The European Commission’s science and knowledge service

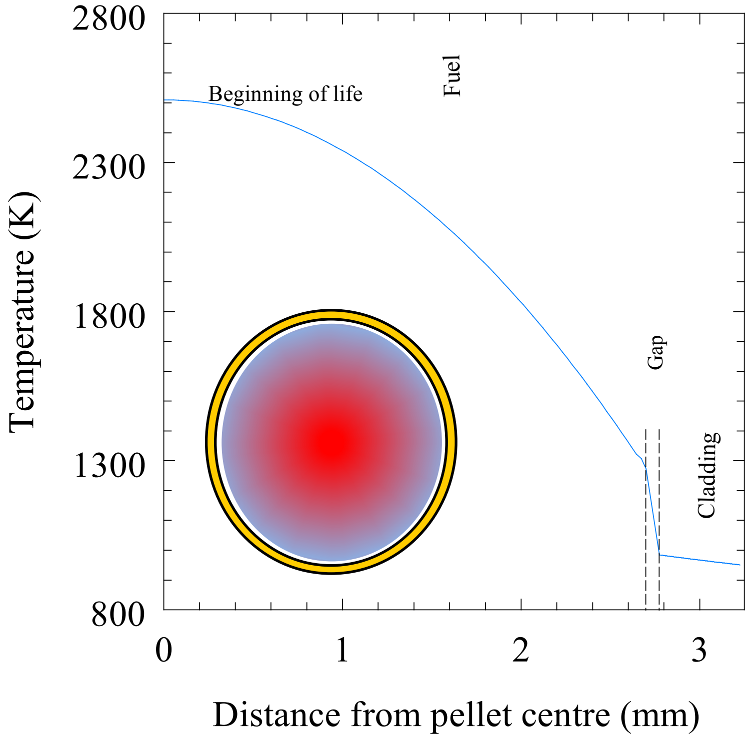
Joint Research Centre

Fuel chemistry and thermodynamic aspects under irradiation

Rudy Konings

European Commission

Thermodynamics and nuclear fuel



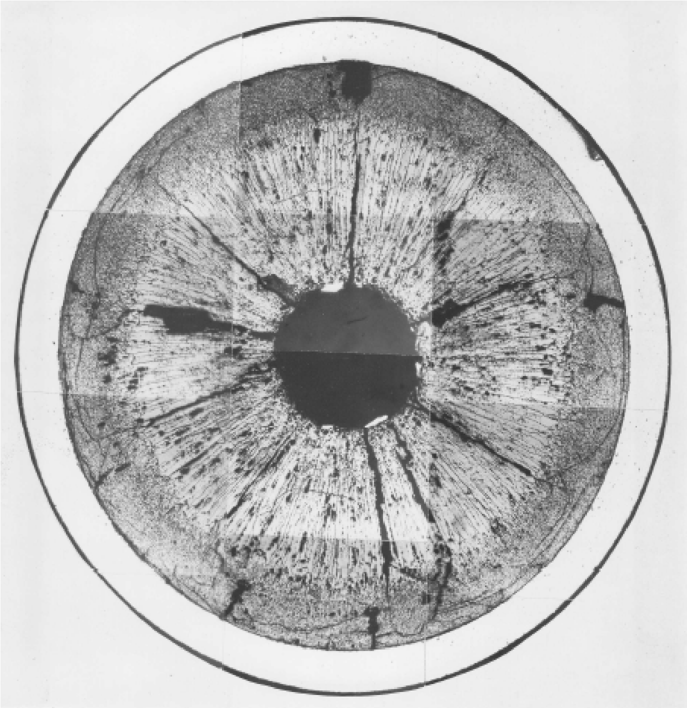
$$\Delta T(r) = T(R) - T(r) = \frac{\chi}{4\pi\lambda R^2}(R^2 - r^2)$$

χ = linear heating rate (W cm^{-1})

R = pellet radius (cm)

λ = Thermal conductivity (W cm^{-1} K^{-1})

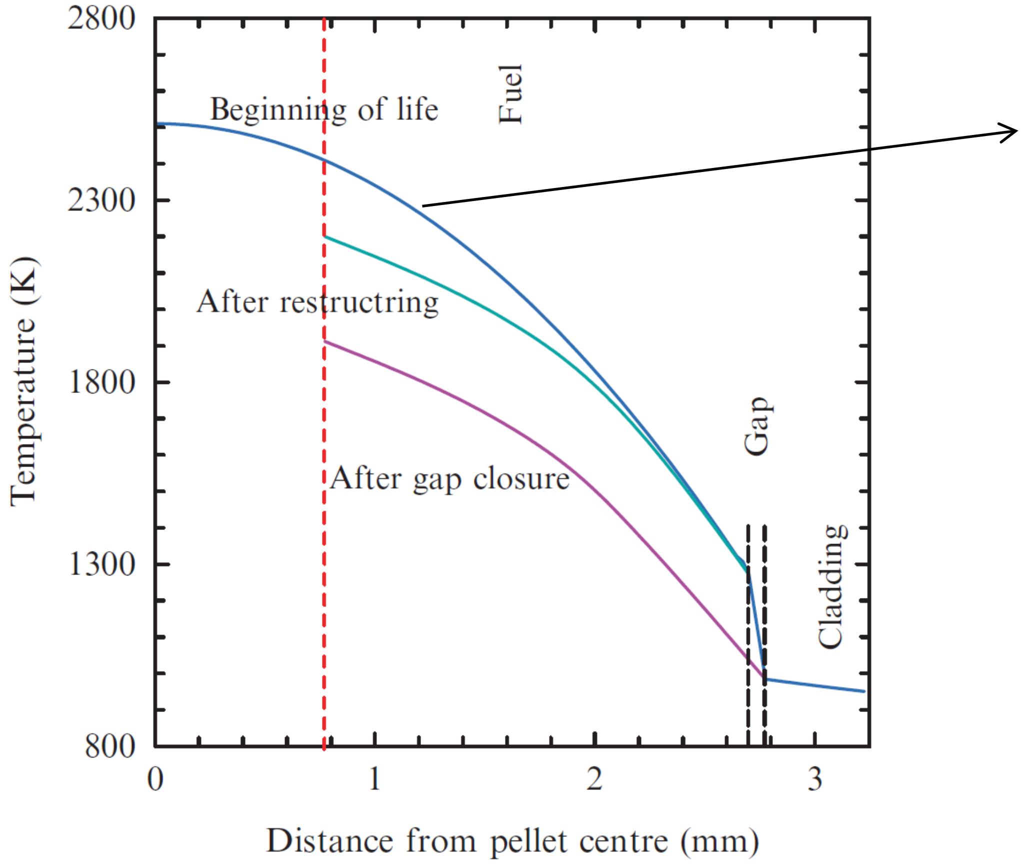
Thermodynamic equilibrium?



- Restructuring takes place during the fist hours
- Massive matter transport
- Fission changes the composition
- Radiation

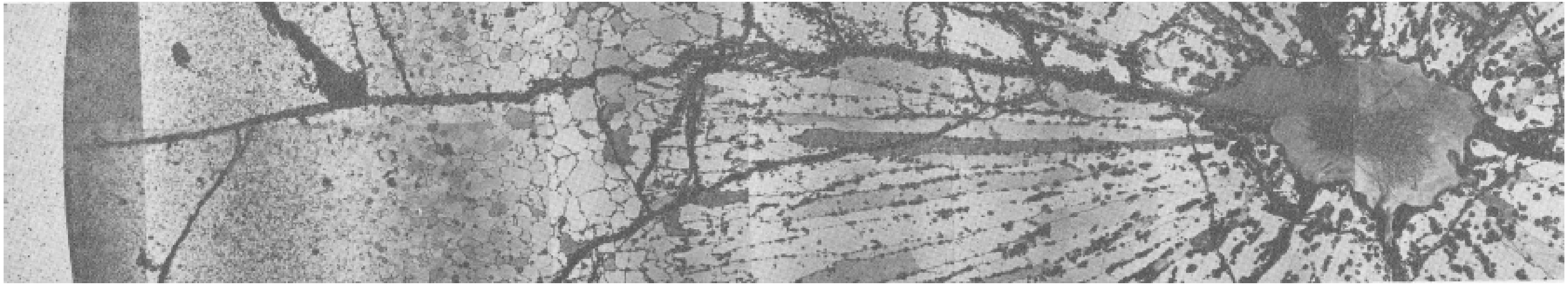
European Commission

Thermodynamics and nuclear fuel



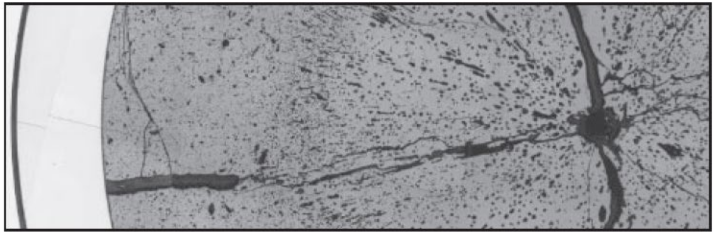
Gradient: 0.7 K/µm

Irradiated fast reactor fuels: restructuring

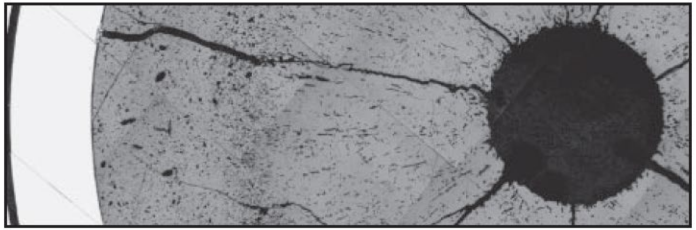


Cross section of a fast reactor mixed oxide fuel from the DS1 experiment

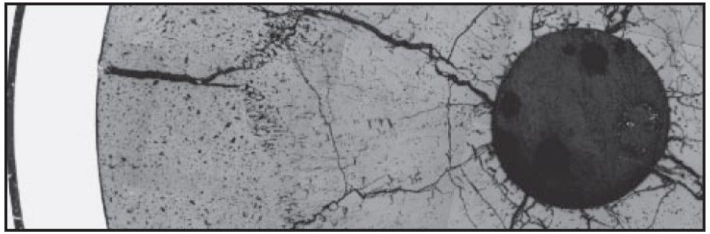
Irradiated fast reactor fuels: restructuring



