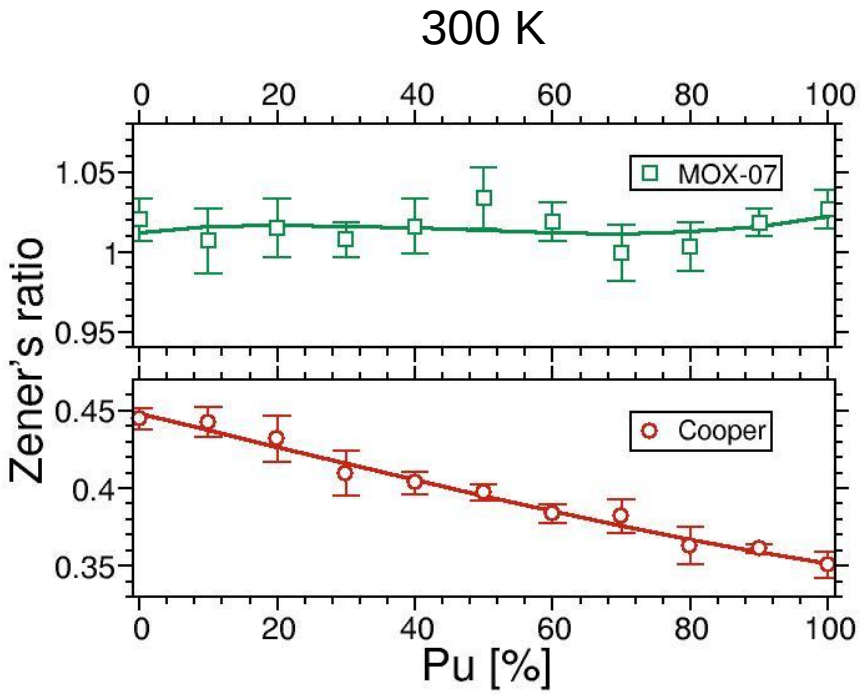
- Cooper fit single-crystal exp.[2] / MOX-07 fit polycrystal recommendation[1]
- Cooper: **anisotropic** / MOX-07: **isotropic**

[1] (Martin High Temp. High Press. 21 (1989) 13)
[2] (Hutchings J. Chem. Soc., Faraday Trans. 83 (1987) 1083)

POTENTIALS: MECHANICAL PROPERTIES II

Influence of the Pu content





- **Bulk modulus**: 5% increase with Pu content for both potentials
- **Zener**: Cooper decreases with Pu content / MOX-07 constant

Empirical potential main difference = **anisotropy**

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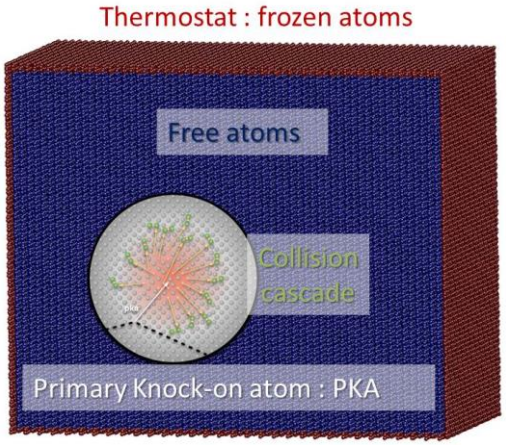
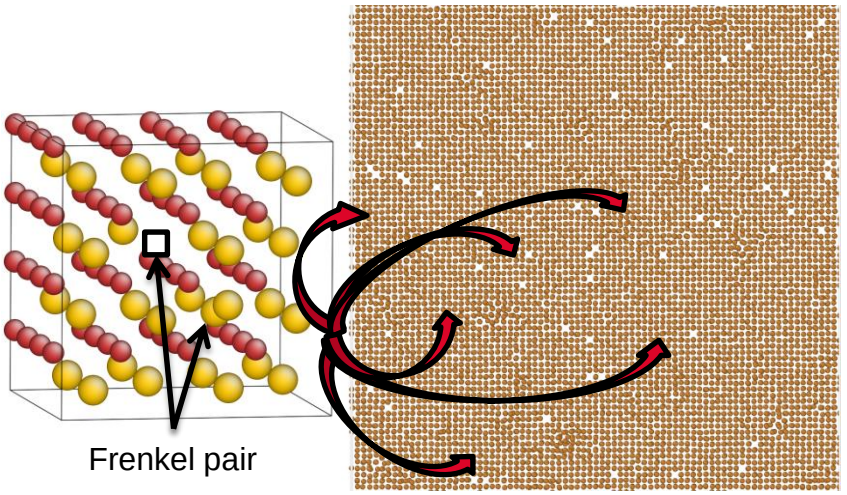
Study of Primary Damage with Molecular Dynamics

- Code: LAMMPS
- $(U,Pu)O_2$: U and Pu randomly distributed



- Primary Damage: **Displacement Cascades**
  Primary Knock-on Atom: Cation
  E_{PKA} = 2 → 75 keV
  T = 300 K
  ⇨ **1 radiation event** = parameter for FPA

- Dose Effects: **Point Defects Accumulation**
  Creation of random Cation Frenkel pairs
  Dose rate: 10^9 dpc/s
  Dose: 0 → 3 dpc
  300 – 1600 K (relaxation NPT) → Swelling

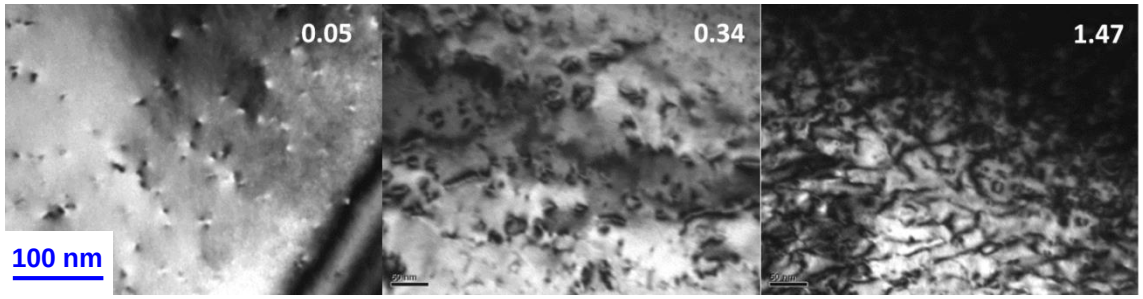


IRRADIATION INDUCED DISLOCATIONS

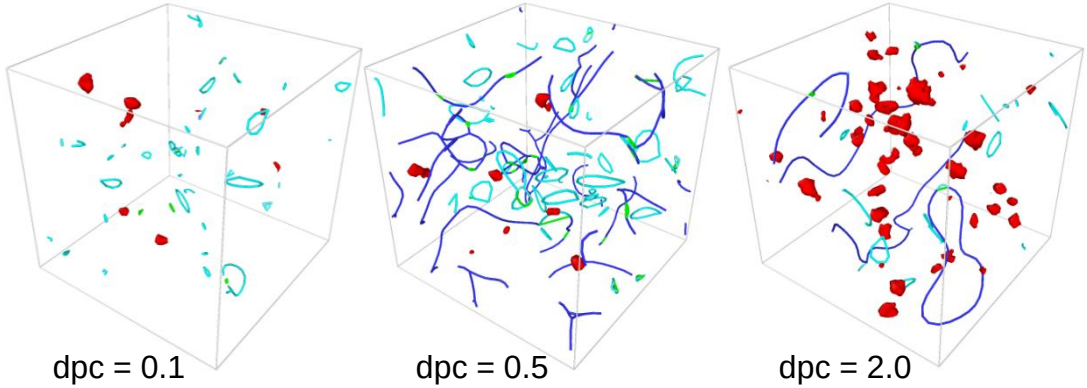
Microstructure evolution

- MD analysis: **OVITO**
  - Point defect: Voronoï cell
  - Dislocation: DXA

Experimental: TEM images of irradiated UO_2



MD simulations: FPA in $(U,Pu)O_2$ calculated with MOX-07



5 stage process

1. point defects creation
2. aggregation into clusters
3. nucleation of Frank loops
4. transformation into unfaulted loops via Shockley that in turn grow
5. reorganization into forest dislocations

