The European Commission's science and knowledge service

Joint Research Centre

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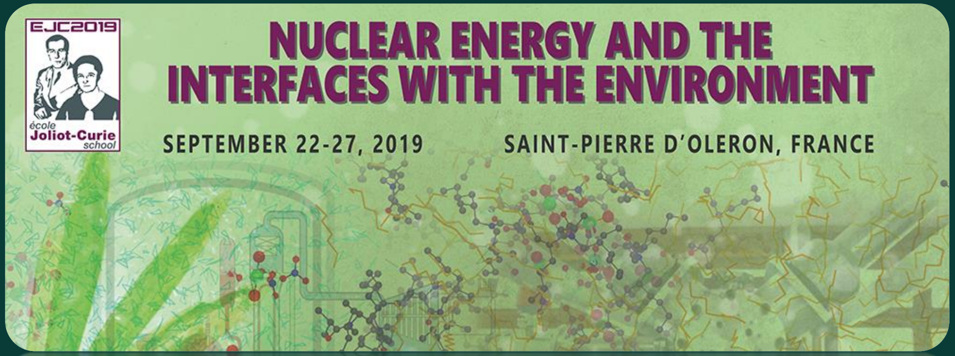
Fuel fabrication and evolution of structural properties under irradiation

Rudy Konings



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Content

1. Introduction
2. Properties of Uranium dioxide (and MOX)
3. Fuel Fabrication
4. Radiation effects
5. Behaviour under irradiation

Thursday morning

Thursday afternoon

Friday morning

1

Introduction

Nuclear Fuel

- ^{235}U

U/Pu

- $^{239}Pu/^{241}Pu$

← The only natural fuel (0.72%)

← Requires reprocessing

- ^{233}U

Th/U

← Breeding from natural ^{232}Th

Nuclear Fuel

- Low neutron capture cross section of non-fissile elements
- High fissile density
- No chemical reaction with cladding or coolant
- Favourable physical properties
- High mechanical stability (*isotropic expansion, stable against radiation*)
- High thermal stability (*no phase transitions, no dissociation*)

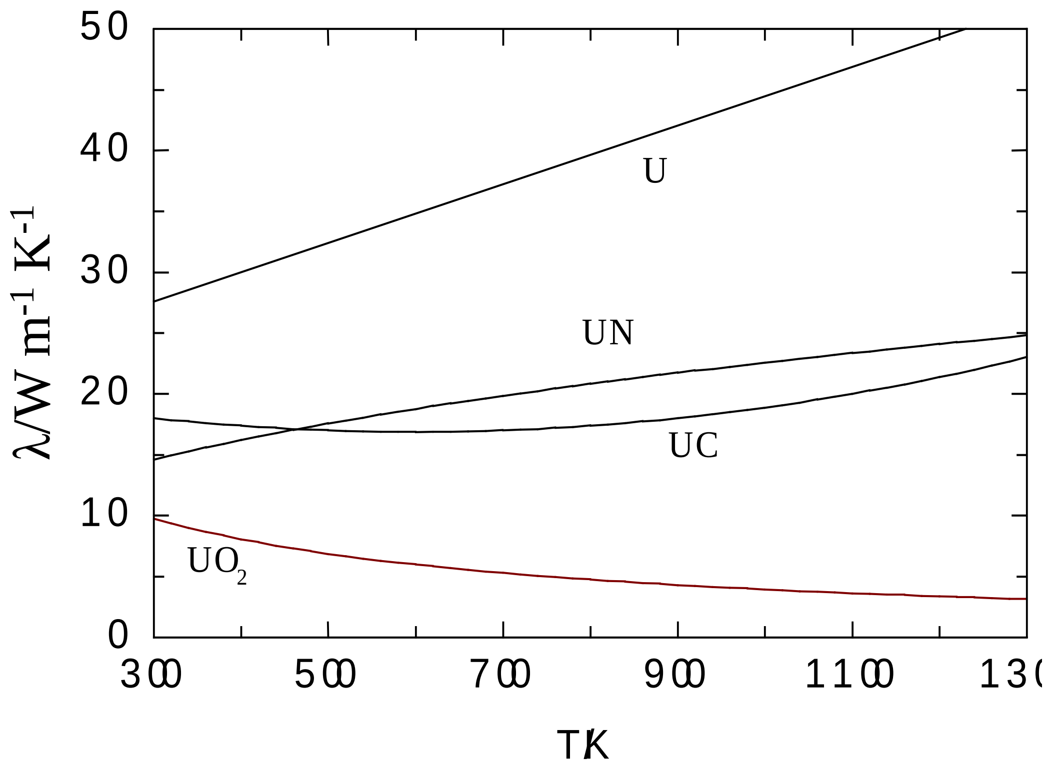
Nuclear Fuel: Requirements

- During normal operation:
  - Keep its shape
  - No melting should occur
  - Interaction with the cladding should not lead to critical mechanical or chemical interaction
- During accidental condition:
  - No excessive exothermal reactions
  - Limit the amount of volatile species
  - Limited interaction with other core and building materials

Nuclear Fuel: Types and variations

Fuel type
Metal
Oxide
Nitride
Carbide
Fluoride (salt)

	Melting point (K)	Density (g cm^{-3})	U-density (g cm^{-3})
U	1308	19.05	19.05
UO_2	3073	10.95	9.6
UC	2798	13.63	12.97
UN	3123	14.32	13.53



Nuclear Fuel: Types and variations

Fuel type
Metal
Oxide
Nitride
Carbide
Fluoride (salt)

Light-water	PWR, BWR	UO_2
Heavy-water	CANDU	UO_2
Graphite-moderated	AGR, RBMK	UO_2
High-temperature gas cooled	HTR	UO_2, (ThO_2, UC)
Sodium-cooled	SPX, Monju	$(U,Pu)O_2$
	EBR-II	(U,Pu) (U,Pu,Zr)
	PBTR	(U,Pu)C
Molten salt	MSRE	$LiF/BeF_2/ThF_4/UF_4$

Nuclear Fuel: Types and variations

e.g. UO_2

Fuel type
Metal
Oxide
Nitride
Carbide
Fluoride (salt)