(a) $(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_{1.98}$ irradiated for 10 min at 427 W/cm



(b) $(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_{1.98}$ irradiated for 24 h at 432 W/cm



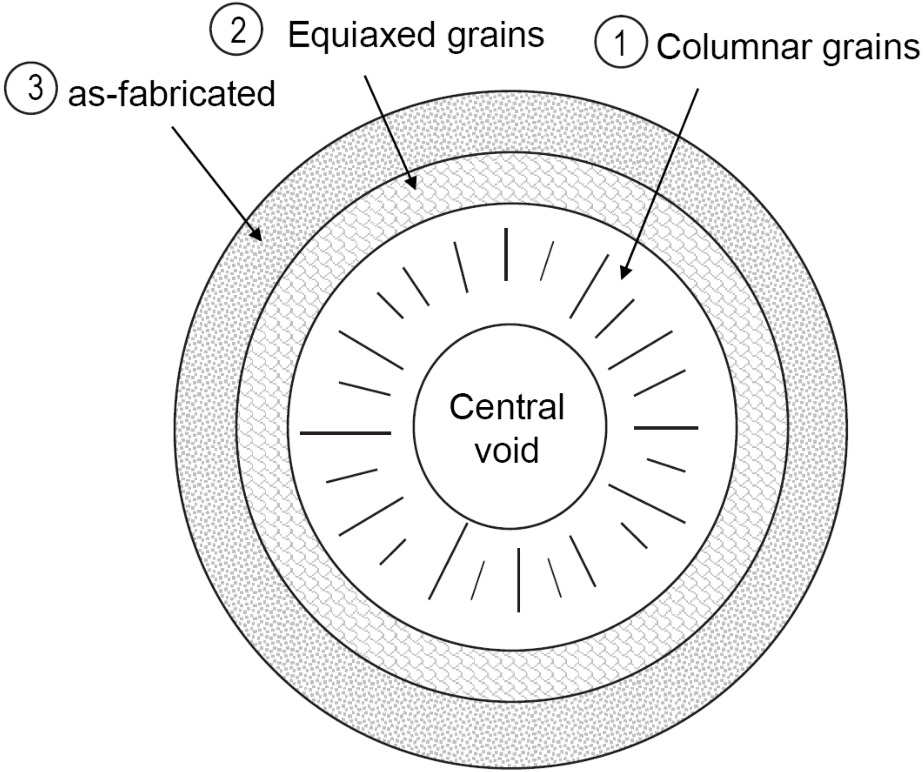
(c) $(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_{1.96}$ irradiated for 24 h at 429 W/cm

Short irradiation tests of minor actinide fuels performed in the JOYO reactor in Japan

Two different O/M ratios

European Commission

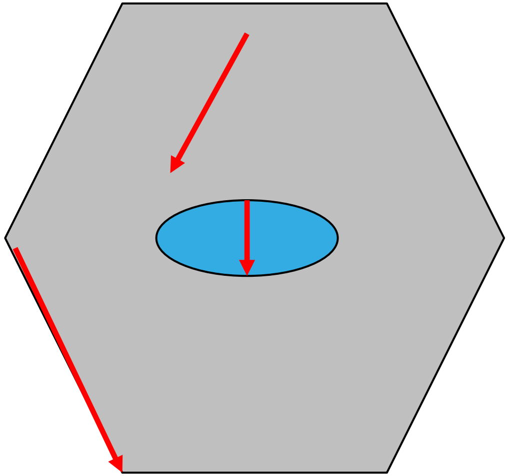
Irradiated fast reactor fuels: restructuring



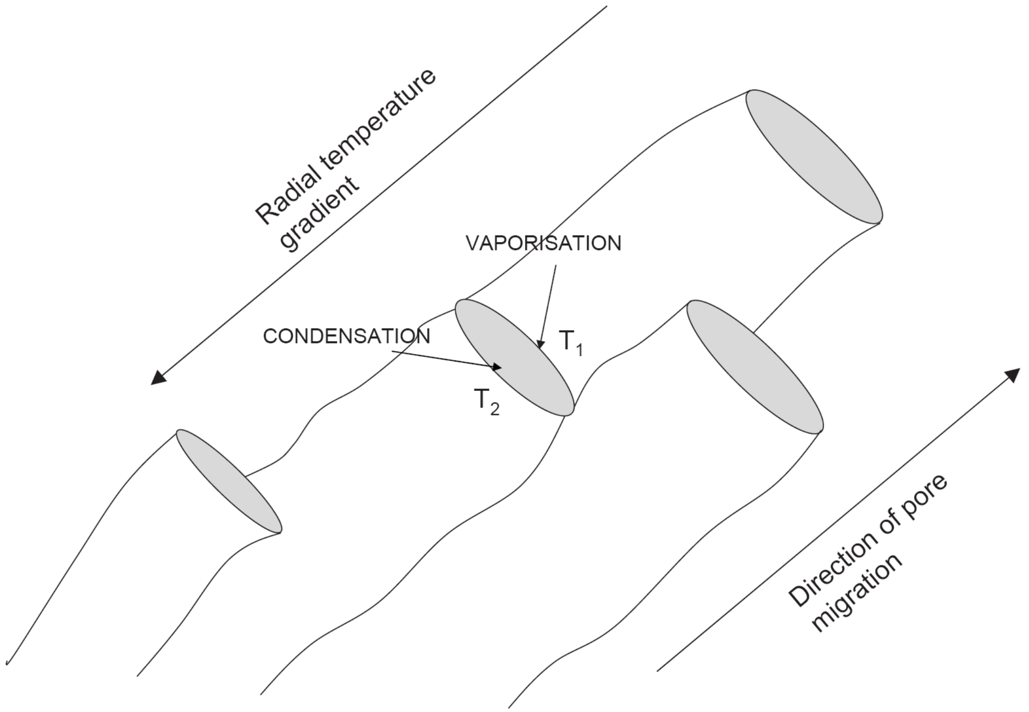
Irradiated fast reactor fuels: restructuring

Several transport processes take place

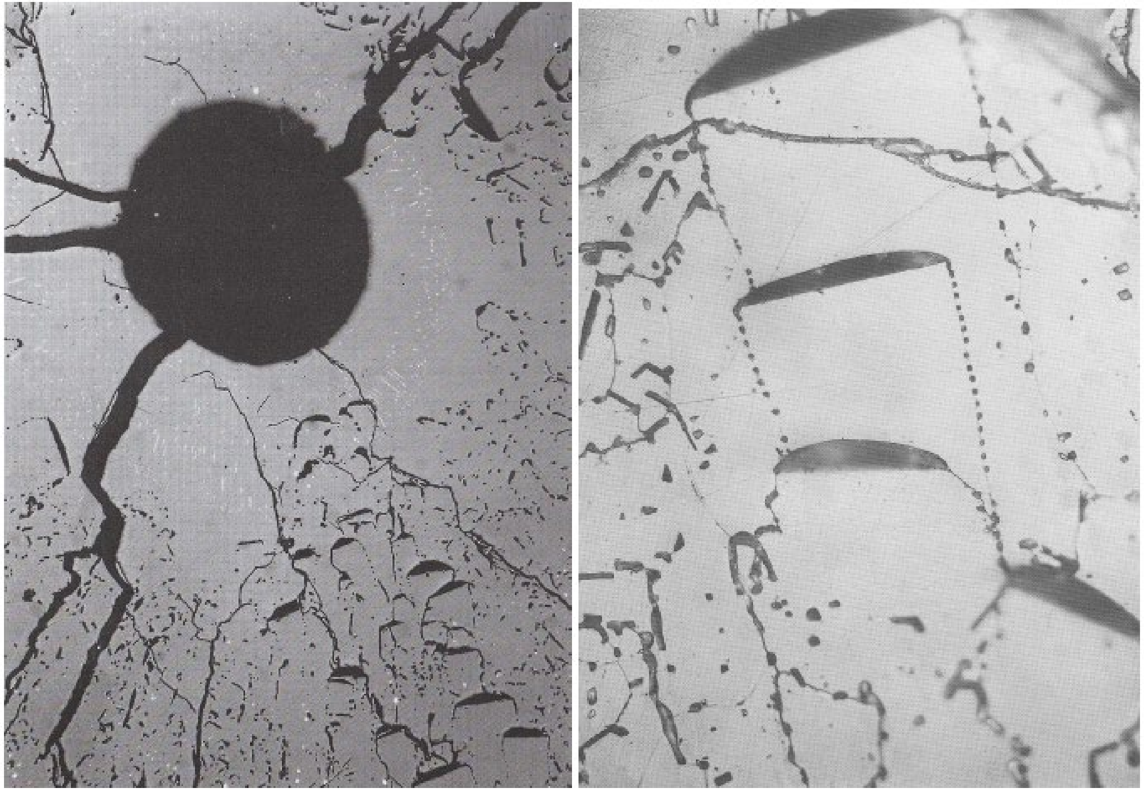
- Vaporisation-condensation
- Matrix diffusion of oxygen and metal
- Grain boundary diffusion