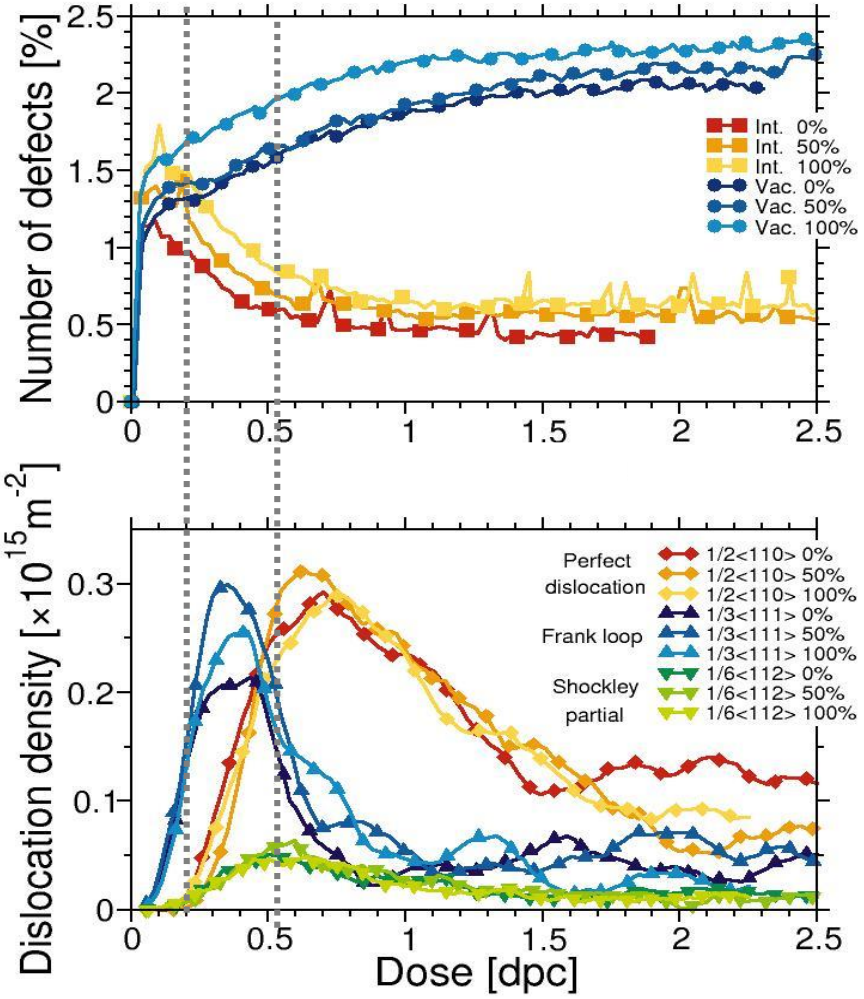
Exp: 2 nm min *(TEM resolution)* **½ <110> loops → ½ <110> lines**

MD: point defects → ⅓<111> loops → ½ <110> loops → ½ <110> lines
Frank loops **perfect loops**

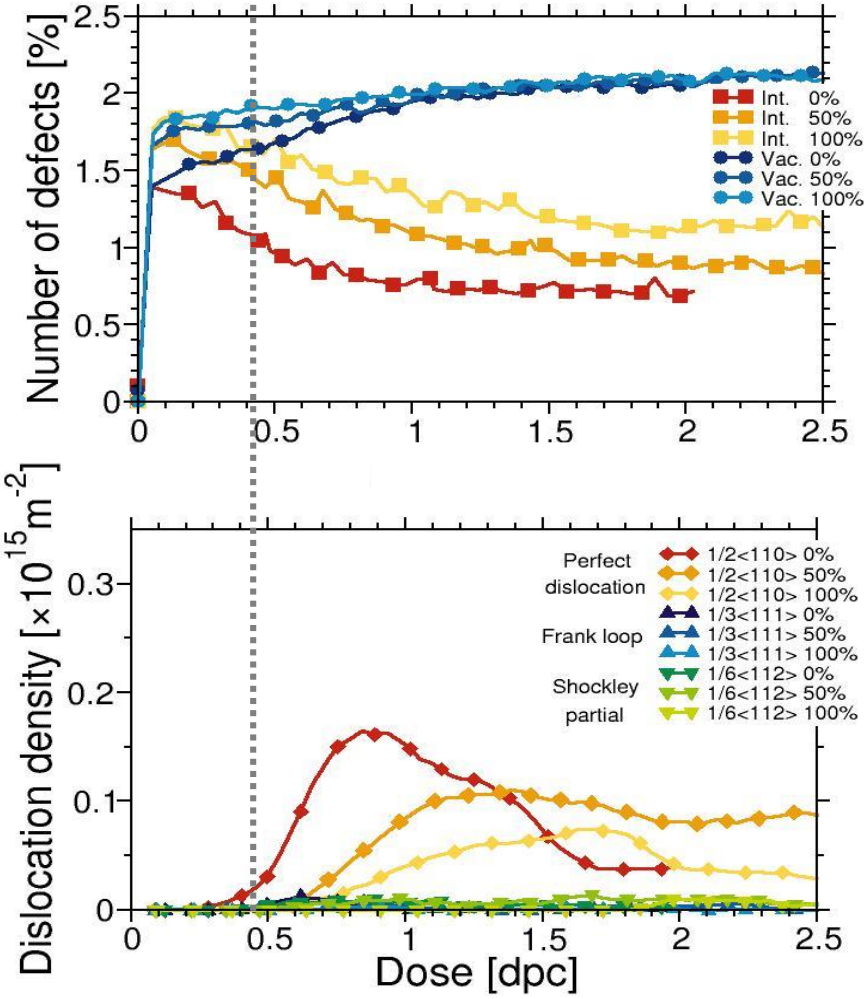
Onofri J. Nucl. Mat. 482 (2016) 105
Onofri J. Nucl. Mat. 494 (2017) 252
Chartier Appl. Phys. Lett. 109 (2016) 181902
Balboa https://doi.org/10.1016/j.jnucmat.2018.07.056

MICROSTRUCTURE EVOLUTION VS DOSE

MOX-07



Cooper



- Cooper: more point defects, less dislocations / more Pu influence

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MECHANICAL PROPERTIES WITH DOSE - METHOD