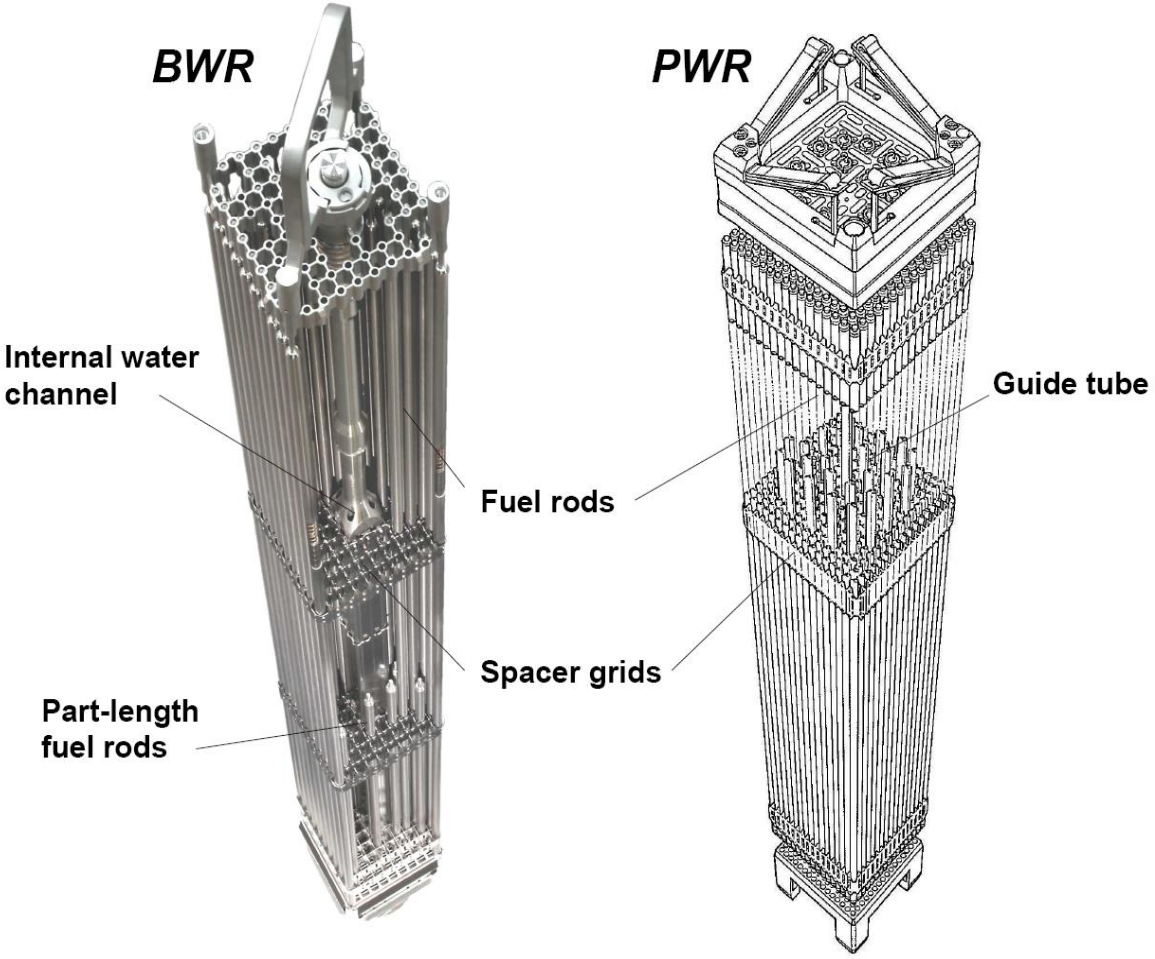
Fuel form
Single phase
Solid solution
Composite

Fuel packing
Pellet
Particle
Liquid

e.g. $(U,Pu)O_2$

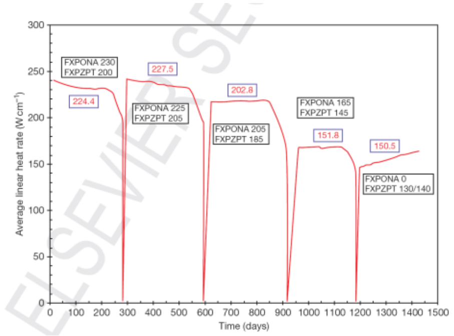
e.g. $Mo+UO_2$

Nuclear Fuel: Light Water Reactors



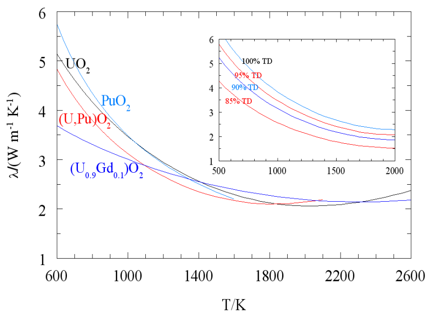
Source: AREVA S.A., Reproduced with permission

Nuclear Fuel: LWR temperature profile



Linear heating (W cm^{-1})

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



Thermal conductivity

Pellet radius (cm)

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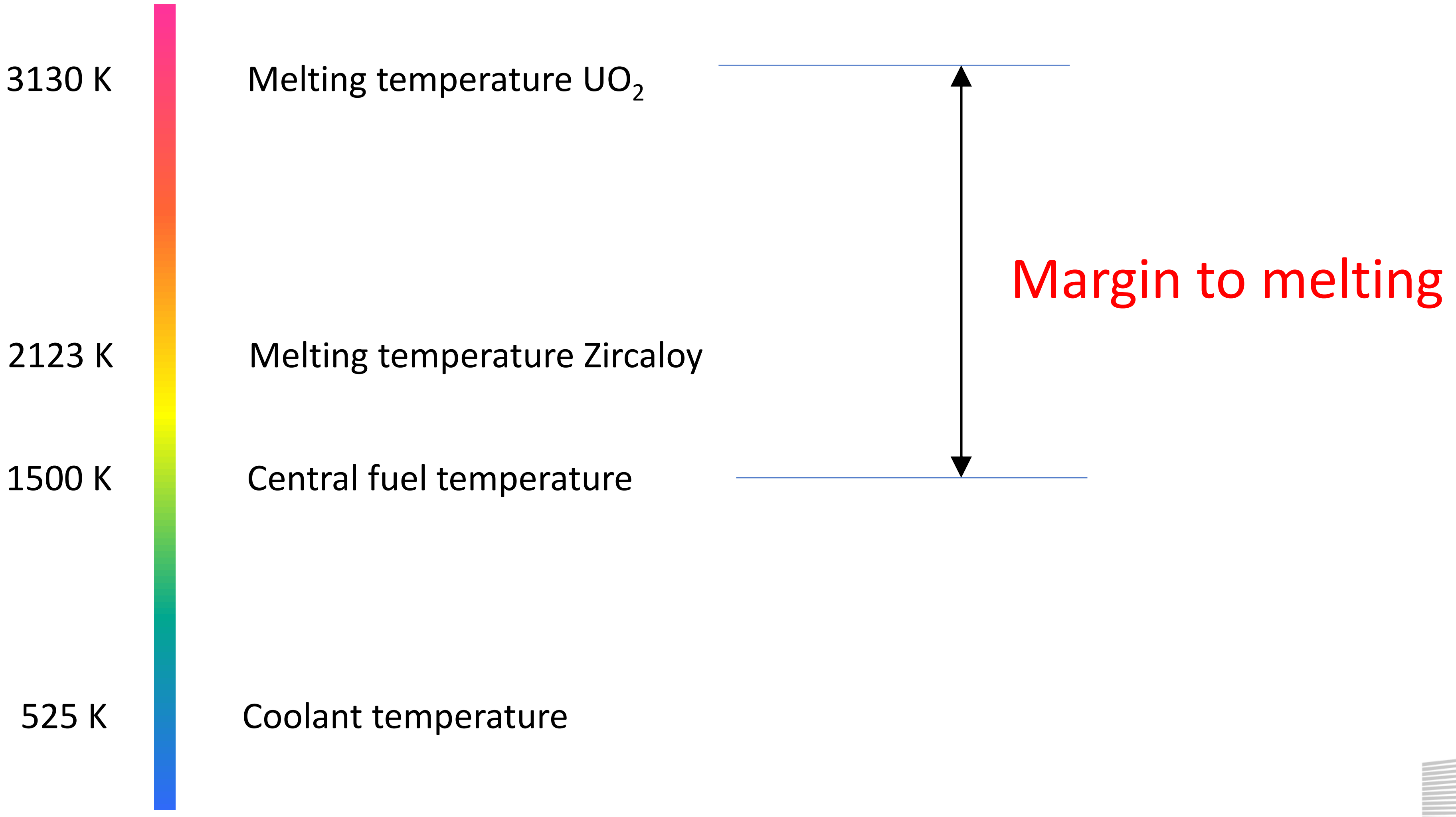
Nuclear Fuel: Margin to melting