Irradiated fast reactor fuels: restructuring

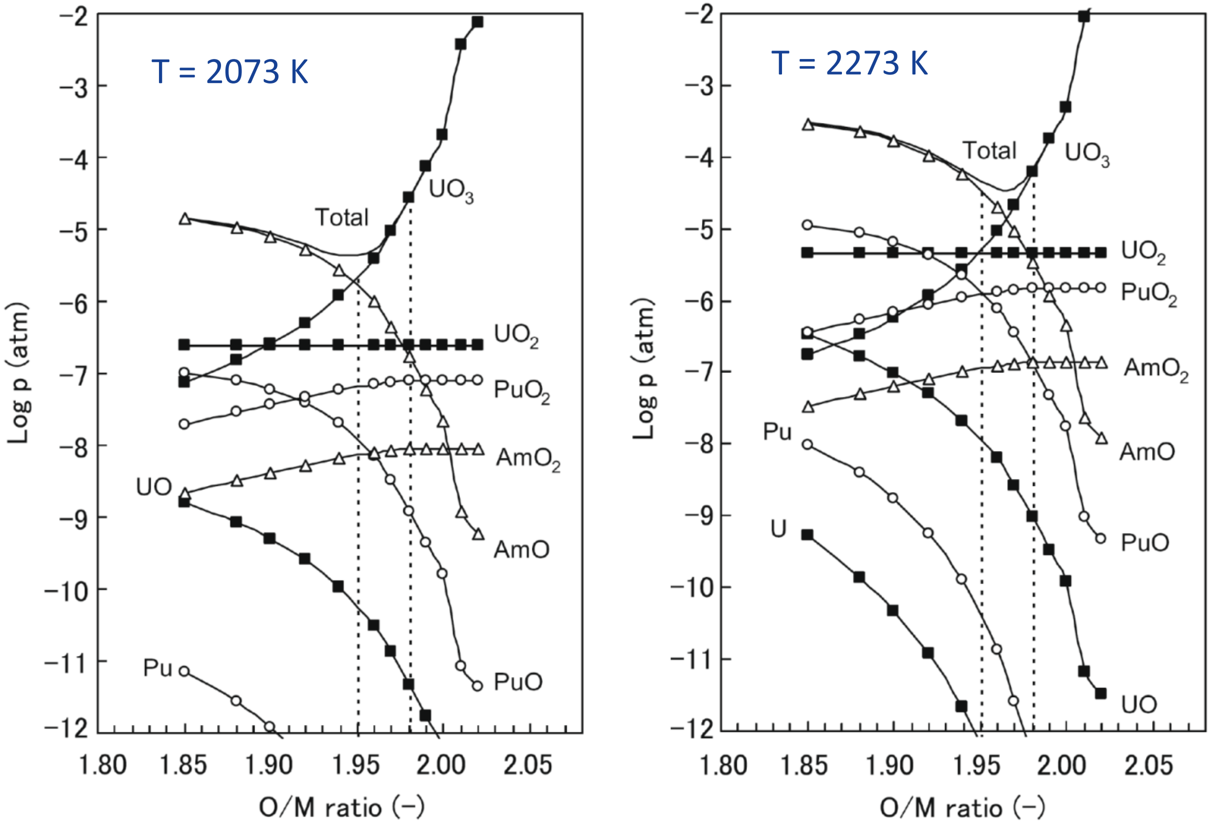


Irradiated fast reactor fuels: restructuring



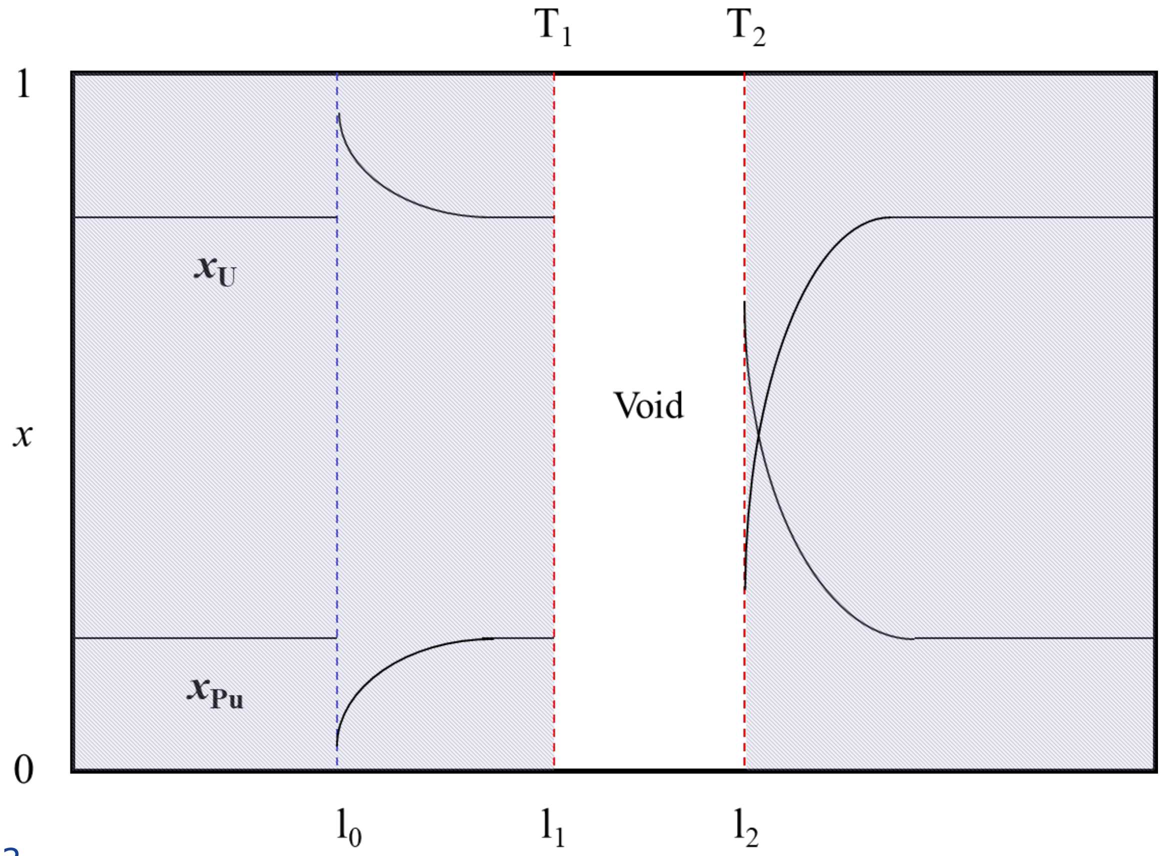
European Commission

Irradiated fast reactor fuels: restructuring



Maeda et al., J. Nucl. Mater. 2009, 389:78

Irradiated fast reactor fuels: restructuring



European Commission

Irradiated fast reactor fuels: restructuring

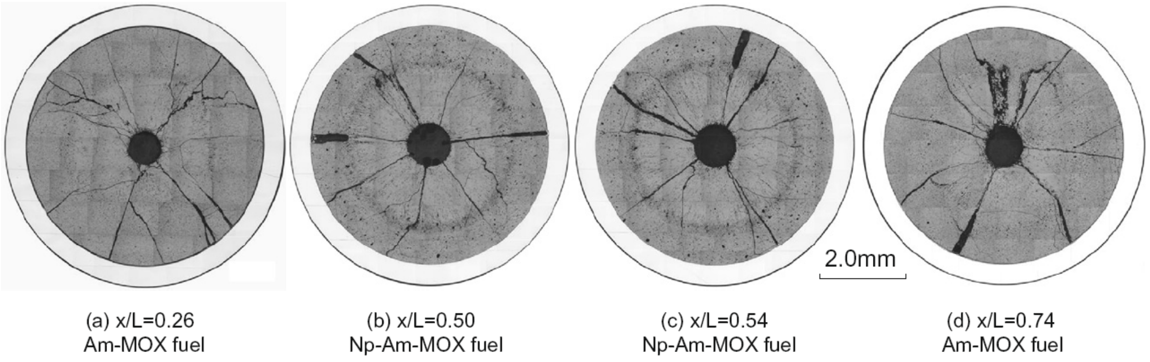


Fig. 3a. Ceramographs of specimens sectioned from the fuel pin of O/M ratio 1.98 irradiated in the Am1-2 experiment.

O/M = 1.98

Am-MOX: 0.5 % Am

Np-Am-MOX: 2% Am+2% Np

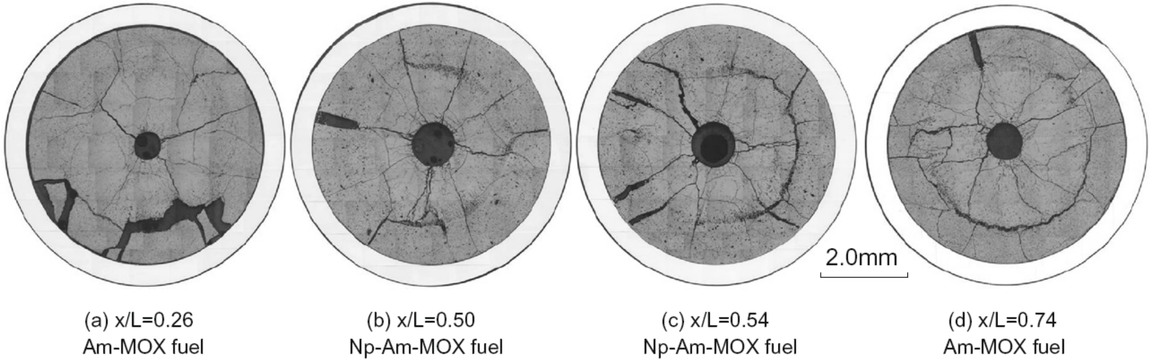


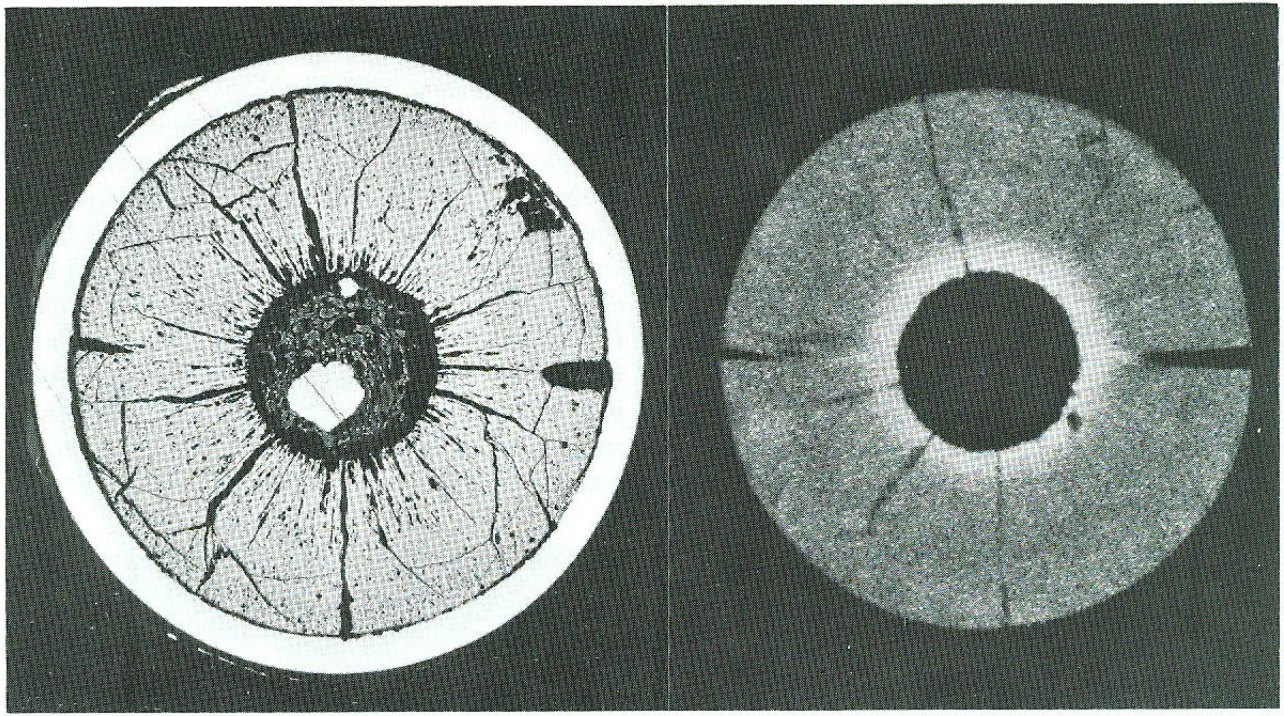
Fig. 3b. Ceramographs of specimens sectioned from the fuel pin of O/M ratio 1.95 irradiated in the Am1-2 experiment.