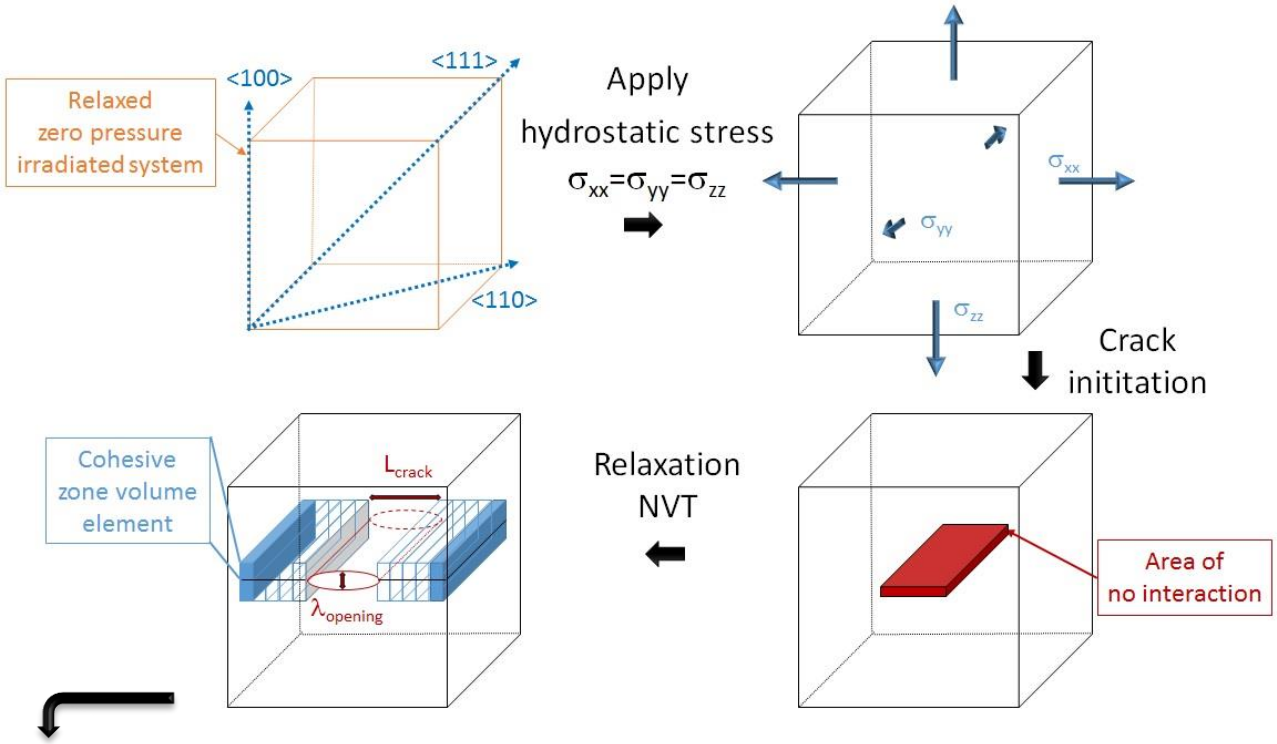
- Elastic constants: small deformation

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl}$$

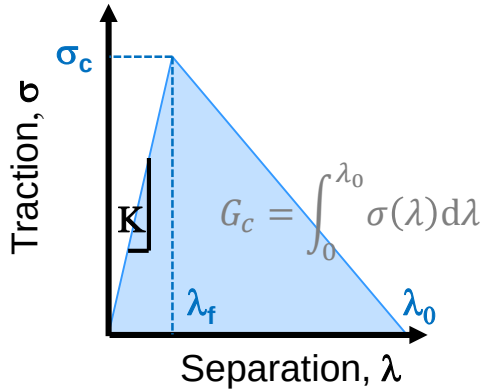
- Cohesive strength: Yamakov method

→ avoid strain-rate issue (phase transition at the crack tip)

Yamakov J. Mech. Phys. Solids, 54 (2006) 1899



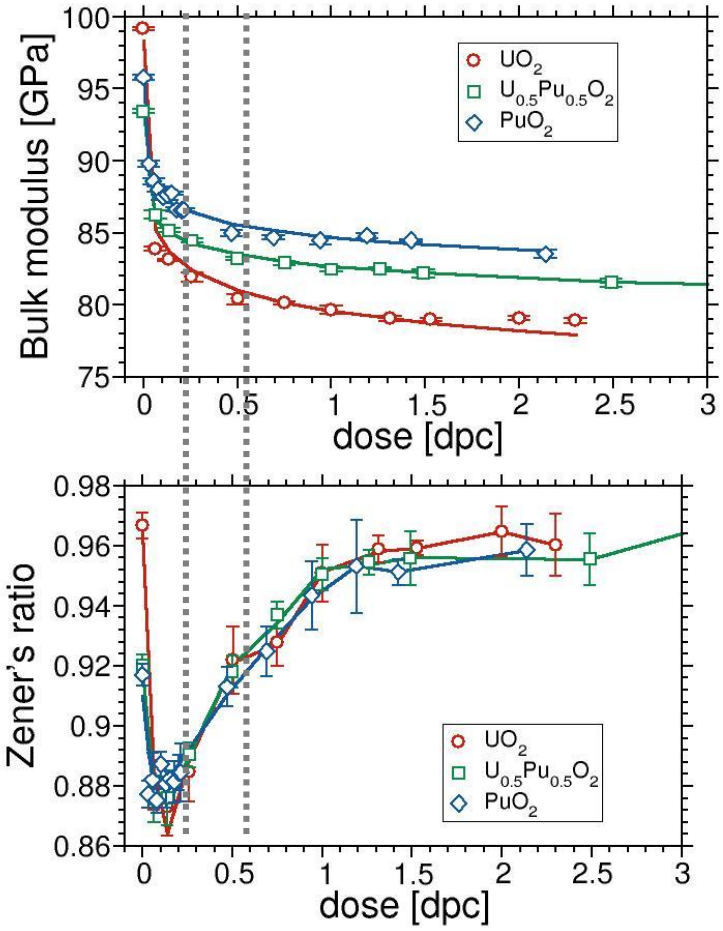
- **Output**
  Traction-separation
  Cohesive Strength: σ_c
  Energy release stresses
  $G_c \rightarrow K_{IC}$