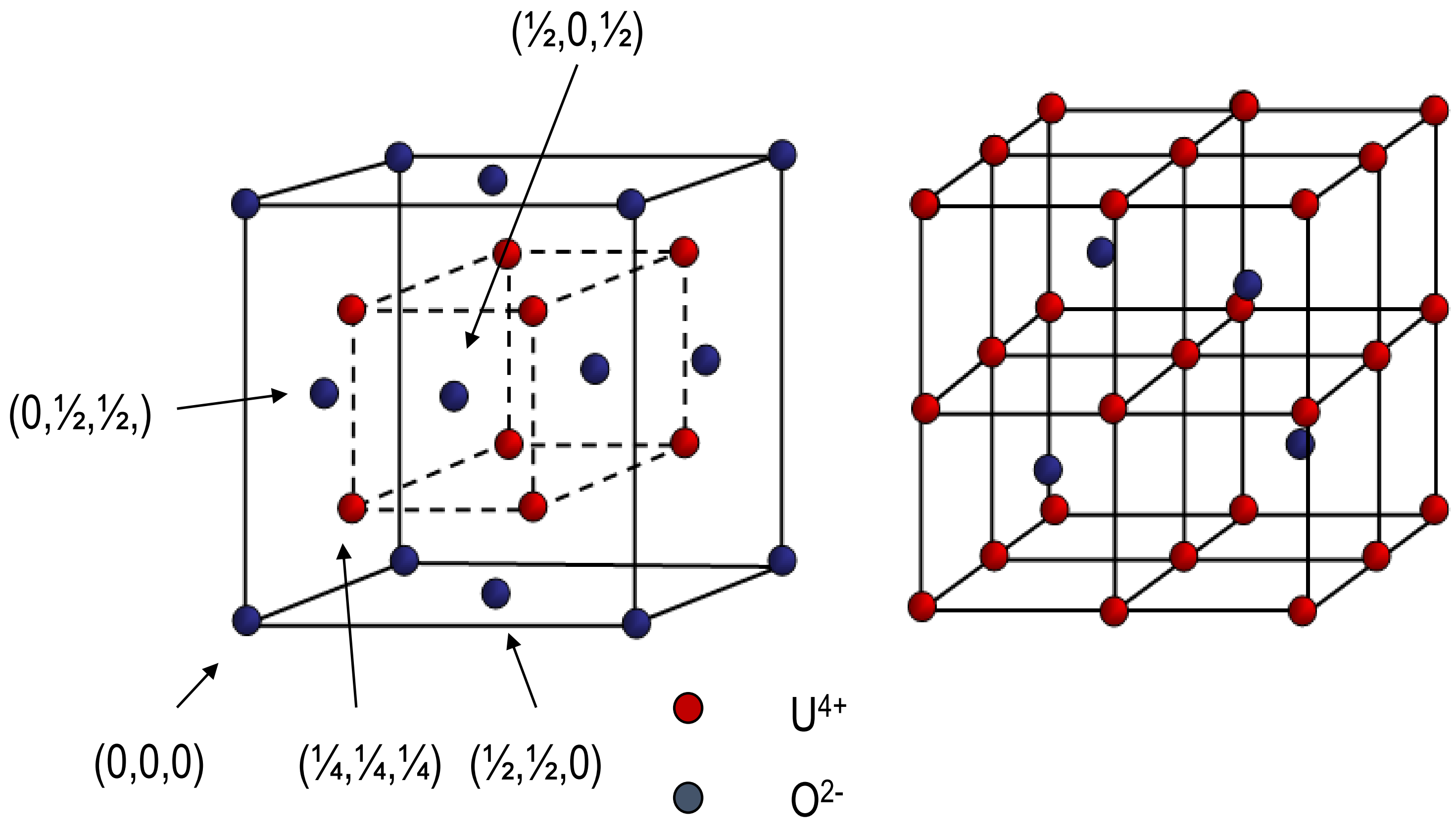
Nuclear Fuel: What affects the margins?

- Fuel Composition
- Microstructure
- Radiation effects
- Fuel Burn up
- Mechanical evolution