O/M = 1.95

European Commission

Irradiated fast reactor fuels: restructuring

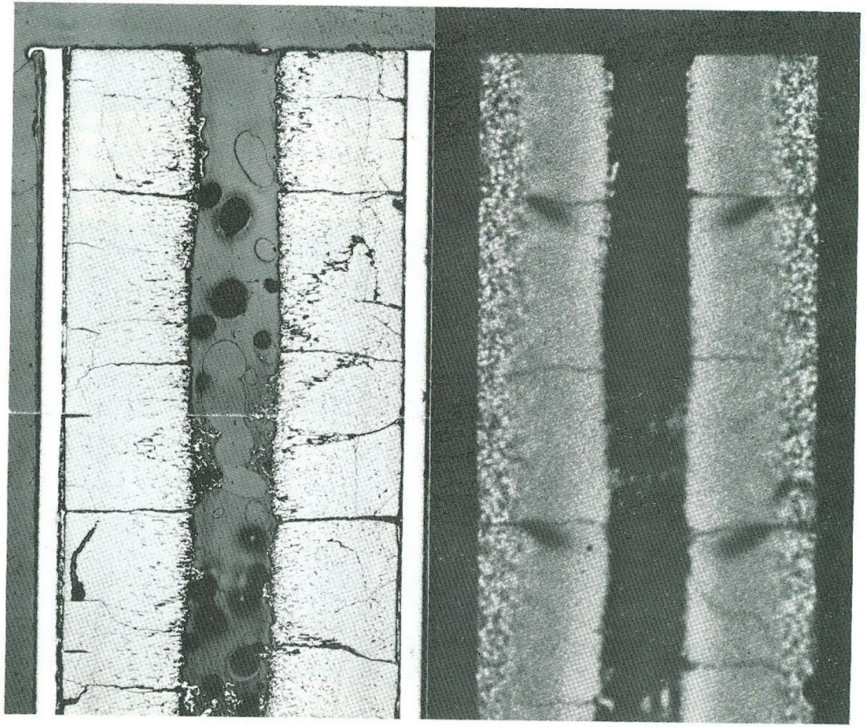
Alpha radiography



O'Boyle et al, 1969

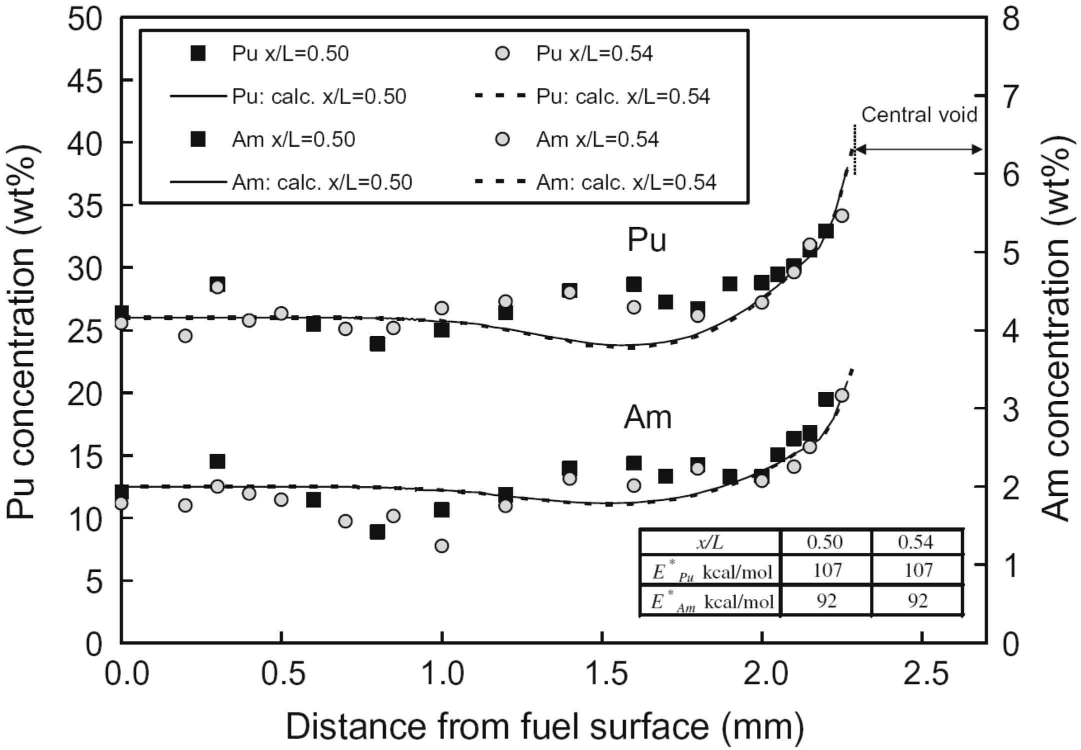
Irradiated fast reactor fuels: restructuring

Alpha radiography



O'Boyle et al, 1969

Irradiated fast reactor fuels: restructuring



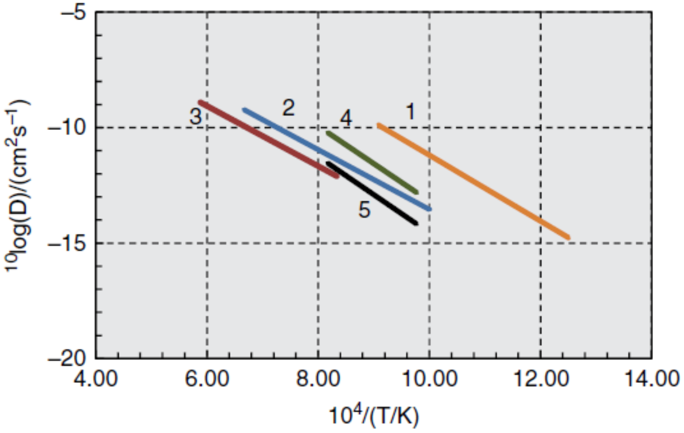
Maeda et al, JNM, 389:78

- Results from the AIM1 irradiation in JOYO

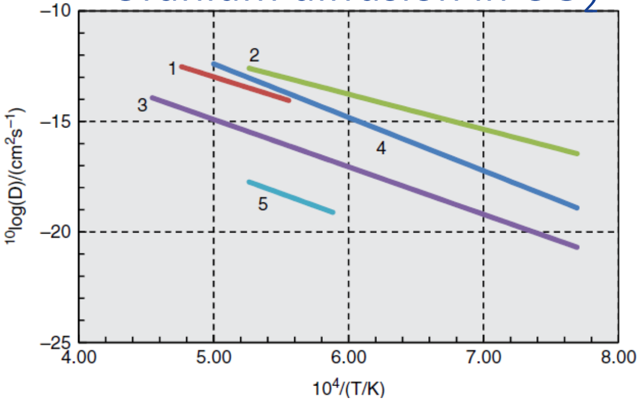
European Commission

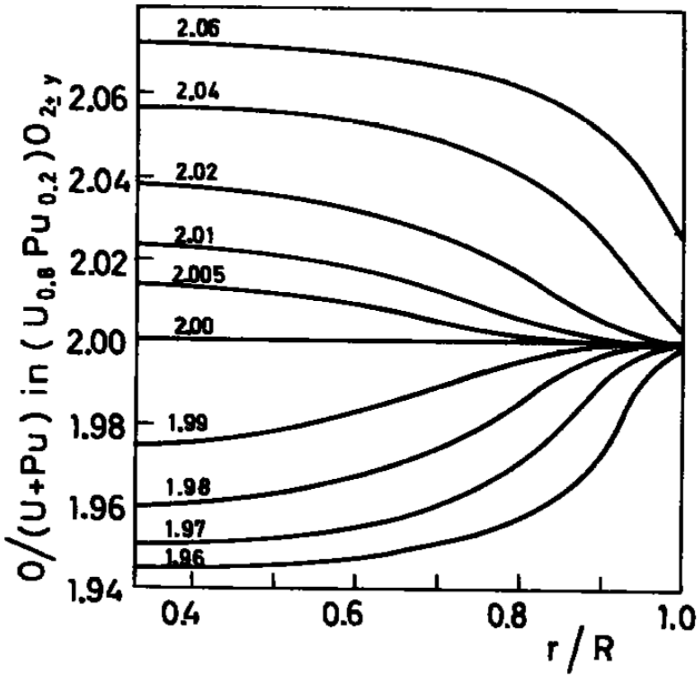
Irradiated fast reactor fuels: restructuring