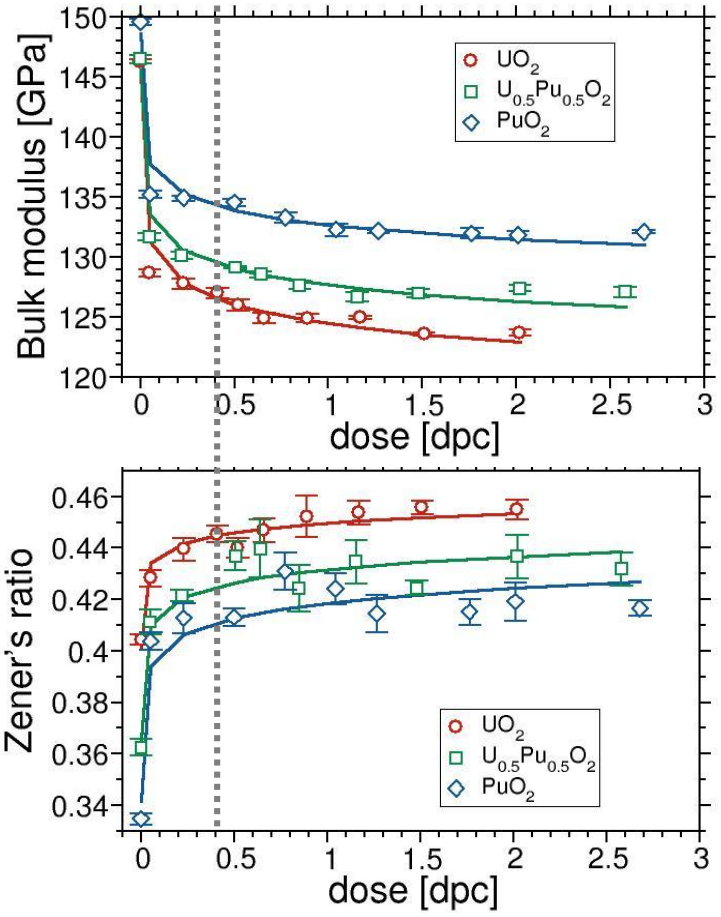
MECHANICAL PROPERTIES WITH DOSE

Elastic stiffness coefficients

MOX-07



Cooper



Point defects ⇨ Bulk modulus decrease & anisotropy changes

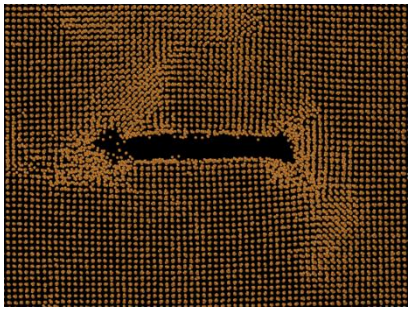
MECHANICAL PROPERTIES WITH DOSE

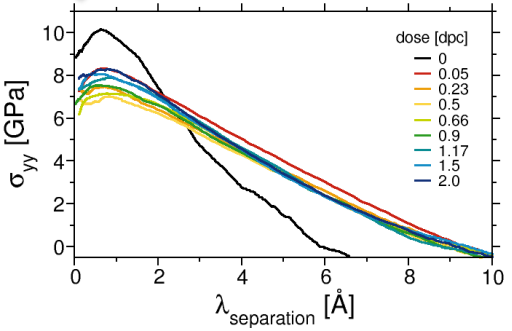
Cohesive Strength

- Both potentials have similar behavior
- Min σ_c ⇔ Max int. + Frank loops

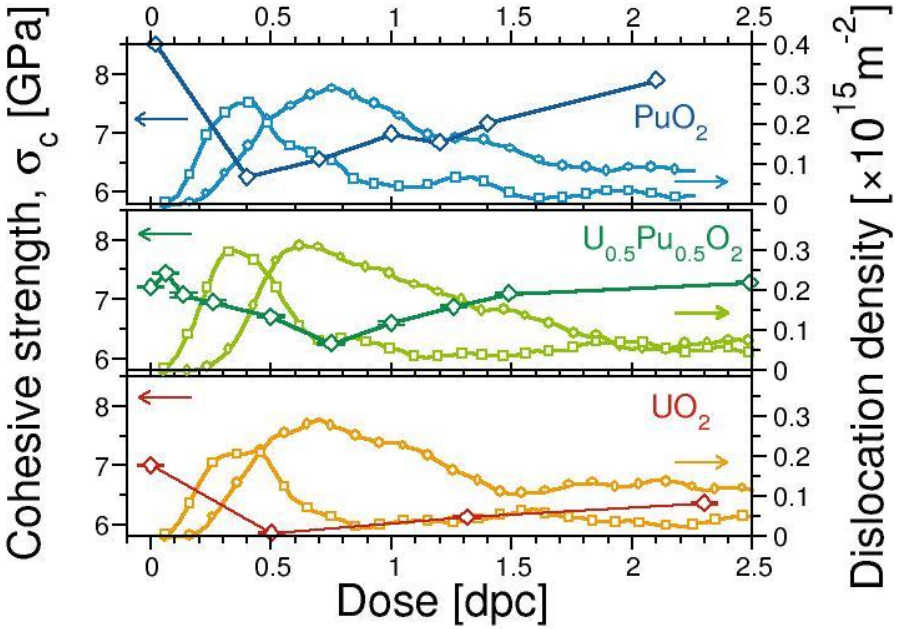
Cohesive strength related to: point defects + Frank loops

Cracking example

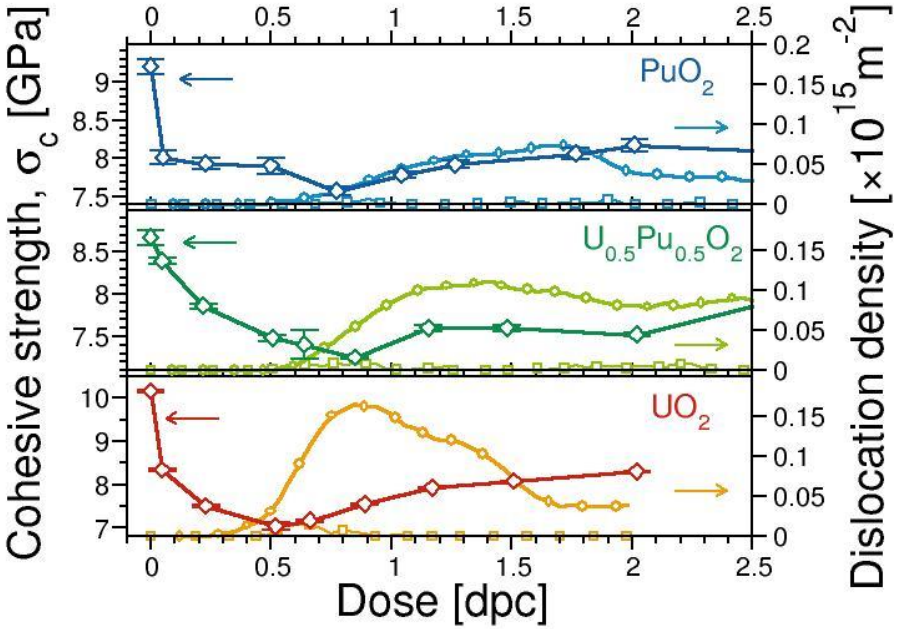




MOX-07



Cooper



(◇) cohesive strength, (○) ½<110> perfect dislocation, (□) ⅓<111> Frank loop

SUMMARY

- **Mechanical behavior assessment** of $(U,Pu)O_2$ with MD simulations
  - Two rigid ion interatomic potentials: Cooper & MOX-07
  - Difference in the elastic stiffness anisotropy
  - Variations but main behavior identical
- **Radiation structure evolution**
  - 5 stage process:

1) point defects 2) clustering 3) Frank loop 4) unfaulted loop 5) forest of dislocations

- **Mechanical properties with dose**
  - Bulk modulus ↘ correlated to point defects (swelling)
  - Cohesive strength ↘ correlated to point defects + Frank loops
- **Pu content**
  - Influence on the absolute values but not on the behavior

Ackwnolegments

Challenge project – C097073

Thank you for listening

Questions ?