2

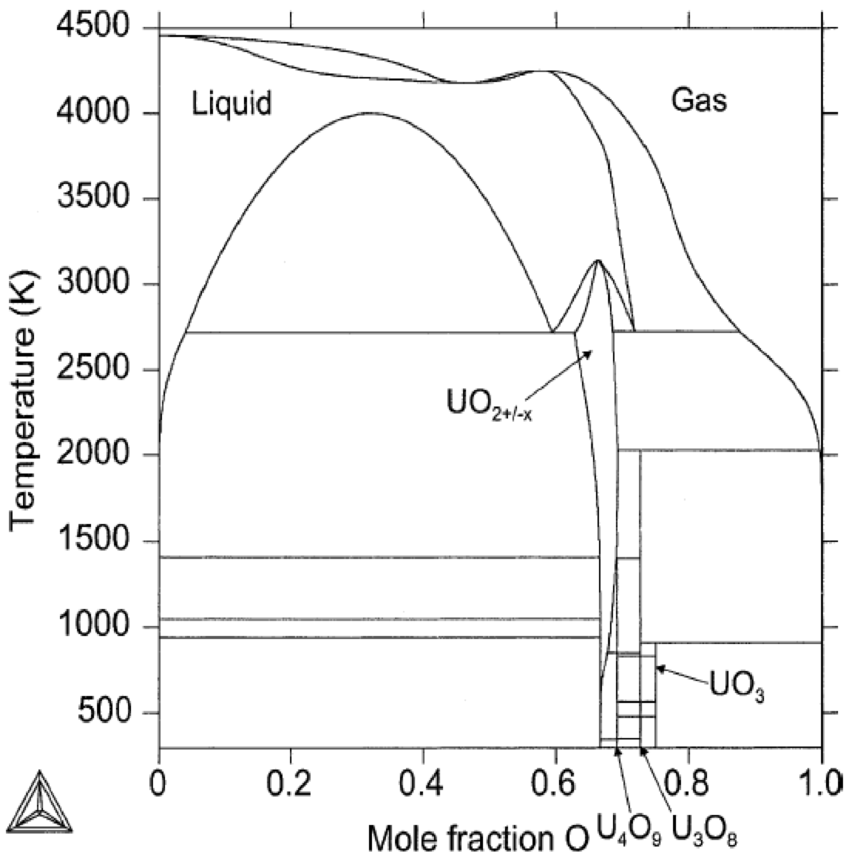
Properties of Uranium dioxide

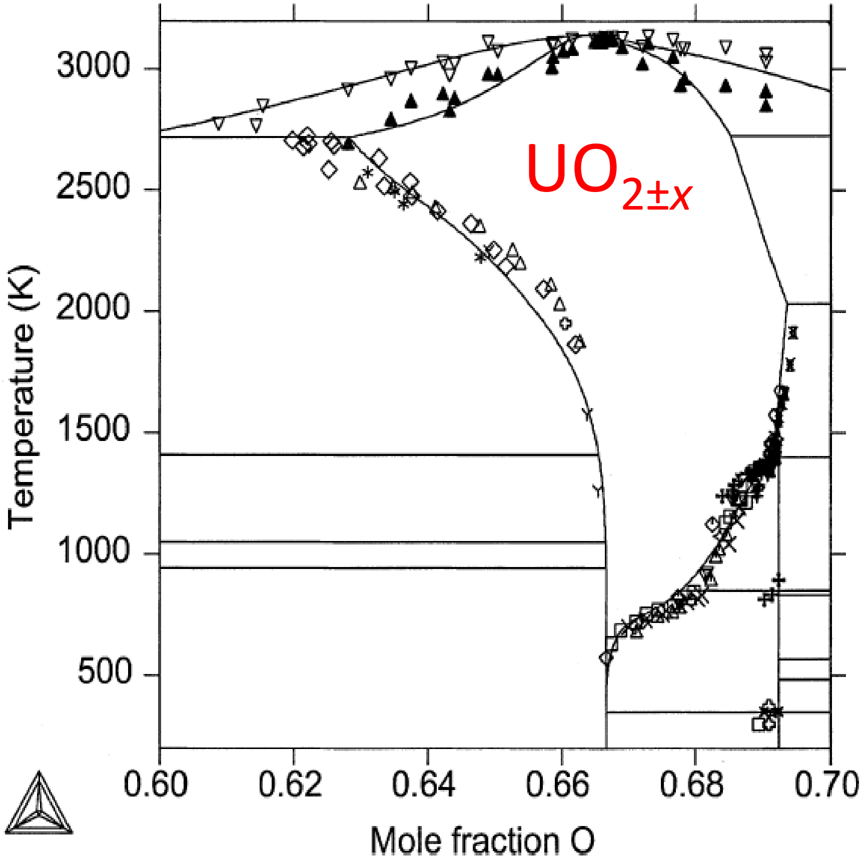
Nuclear Fuel:Uranium dioxide



Fluorite-type face-centered cubic structure

Nuclear Fuel: Uranium dioxide



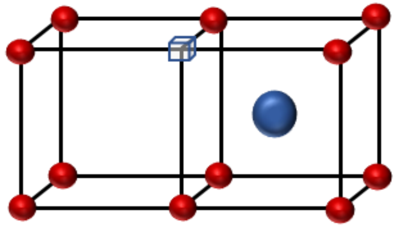


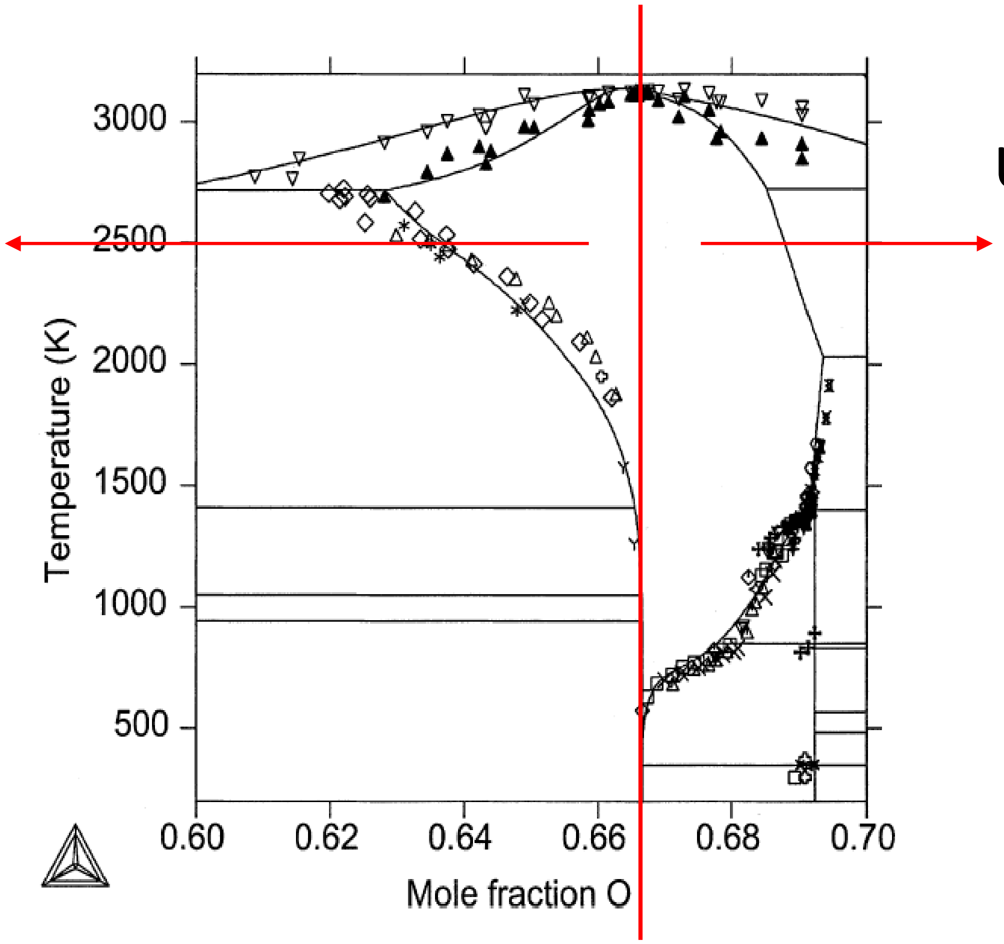
Source: Guéneau et al. (2002), J. Nucl. Mat. 304:161