Oxygen diffusion in UO_2



Uranium diffusion in UO_2

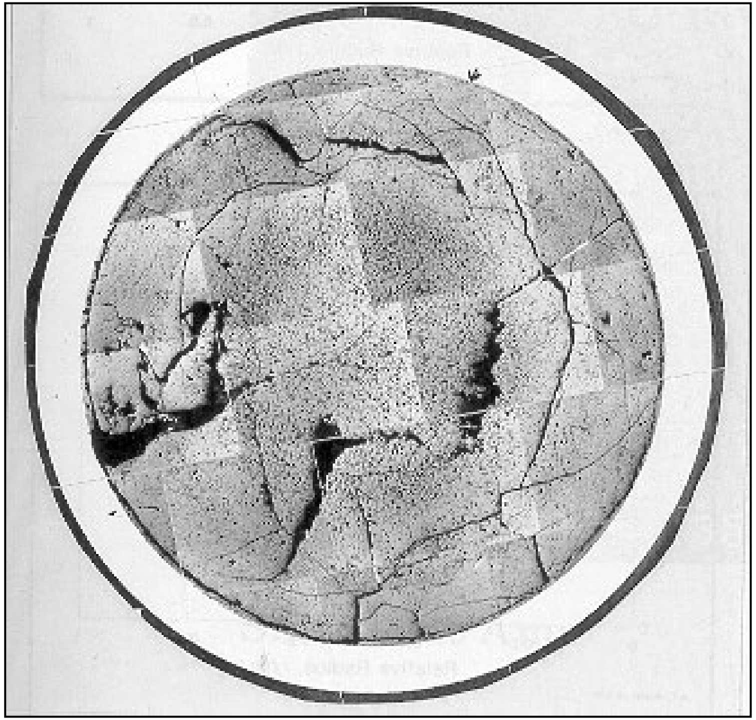




Sari anmd Schumacher, JNM, 41:192

European Commission

Irradiated fast reactor fuels: restructuring

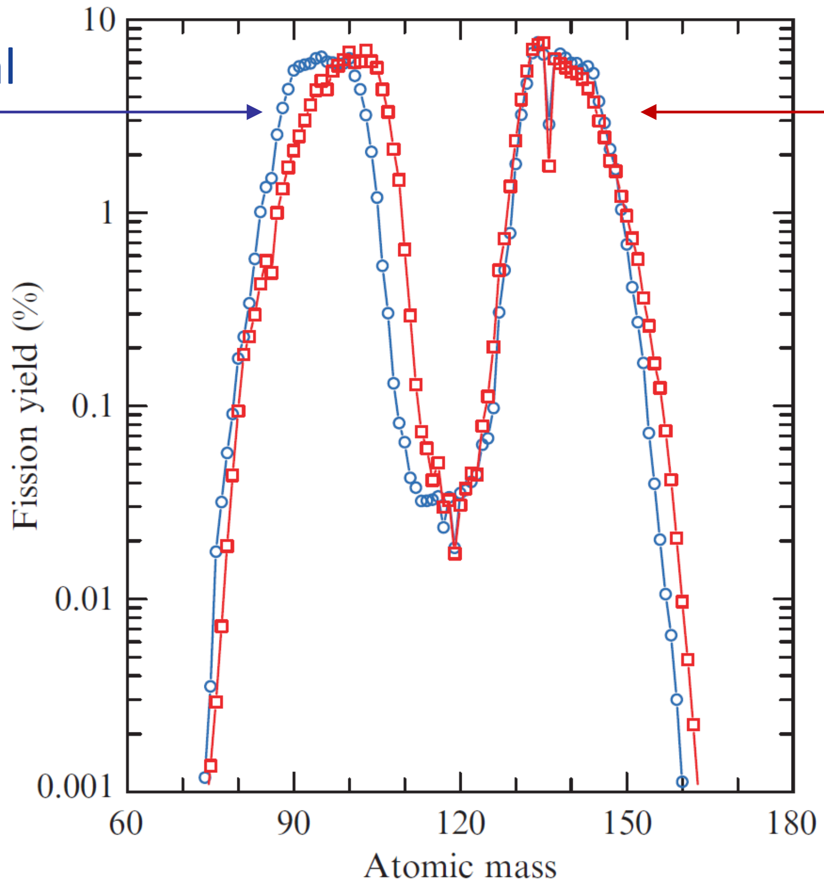


SUPERFACT $(U_{0.6}Np_{0.2}Am_{0.2})O_2$ fuel

- Actinide burnout 31% (actinide fission not known)
- Swelling (axial expansion 2.3%, radial expansion 3.3%)
- High helium production & release (high porosity)

Fission product behaviour

^{235}U in thermal spectrum



^{239}Pu in fast spectrum

European Commission

Fission product behaviour

1) Dissolved in the matrix: Rb, Sr, Y, Zr, Nb, Te, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu

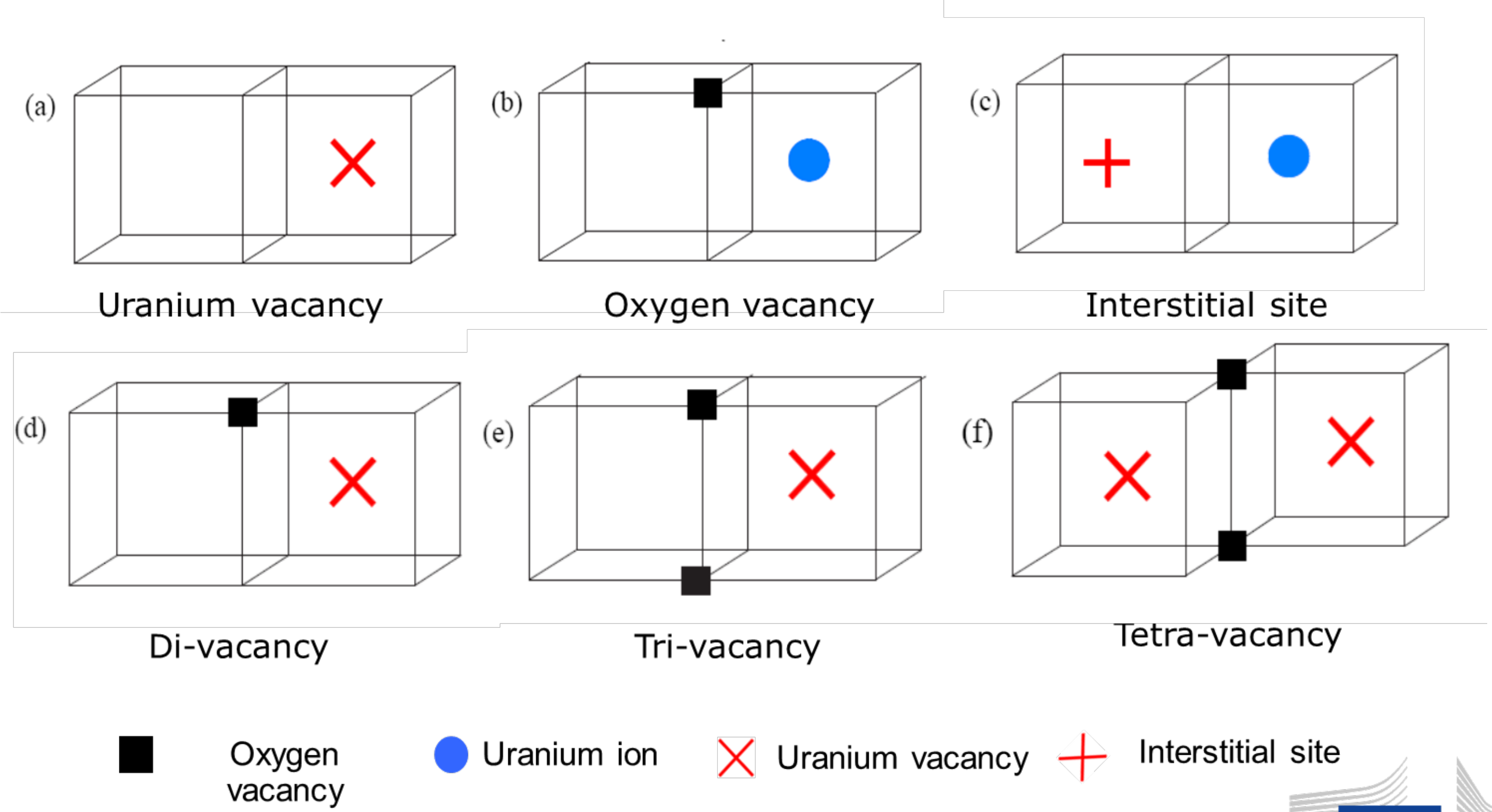
2) Oxide precipitates at grain boundaries: Rb, Sr, Zr, Nb, Mo, Se, Te, Cs, Ba

3) Metallic precipitates: Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Se, Te