Commissariat à l'énergie atomique et aux énergies alternatives
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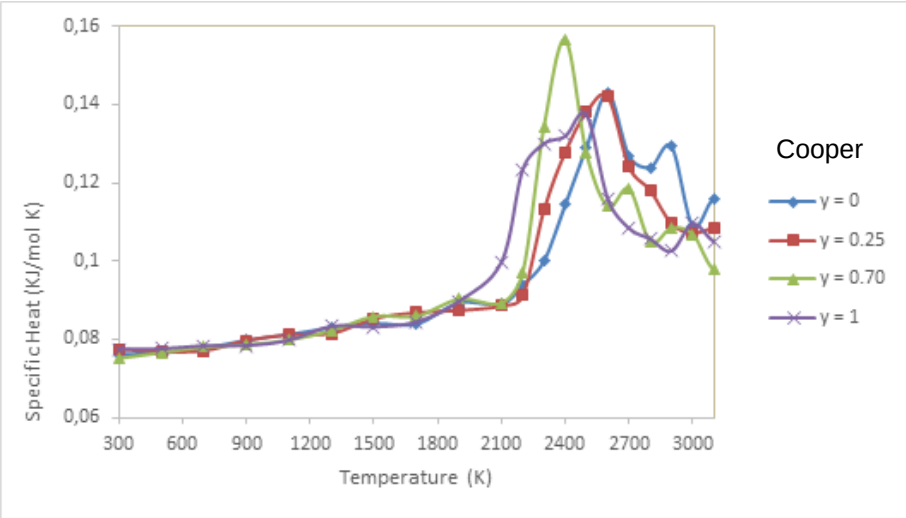
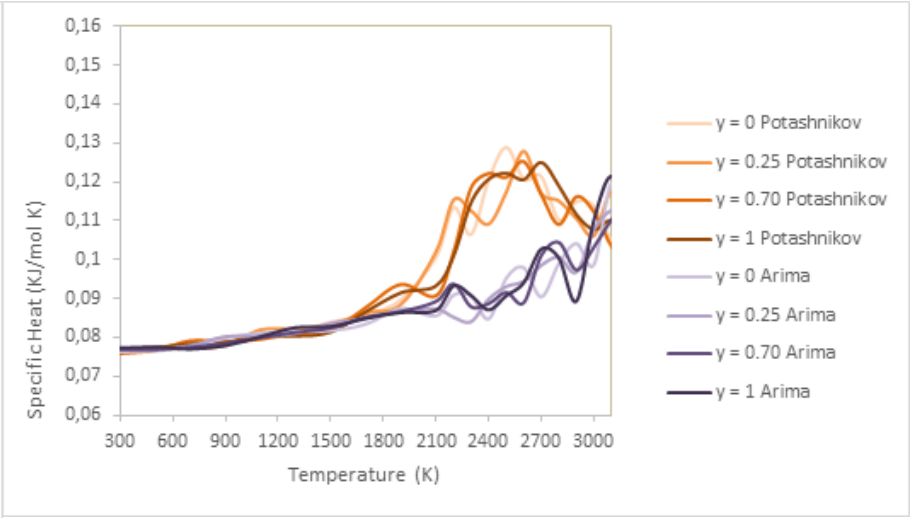
Direction de l'Energie Nucléaire
Département de Physico-Chimie
Service de la Corrosion et du Comportement des Matériaux dans leur Environnement

ASSESSMENT POTENTIAL - THERMODYNAMICS

Assessment of $U_{1-y}Pu_yO_2$: $0 < y < 1$ and $300\ K < T < T_m$

- Lattice parameter, thermal expansion fit well Fink's recommandation & Uchida → 2100 K
- Vegard's law verified → 2100 K
- Specific heat: $C_p = \frac{1}{n}\left(\frac{\partial H}{\partial T}\right)_P$



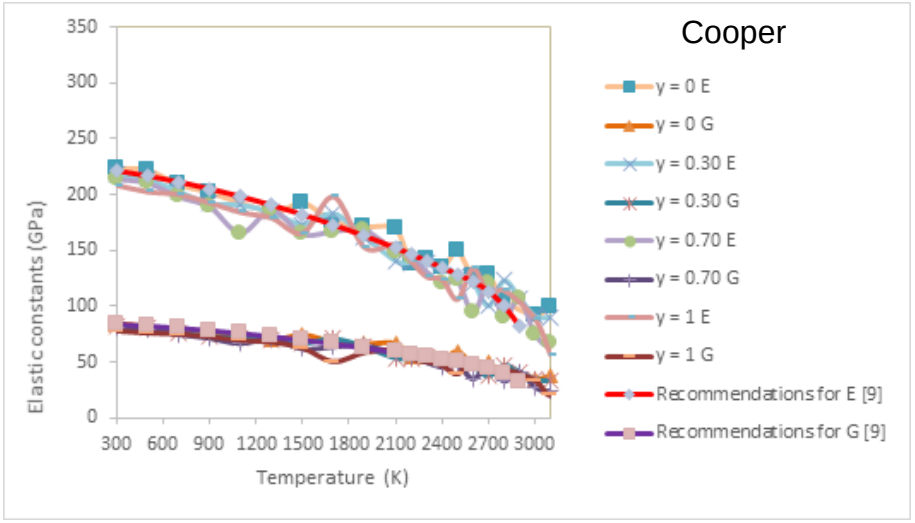


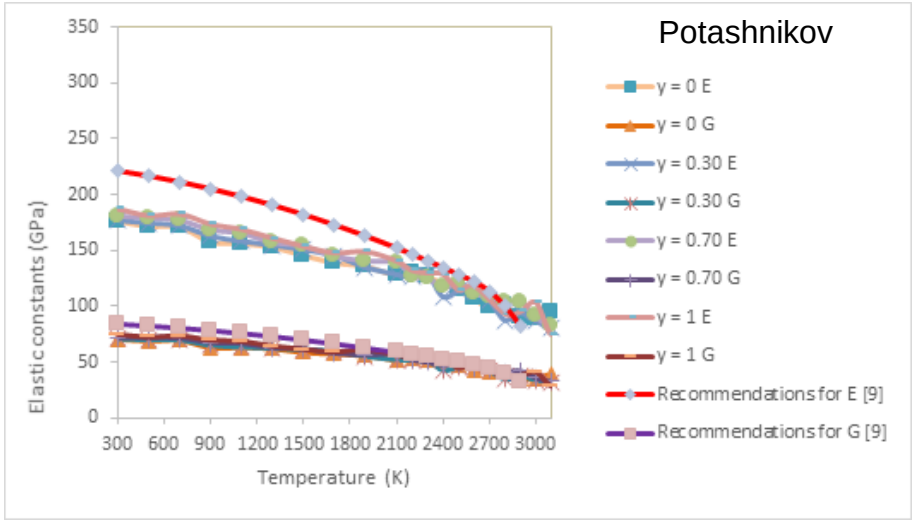
- λ-peak = superionic transition[1,2] at $T \approx 0{,}8\ T_m$
- Cooper & Potashnikov = OK, Arima no λ-peak

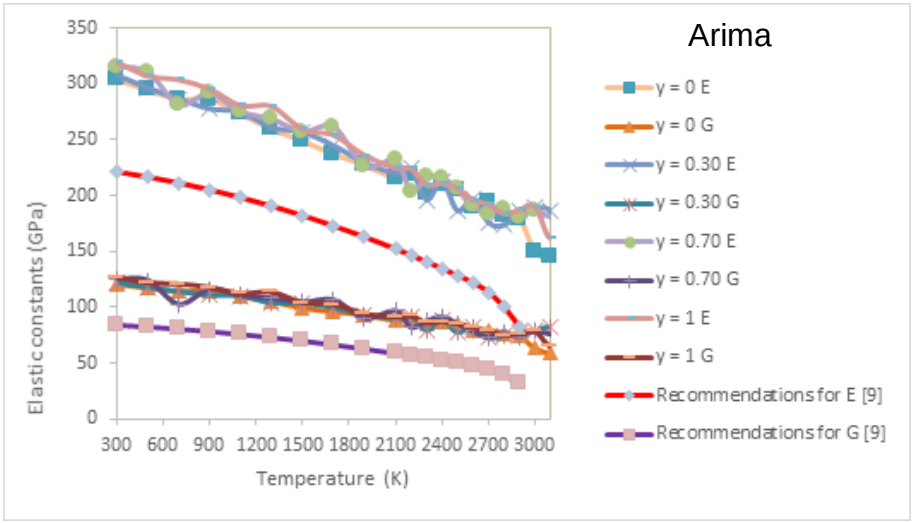
[1] J Ralph, J of Chemical Society 83, 1253 (1987)
[2] C Ronchi, J Alloys Compd, 213, 159 (1994)