Nuclear Fuel: Uranium dioxide

$UO_2 = UO_{2-x} + x/2O_2(g)$

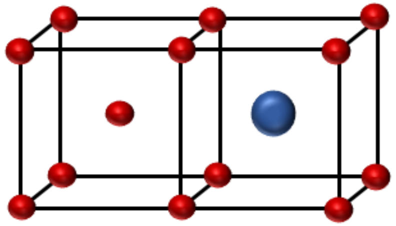
O vacancies



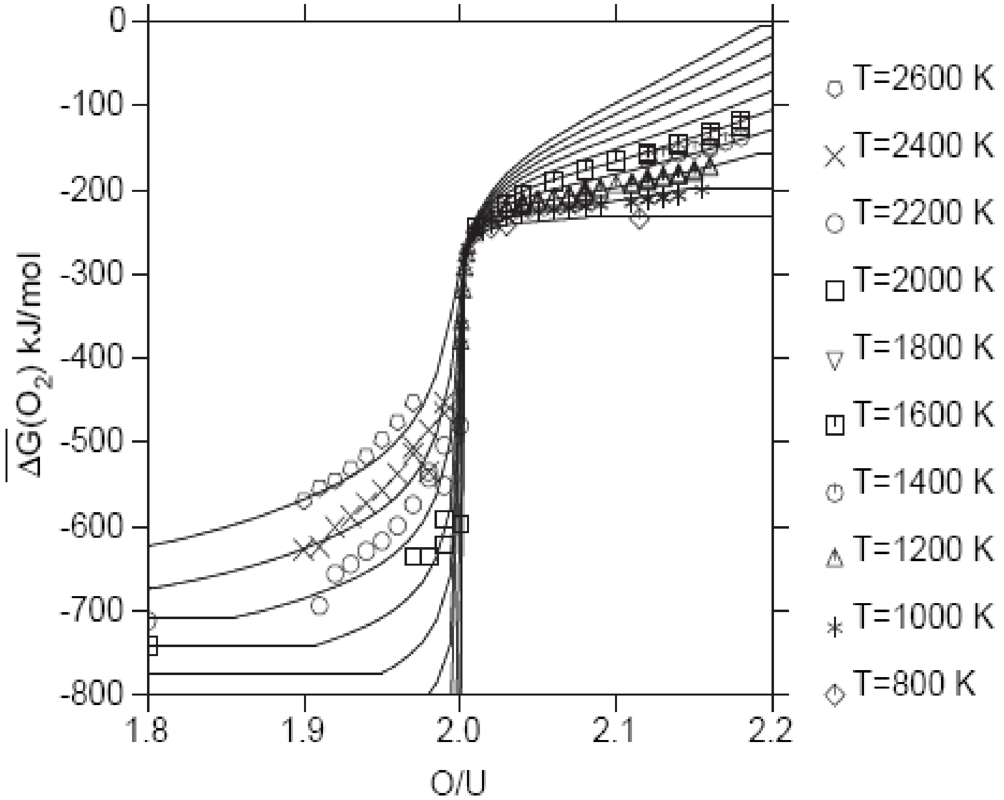


$UO_2 + x/2O_2(g) = UO_{2+x}$

O occupying “hole” positions in the fcc lattice



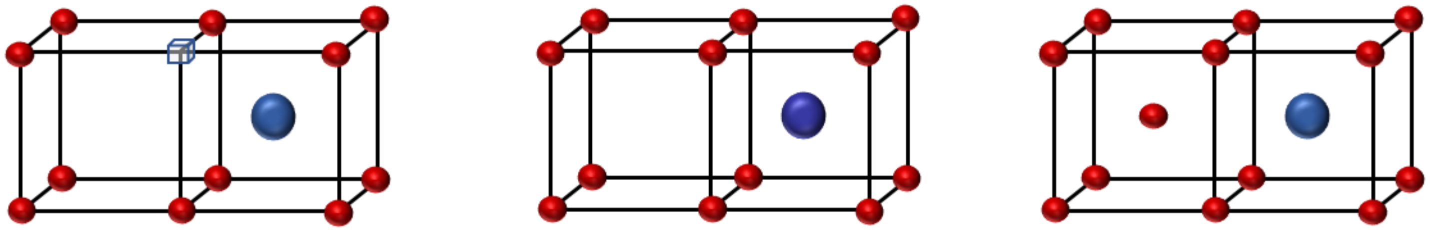
Nuclear Fuel: Uranium dioxide



$$2\left[O^{2-}\right]_{lattice} \rightleftarrows O_2(g) + 4e^-$$

$$\Delta\overline{G}(O_2) = RT \ln p(O_2)$$

The oxygen potential of UO_2 as a function of the O/U ratio and temperature.



Nuclear Fuel: Uranium dioxide

Key thermal properties for safety assessment:

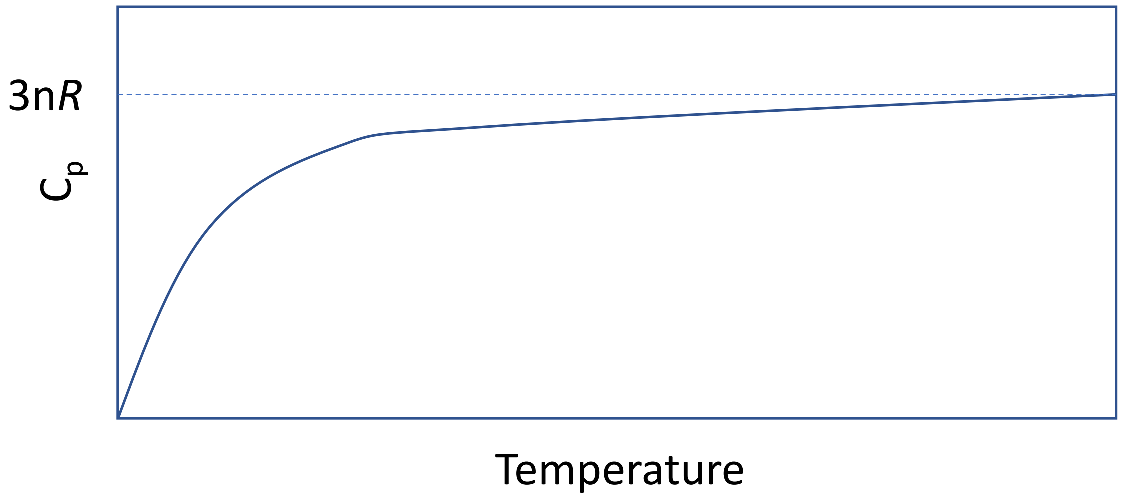
- Thermal expansion
- Melting temperature/Liquidus temperature
- Heat capacity
- Thermal diffusivity

Thermal conductivity

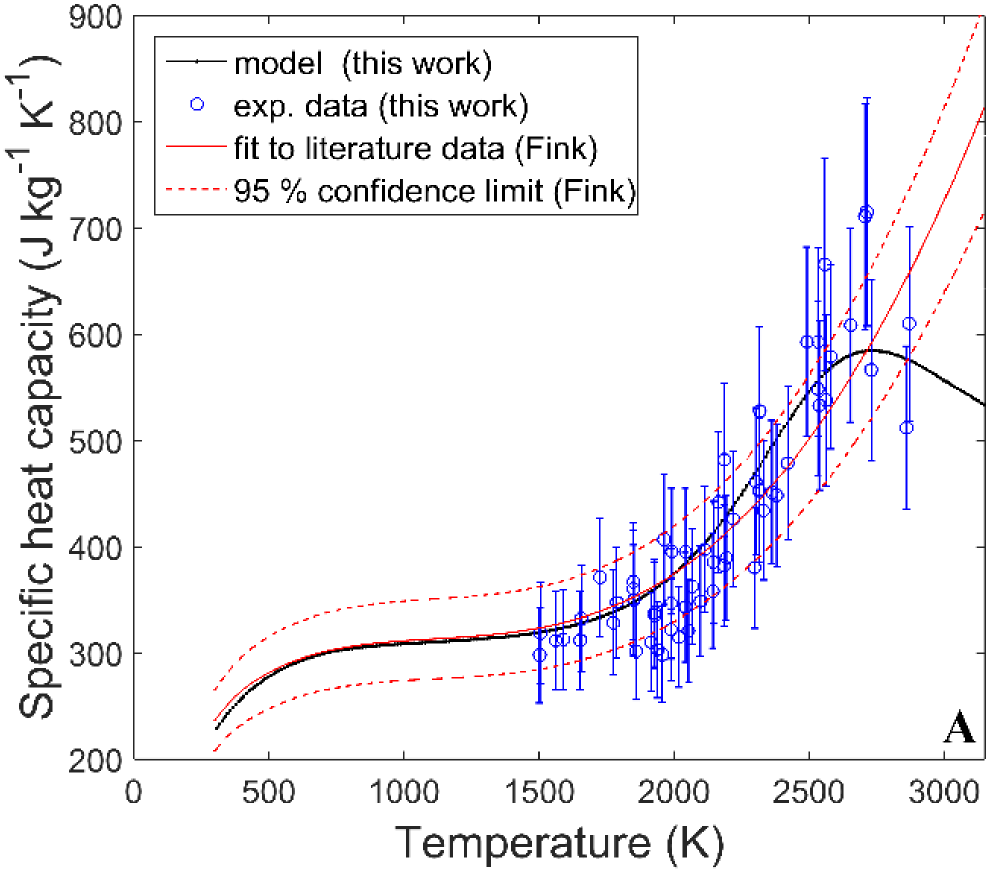
Strongly related to the phonon structure of the material