4) Volatiles: Br, Rb, I, Cs, Te

5) Gases: Kr, Xe

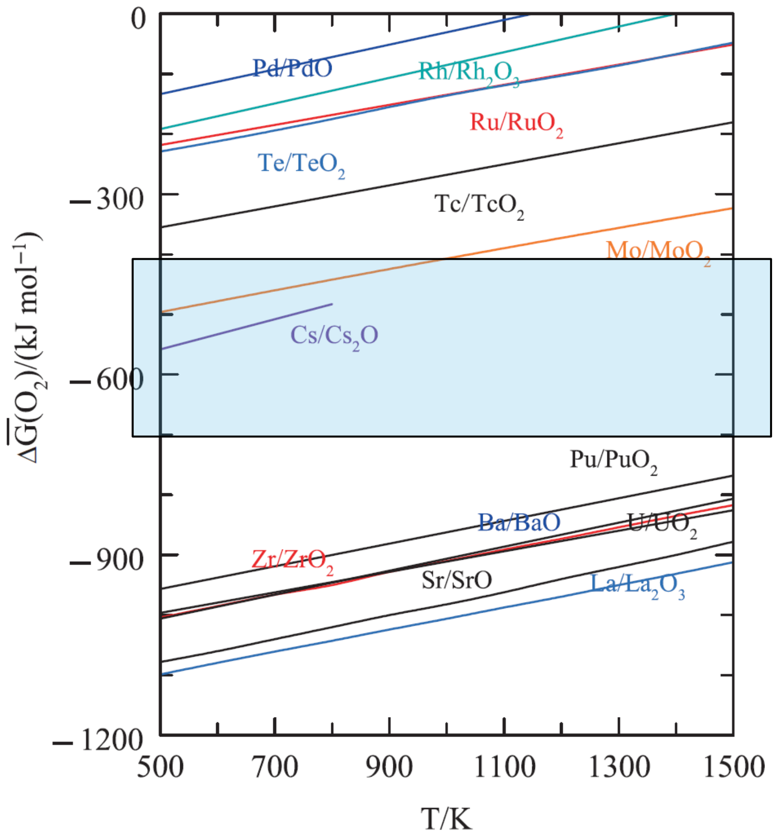
Fission product behaviour

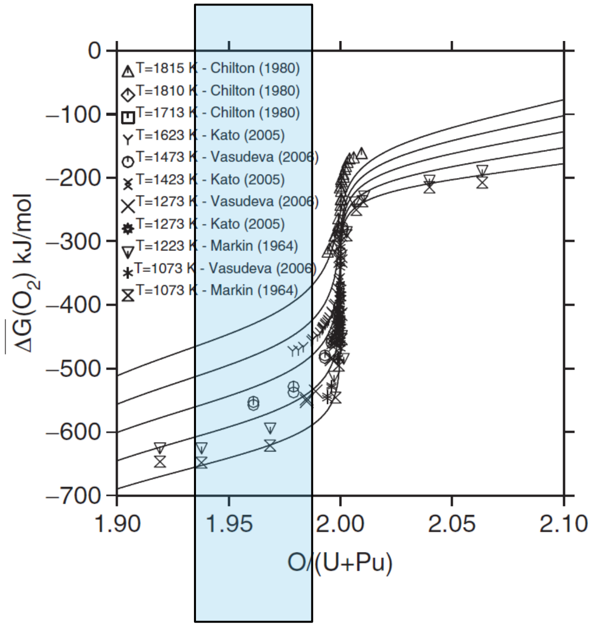


Fission product behaviour

Metallic

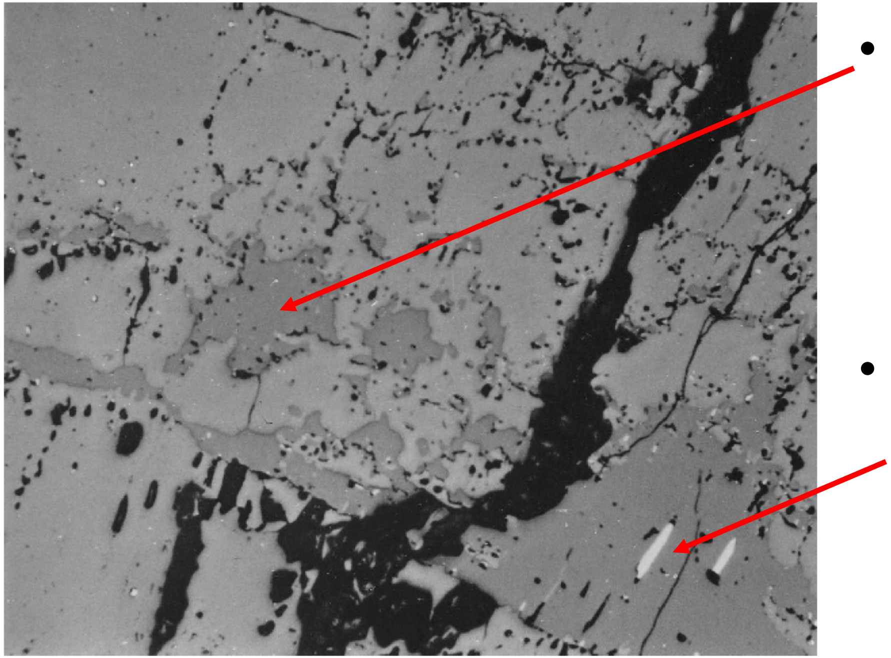
Oxidised





European Commission

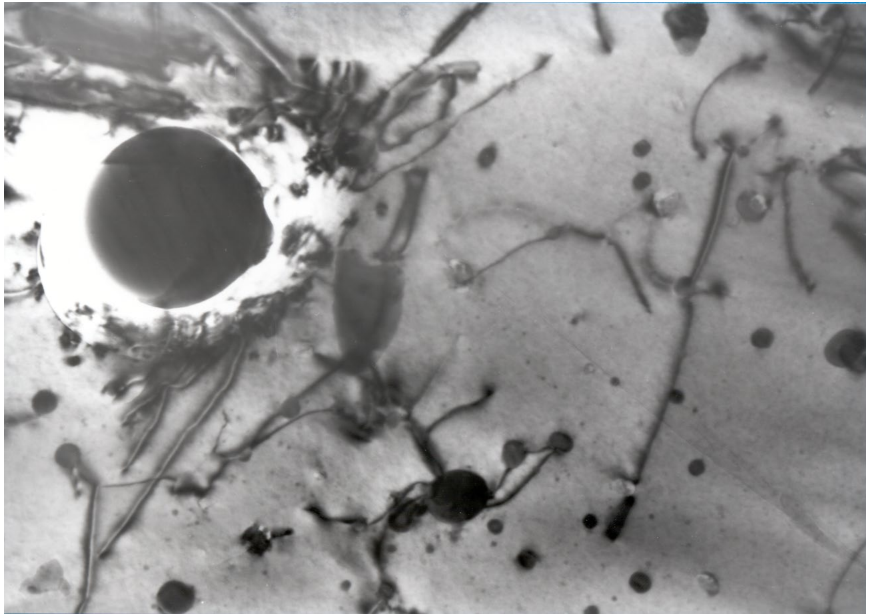
Fission product behaviour



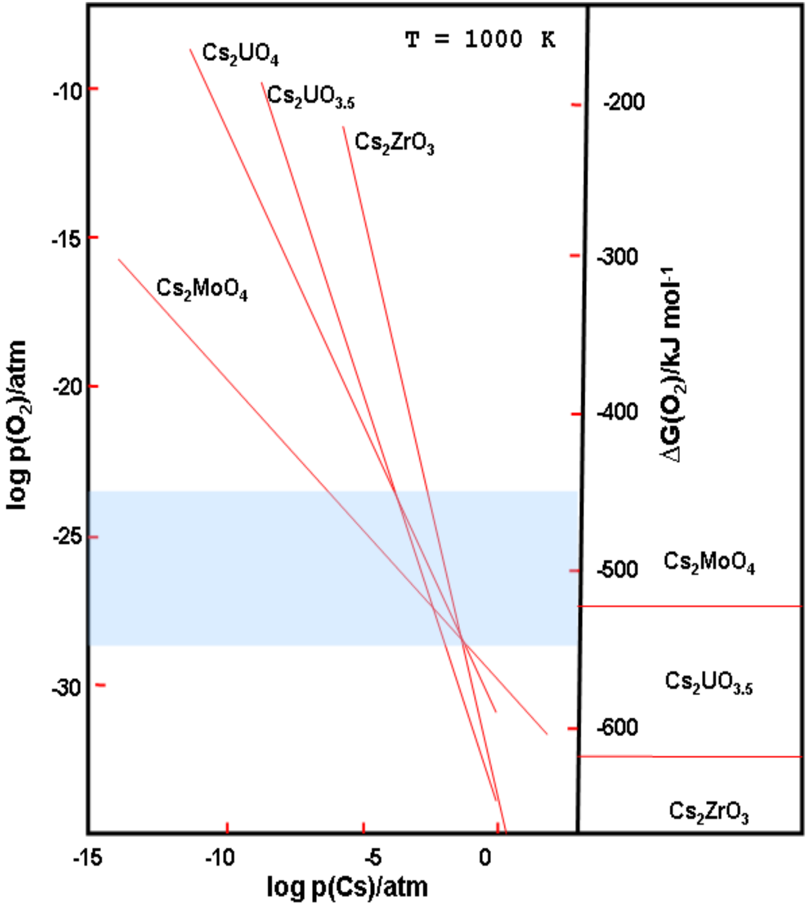
- "Grey phase" adjacent to columnar grains
  - Perovskite-type phase (Ba,Sr,Cs)(Zr,U,Pu,Mo)O_3
- "White inclusions"
  - (Pd,Tc,Rh,Tc,Mo)