ASSESSMENT POTENTIAL – MECHANICS I

Elastic stiffness coefficients





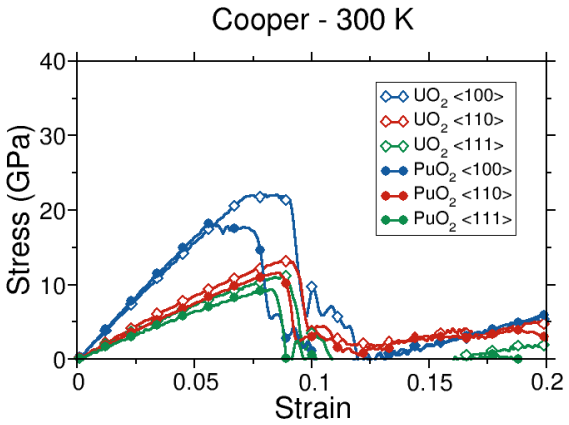


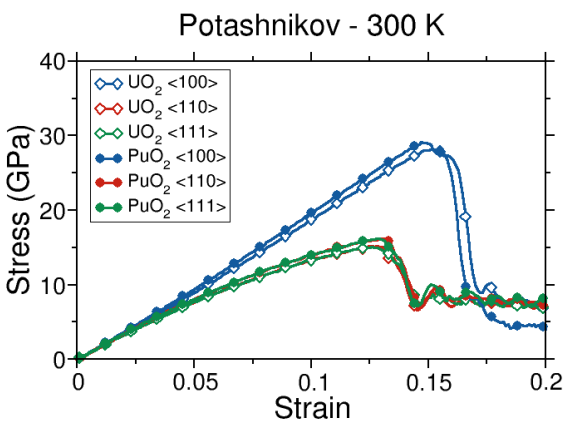
E: Young's modulus
G: shear modulus

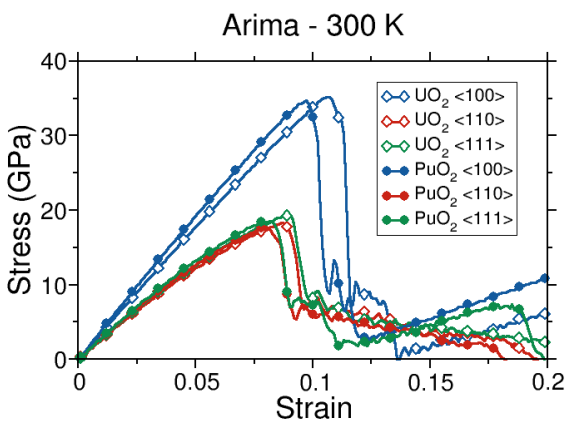
All potentials ↘ with temperature & independent to Pu content
but
Cooper > Potashnikov > Arima

Stress-Strain curves in bulk

- All potentials: σ_{yield}<100> » σ_{yield}<111> & σ_{yield}<110> ⇨ agreement with experiments
- For <111> & <110> few influence of the Pu content





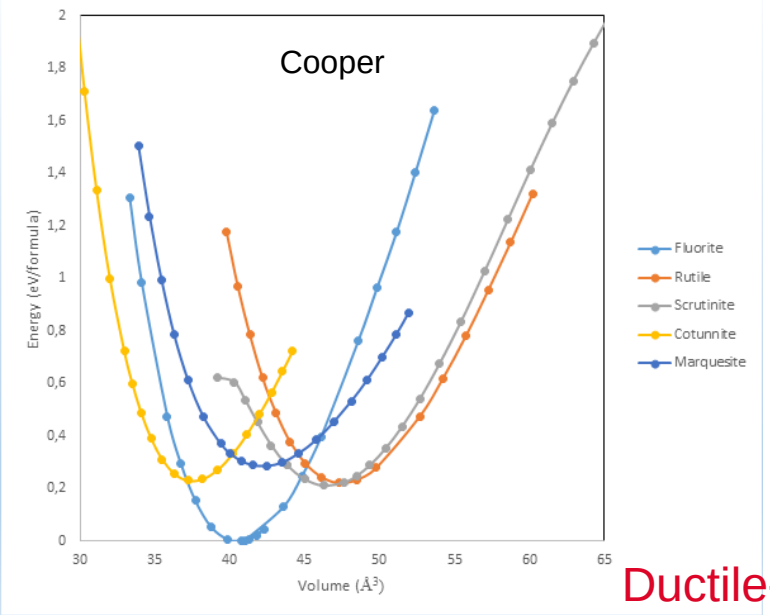


		Cooper		Potashnikov		Arima	
		σ_{yield} (GPa)	$\varepsilon_{fracture}$ (%)	σ_{yield} (GPa)	$\varepsilon_{fracture}$ (%)	σ_{yield} (GPa)	$\varepsilon_{fracture}$ (%)
<100>	UO_2	22.0	7.6	28.1	15.2	35.1	10.6
	PuO_2	18.5	5.6	29.0	14.8	34.2	9.6
<110>	UO_2	13.2	9.0	15.1	13.0	18.0	8.8
	PuO_2	11.6	8.5	16.1	13.1	17.8	8.0
<111>	UO_2	10.9	8.8	15.0	12.7	19.3	9.0
	PuO_2	9.3	8.3	16.1	12.9	18.4	8.3

σ_{yield}: Arima > Potashnikov > Cooper

$\varepsilon_{fracture}$: Potashnikov > Cooper ≥ Arima

ASSESSMENT POTENTIAL – MECHANICS III

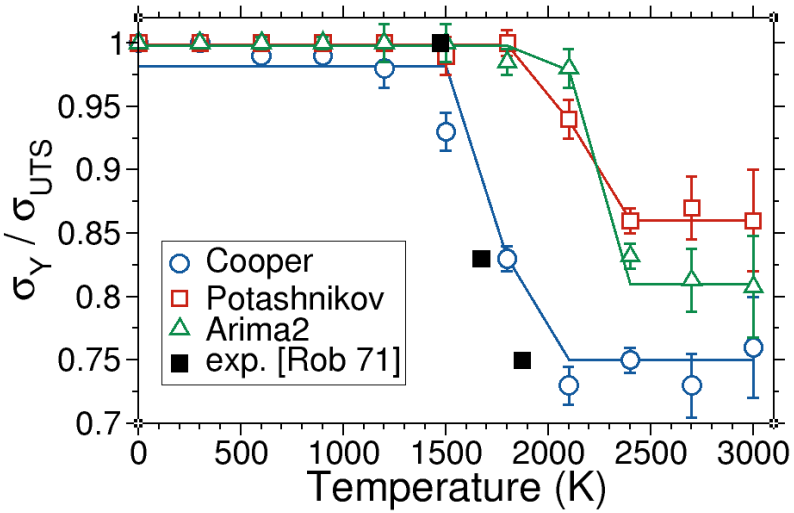


Transition pressure	Fluorite to			
	Cotunnite (GPa)	Rutile (GPa)	Scrutinite (GPa)	Marquesite (GPa)
Cooper	2.95	-6.84	-8.02	-15.99
Potashnikov	16.03	-10.57	-12.08	-
Arima	44.34	-8.30	-9.61	-