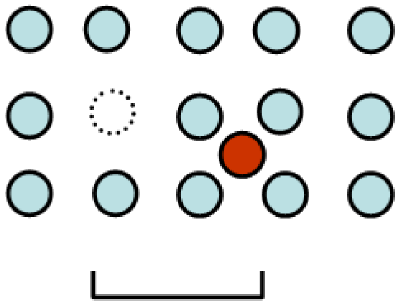
Nuclear Fuel: Uranium dioxide

Phonon theory predicts that for an **ideal crystal** the heat capacity is **zero** at T = 0 K and approaches the value **3n*R*** *(**Dulong-Petit** limit)* at high temperature



Nuclear Fuel: Uranium dioxide



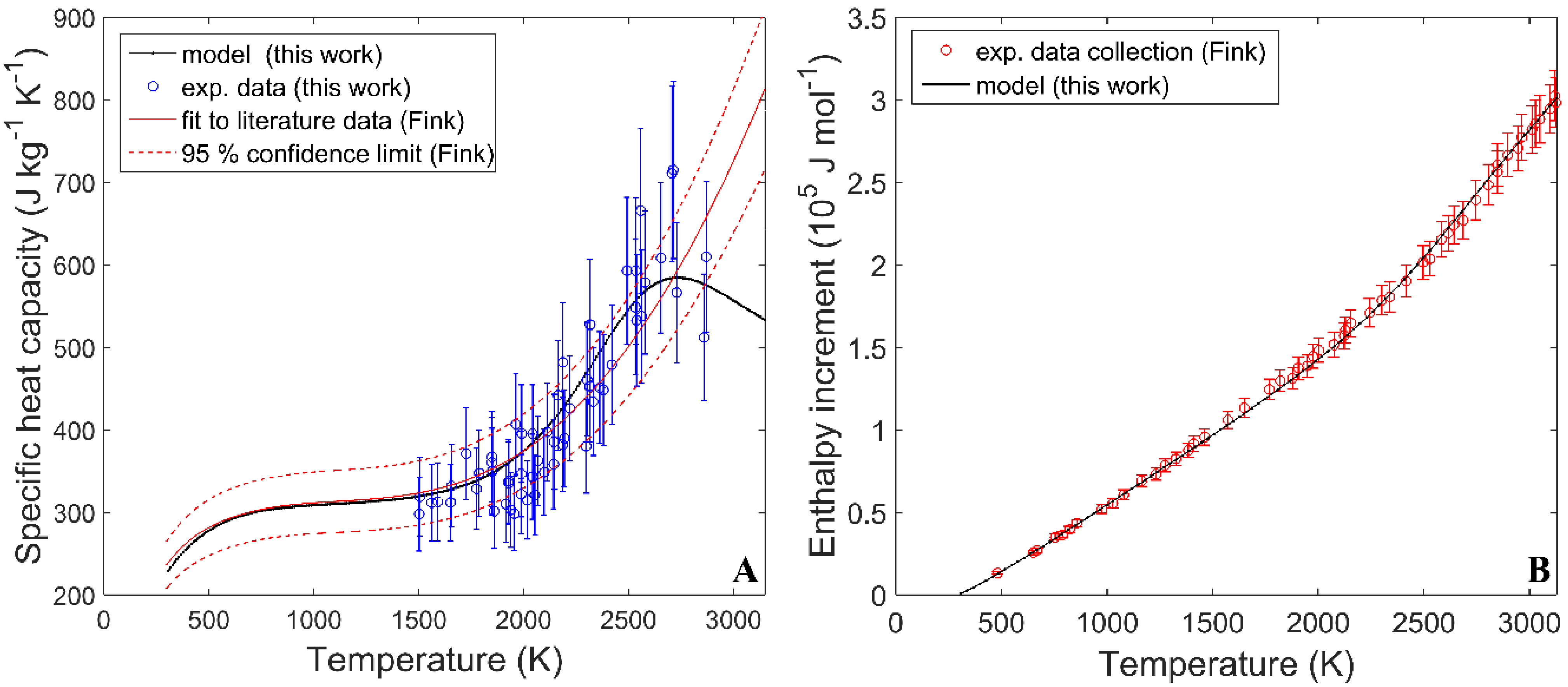


Oxygen Frenkel Pair (OFP) formation on the O sublattice

The heat capacity of UO_2 as a function of temperature.

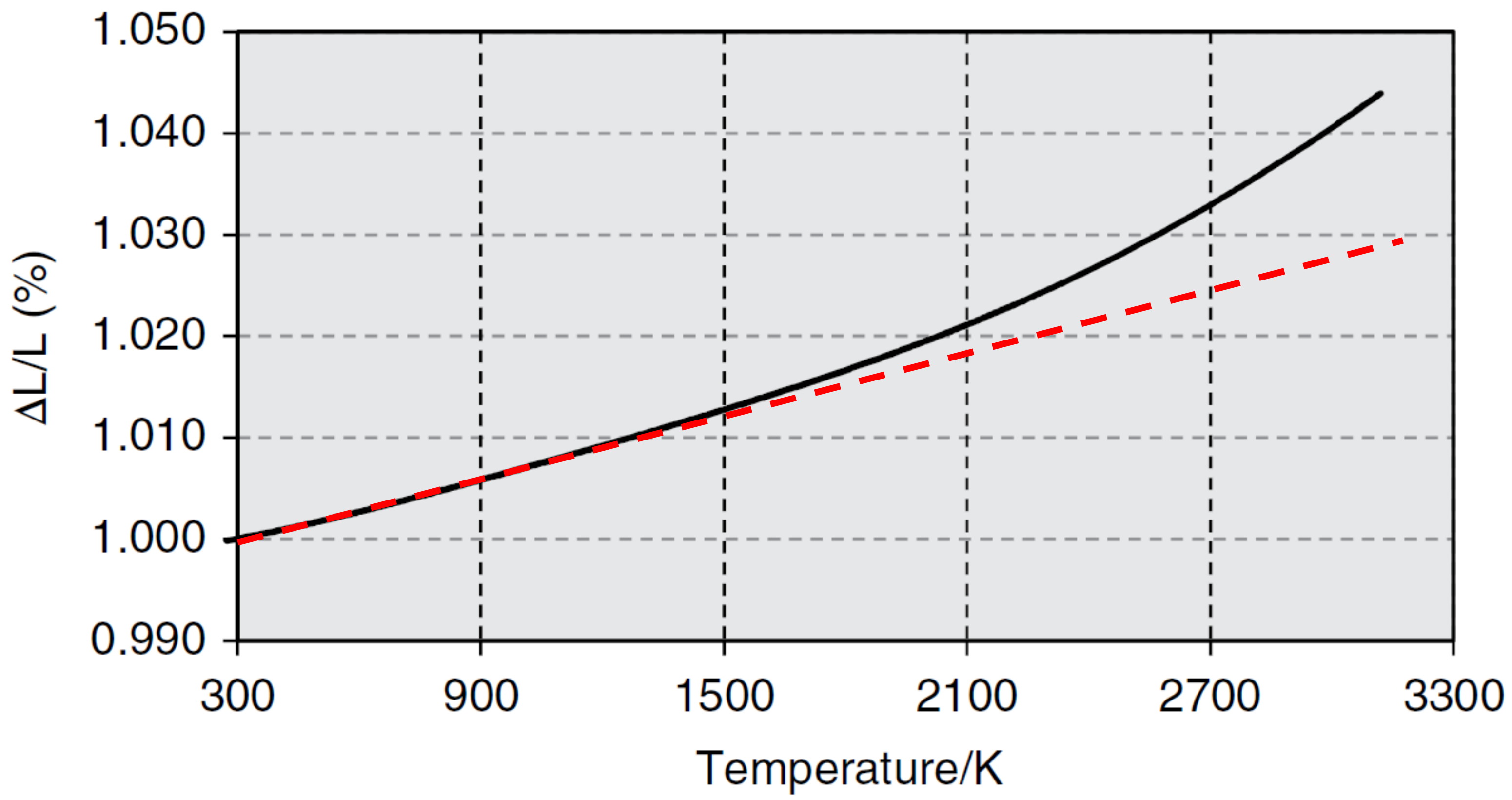
Source: Pavlov et al. Acta Mater. 139 (2017) 138.