European Commission

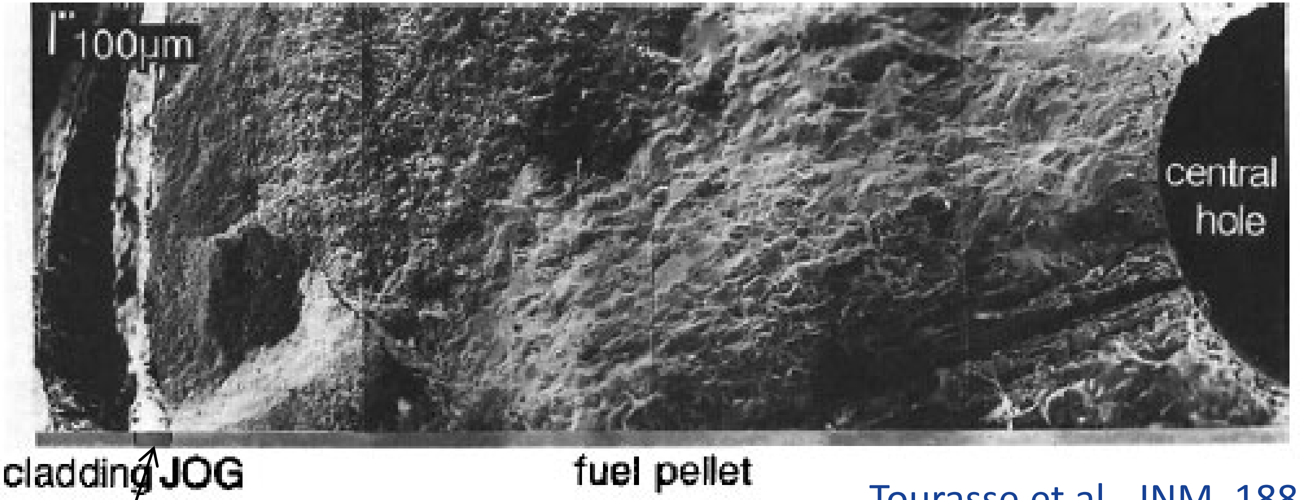
Fission product behaviour



Fission product behaviour



Fission product behaviour

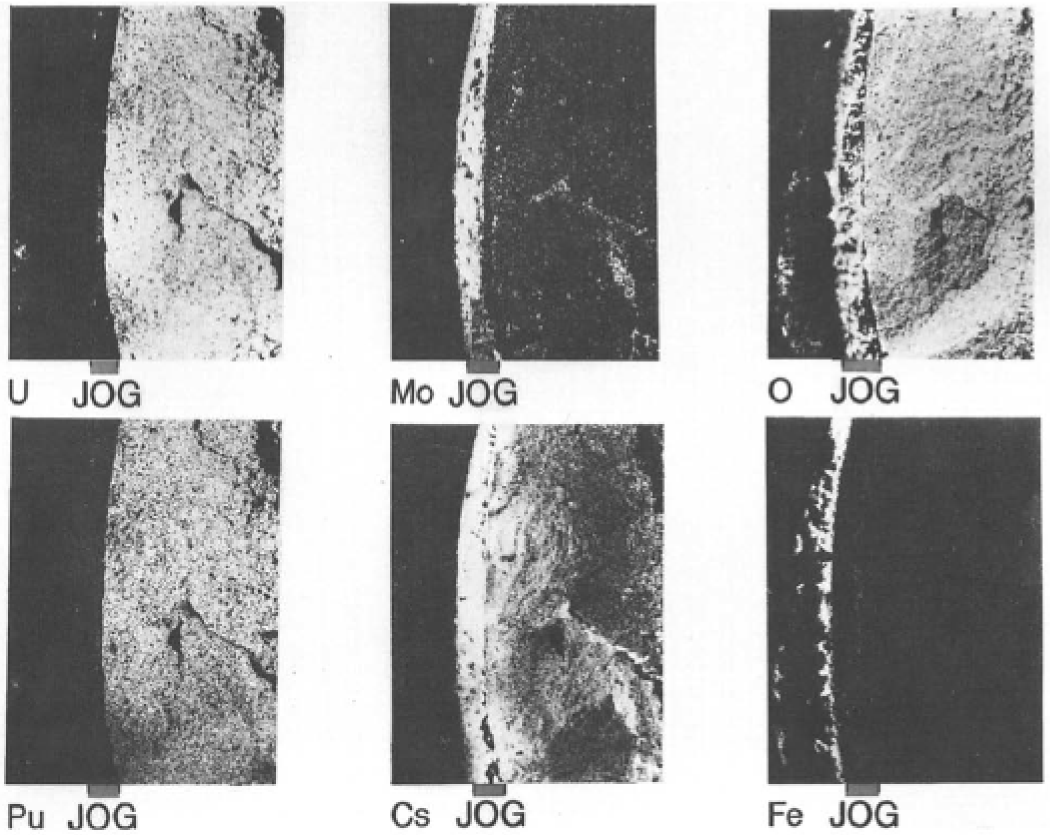


Tourasse et al., JNM, 188:49

JOG: Fuel-to-clad joint

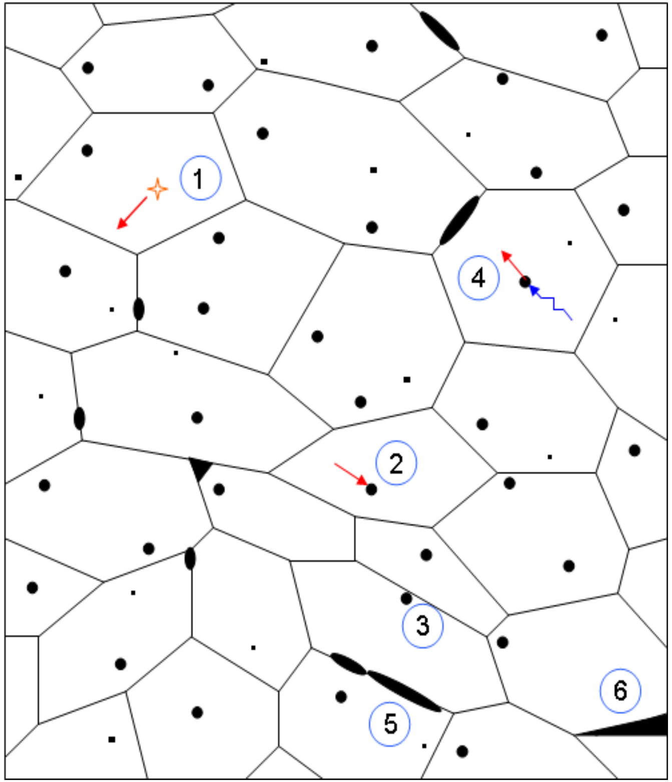
European Commission

Fission product behaviour



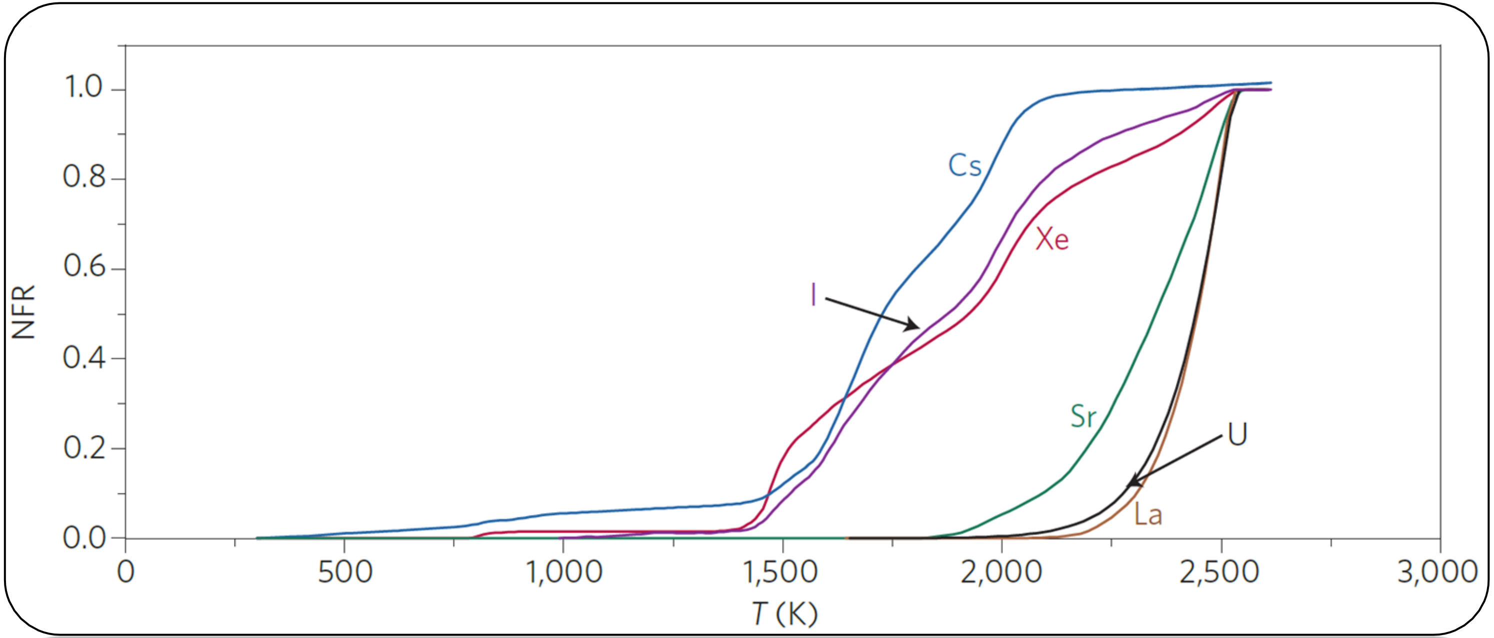
X-ray mapping of the JOG

Fission product behaviour



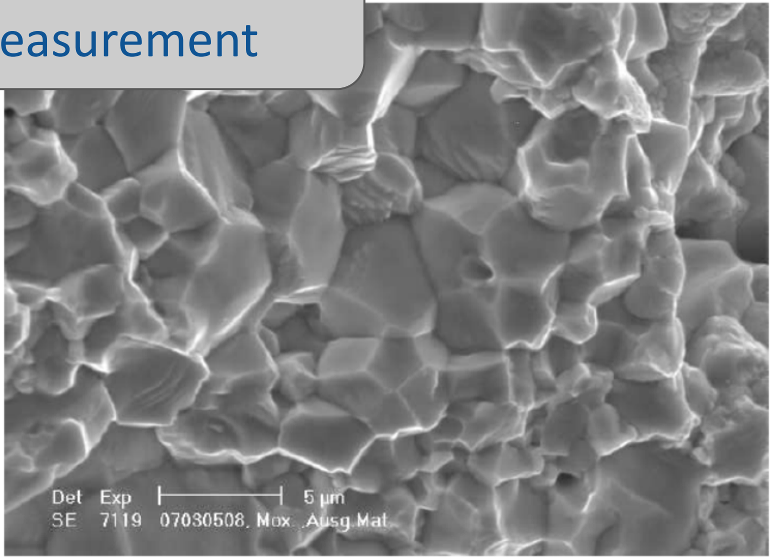
1. Atomic diffusion in the lattice (thermal and radiation induced)
2. Capture in intergranular bubbles
3. Migration of bubbles to grain boundaries
4. Resolution of gas
5. Aggregation to closed porosity
6. Venting via open porosity channels