Cooper shows lower transition pressures than the other potentials

Ductile-brittle transition temperature

MOX, PuO2 20% wt



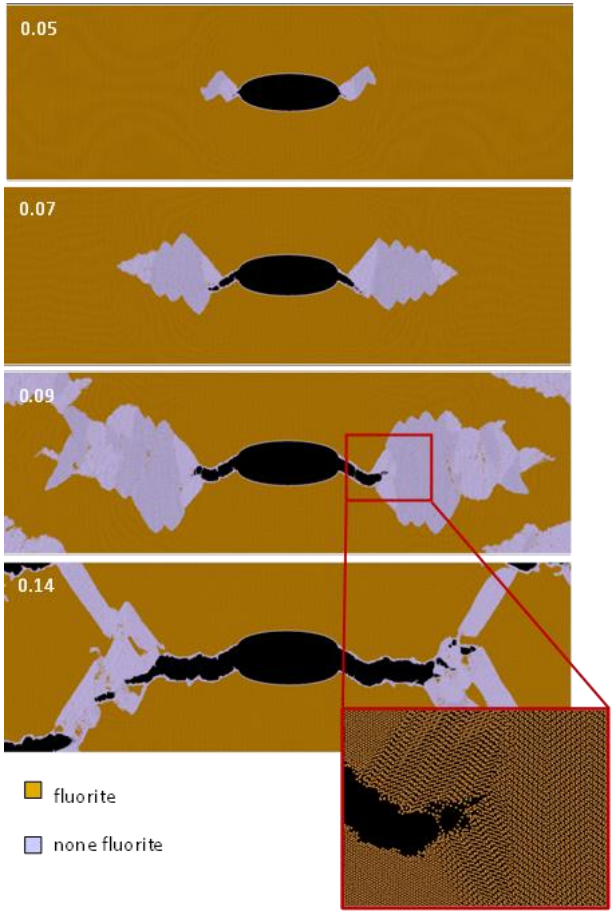
Cooper potential shows a behaviour closer to experiments

ASSESSMENT POTENTIAL – MECHANICS IV

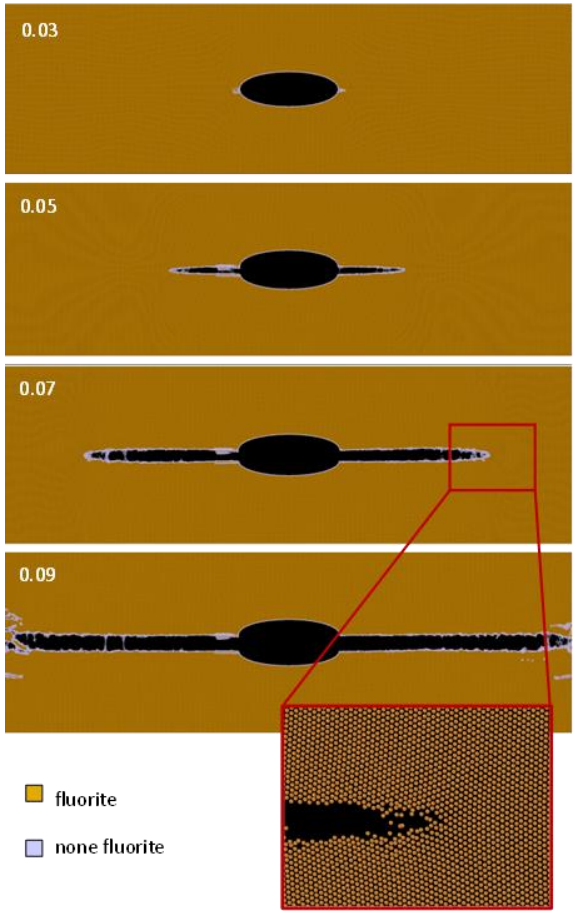
- Crack propagation – 15% Pu, <111>, 300 K
- The system included 4.4×106 atoms with box size equal to 240×70×4 nm