Nuclear Fuel: Uranium dioxide



Source: Pavlov et al. Acta Mater. 139 (2017) 138.

Nuclear Fuel: Uranium dioxide



The thermal expansion of UO_2 expressed as ΔL/L a function of temperature.

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Nuclear Fuel: Uranium dioxide

$$\lambda = \lambda_{phonon} + \lambda_{electronic}$$

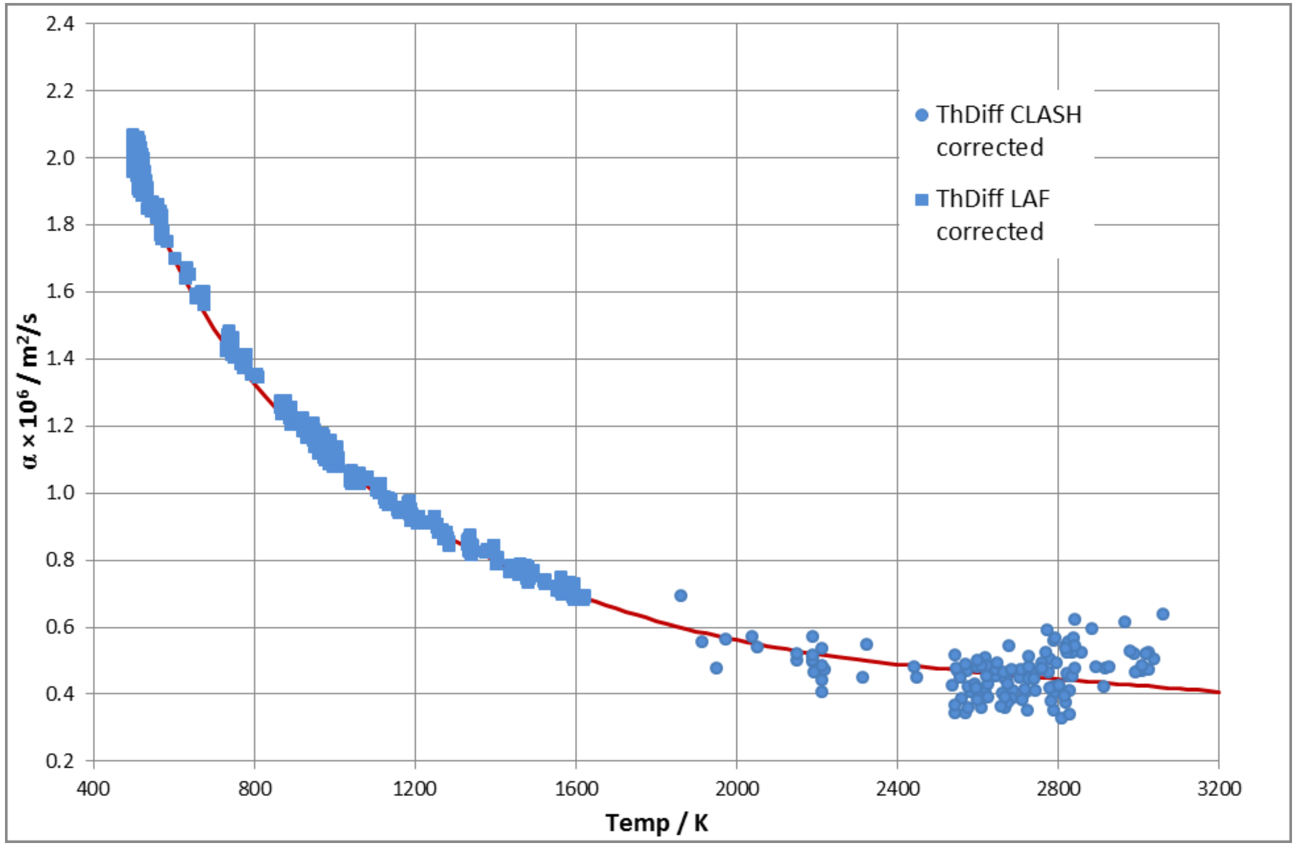
- The phonon contribution dominates at low temperature (< 2000 K)

$$\lambda = \frac{1}{A + BT}$$

Phonon scattering ← A BT → Phonon-phonon

- The electronic contribution becomes significant at high temperatures
- The phonon contribution is affected by impurities and microstructure changes

Nuclear Fuel: Uranium dioxide



The thermal diffusivity of UO_2 determined by LAF and CLASH instruments in JRC Karlsruhe.

Corrected to 95% theoretical density

Source: Vlahovic et al. J. Nucl. Mater. 499 (2018) 504.