Fission product behaviour

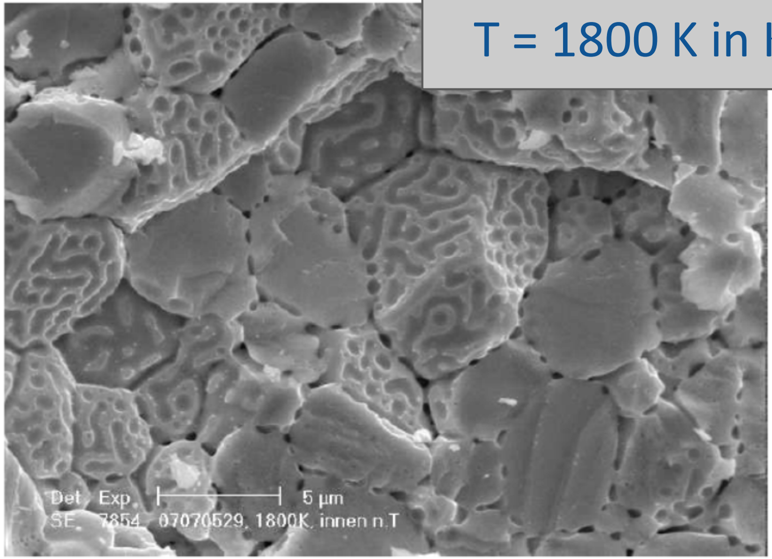


Fission product behaviour

SEM of before KEMS measurement



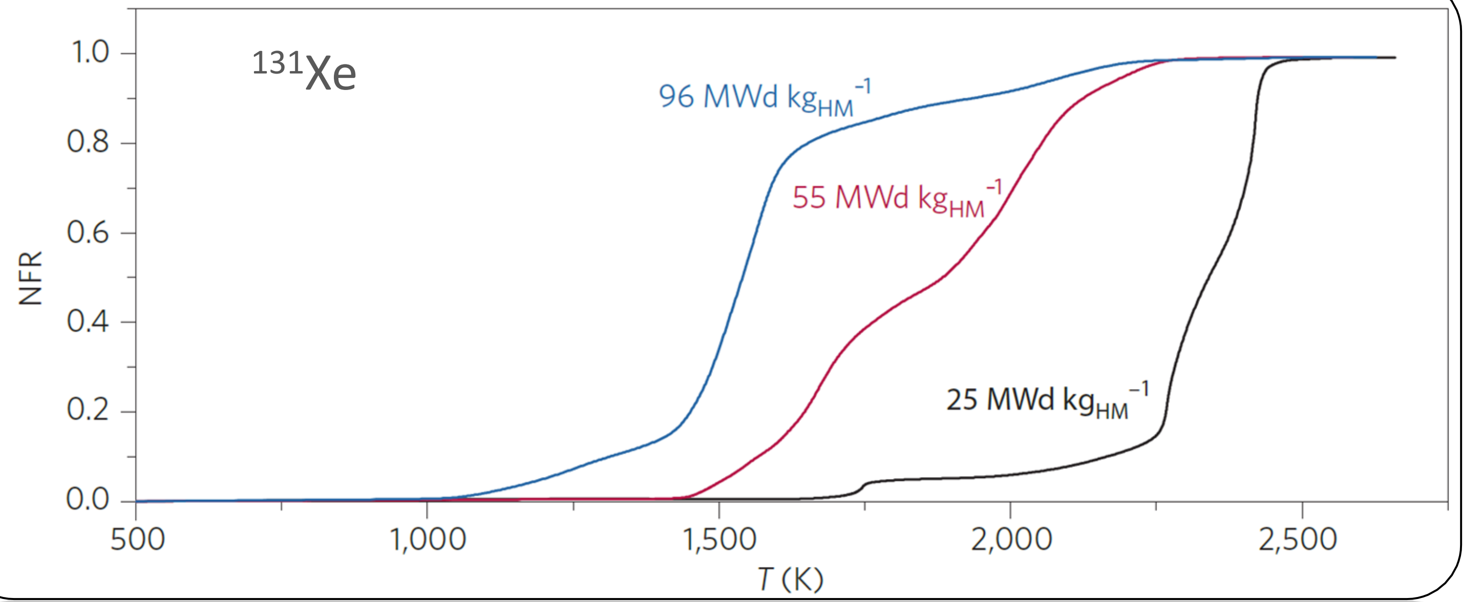
SEM after heating at T = 1800 K in KEMS



Konings et al., In: The Chemistry of Actinides and Transactinide Elements, 4th Edition., Volume 6, Chapter 34, p. 3665-3812, Springer Netherlands, 2010

European Commission

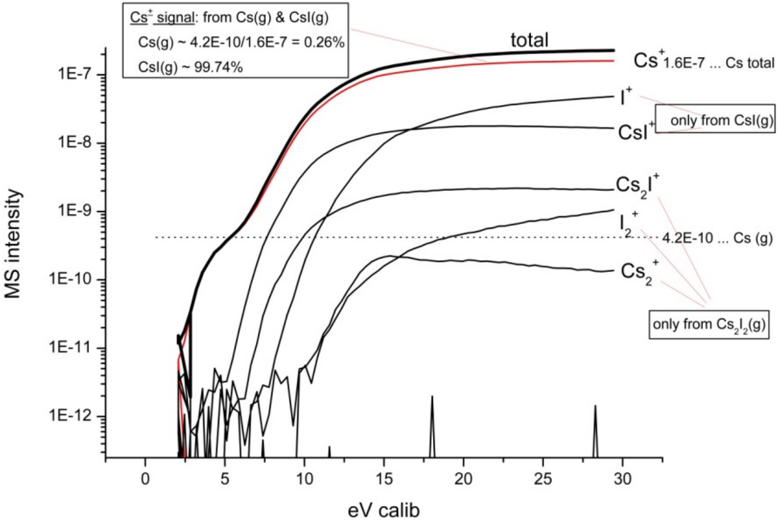
Fission product behaviour



Fission product behaviour

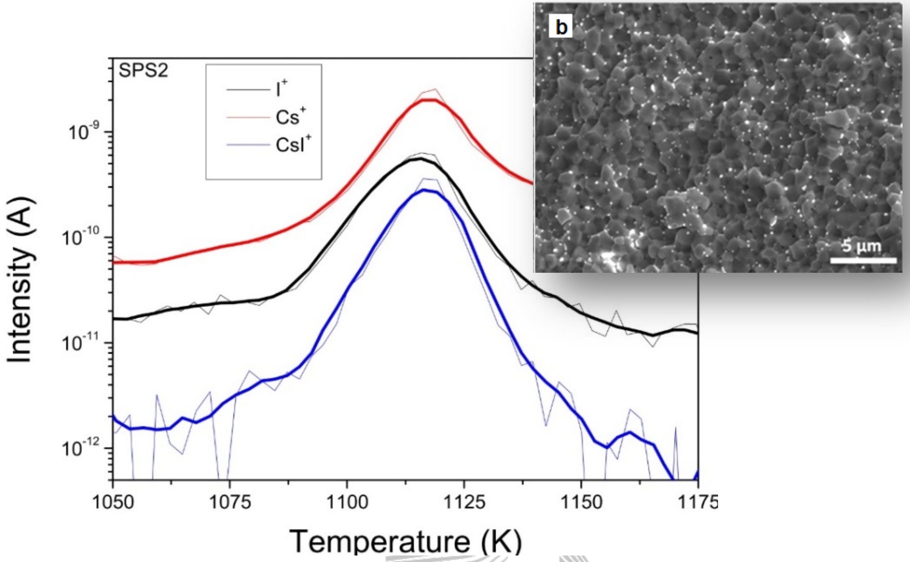
Cesium iodide

- Fragments: Cs^+, I^+, CsI^+, Cs_2I^+, Cs_2^+, I_2^+
- $I^+/CsI^+ \approx 3:1$
- Parallel release profiles



Simulated Fuel (UO_2+CsI)

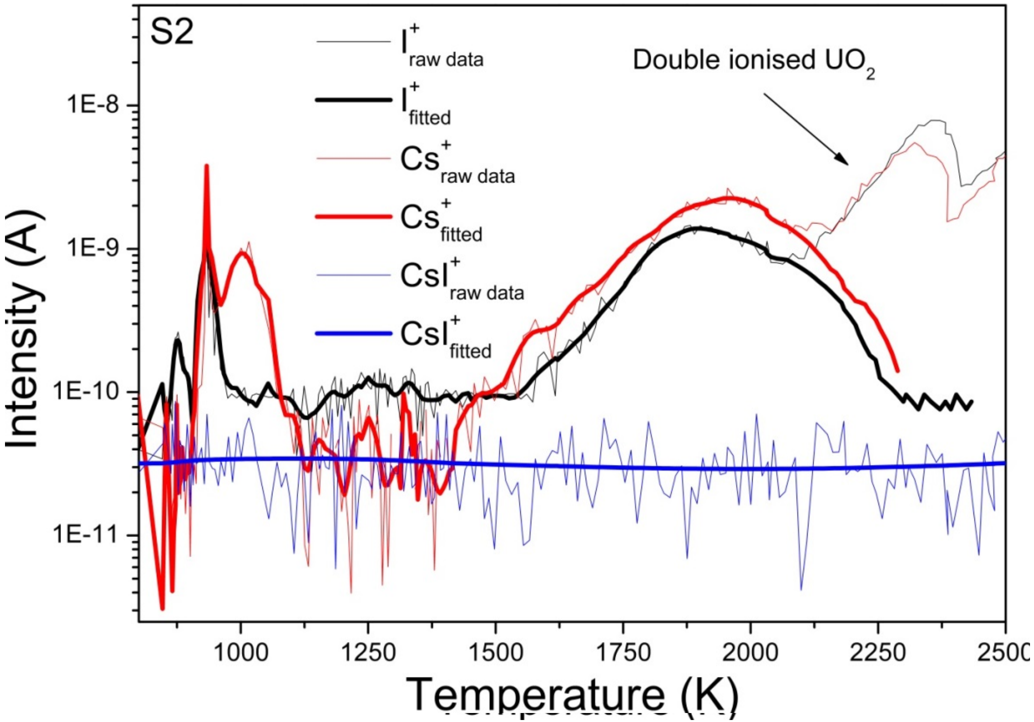
- Fragments: Cs^+, I^+, CsI^+
- $I^+/CsI^+ \approx 3:2$
- Parallel release profiles



Zappey et al., to be published

European Commission

Fission product behaviour



Irradiated fuel (BWR, 55 MWd/kg)

- Absolute intensities similar to Simfuel
- No CsI^+ ions (but higher background)
- Some temperature regions with parallell release

Conclusion: No clear evidence for CsI formation in irradiated fuel

- *Insufficient reaction sites?*
- *Gamma radiation?*

Summary:

- Nuclear fuel is not in chemical equilibrium
- But at microscopic scale processes can be described by equilibrium thermodynamics (local equilibrium)
- Chemical thermodynamics give the boundary conditions; kinetics need to be considered also

Stay in touch

EU Science Hub: ***ec.europa.eu/jrc***

Twitter: ***@EU_ScienceHub***

Facebook: ***EU Science Hub - Joint Research Centre***

LinkedIn: ***Joint Research Centre***

YouTube: ***EU Science Hub***

European Commission