Strain

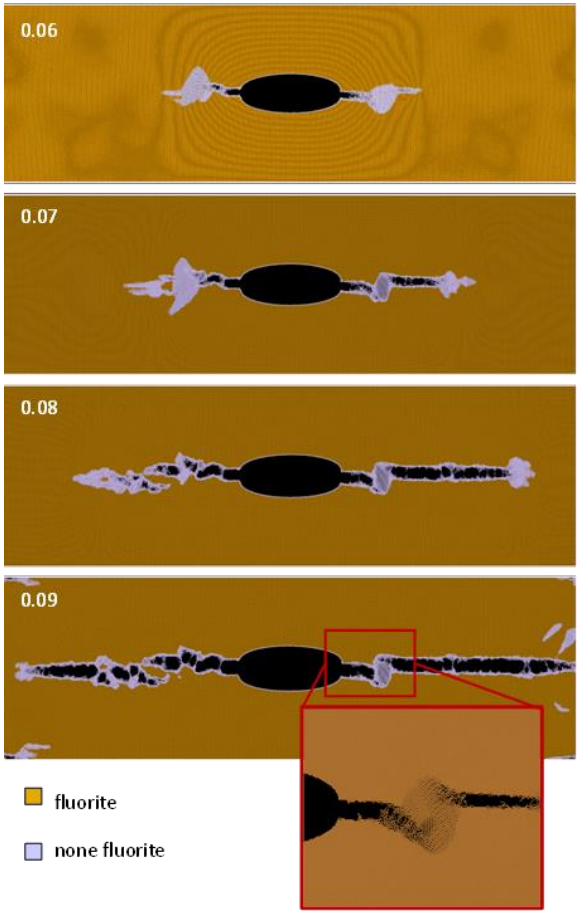
Cooper



MOX-07



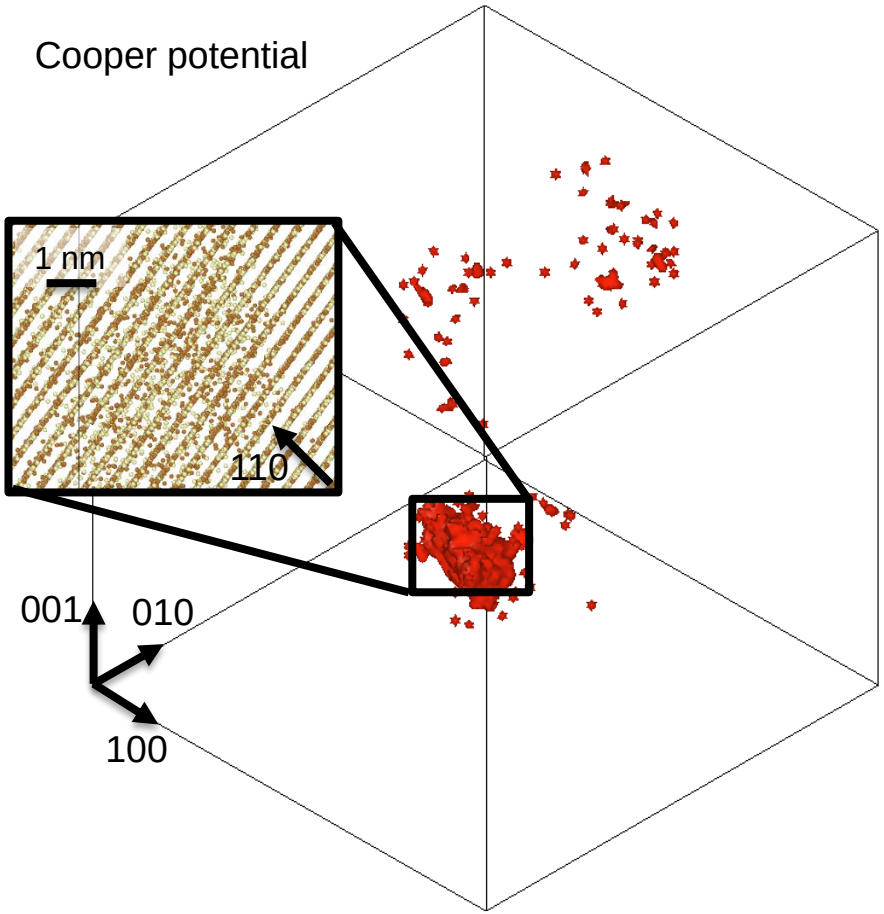
Arima

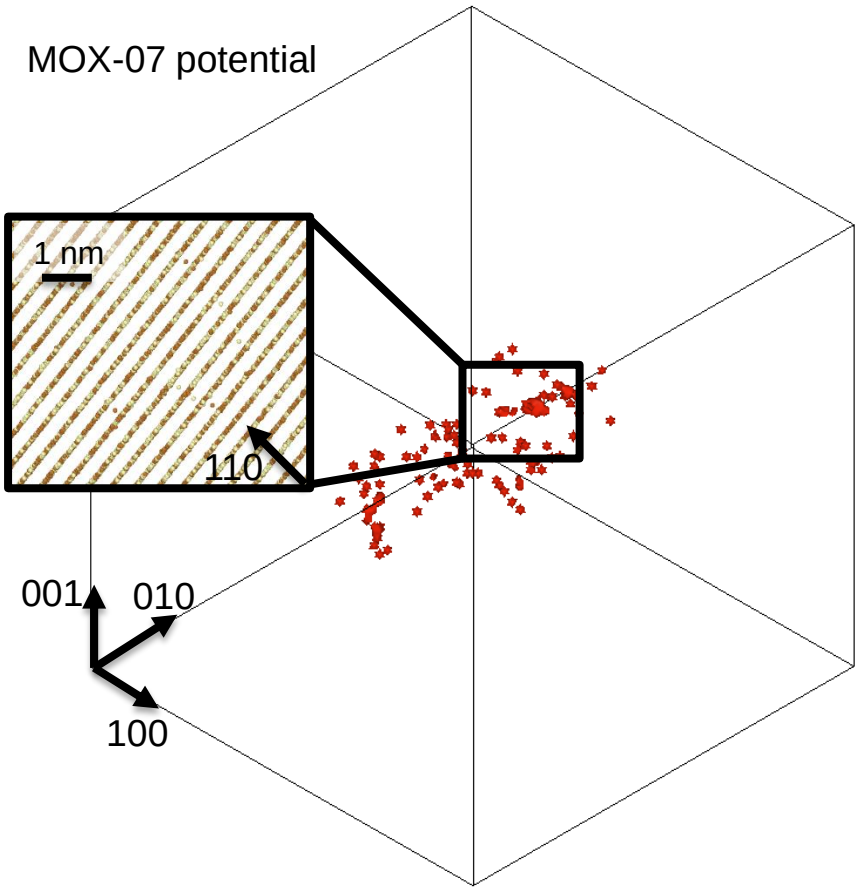


- Cooper & Arima ⇨ "plastic-like" ; MPX-07 ⇨ brittle behaviour

DISPLACEMENT CASCADES

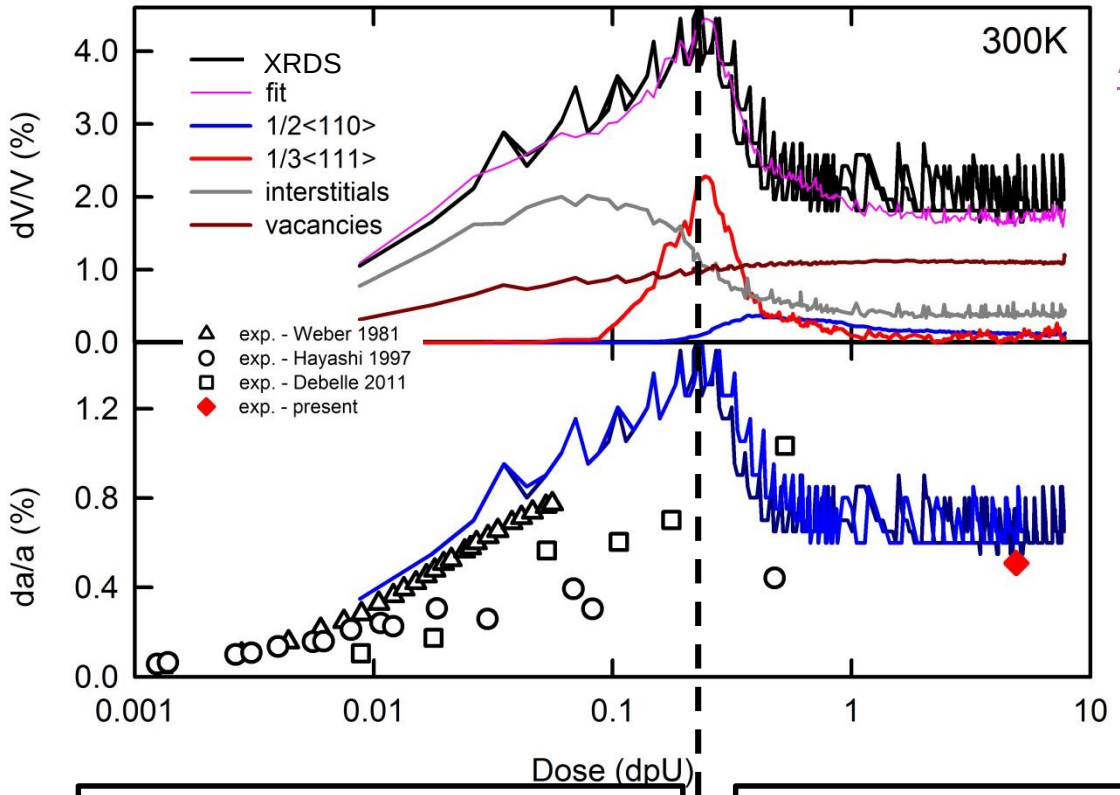
Damage created by 75 keV displacement cascade in $U_{0.5}Pu_{0.5}O_2$





LATTICE EVOLUTION WITH IRRADIATION DOSE

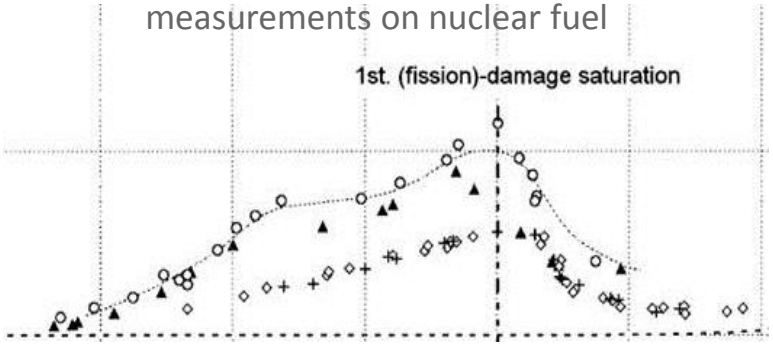
Deconvolution of the Lattice evolution under irradiation



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

with $V_I^f = 5.6\ \text{Å}^3$; $\overline{R_{loop}} \sim 3\ \text{nm}$; $V_{rel}^I = V_{rel}^V = 1.\Omega$

Lattice expansion
contraction comparable to measurements on nuclear fuel



Swelling:

- **interstitials** and **vacancies**
- 1/3<111> **Frank loops**

Contraction:

- transformation of **Frank loops** into perfect loops
- **vacancies** at steady state

Chartier *et al.*, Appl. Phys. Lett. 109 (2016) 181902