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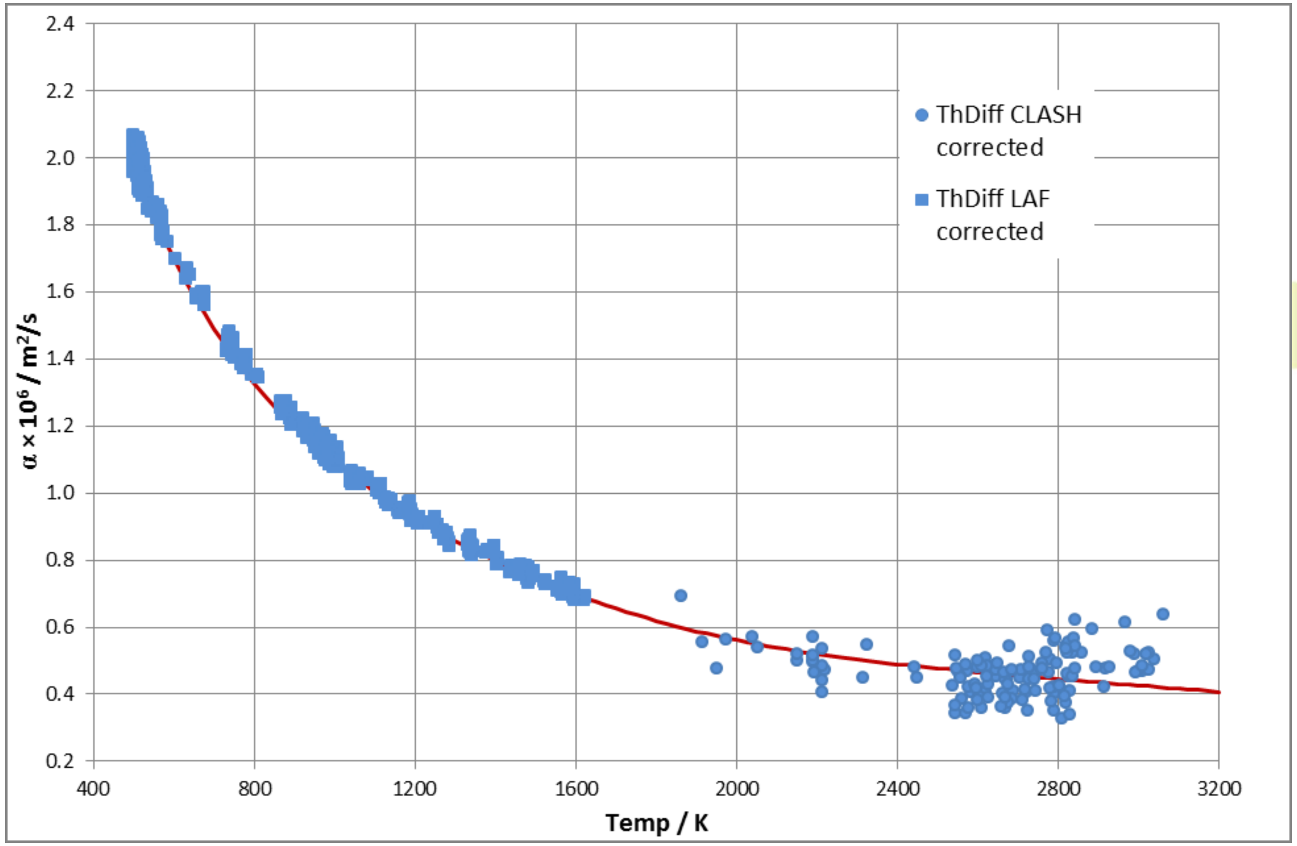
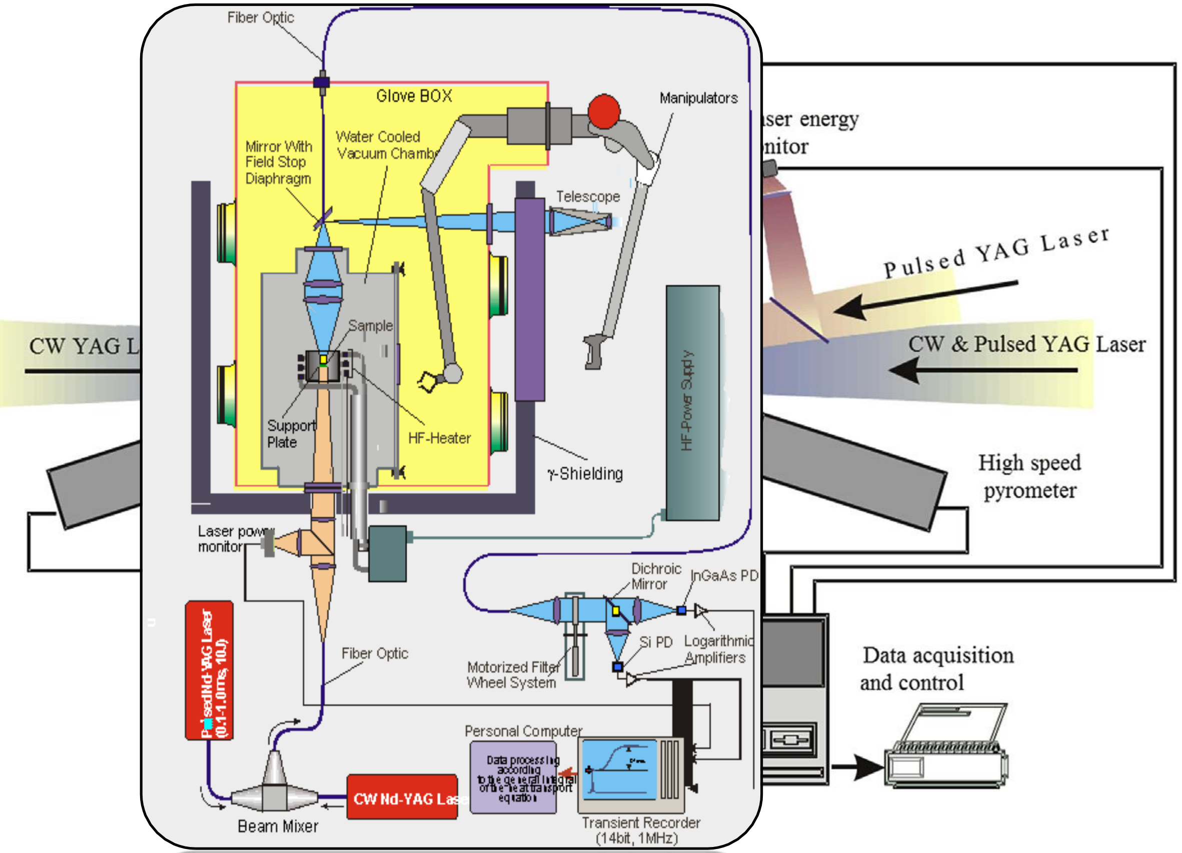
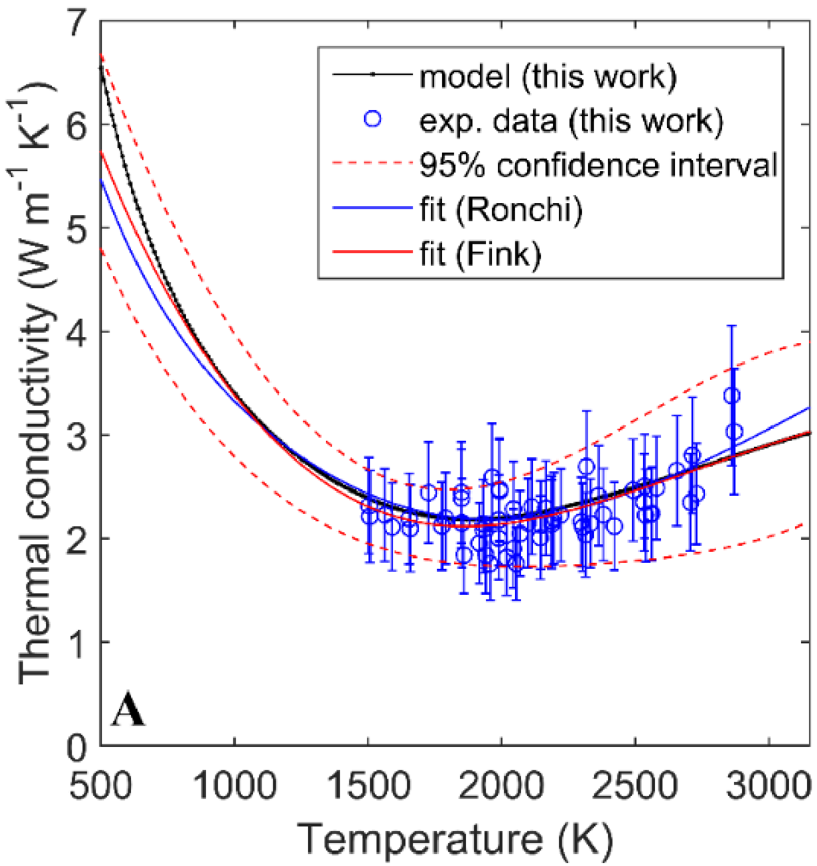
Nuclear Fuel: Uranium dioxide

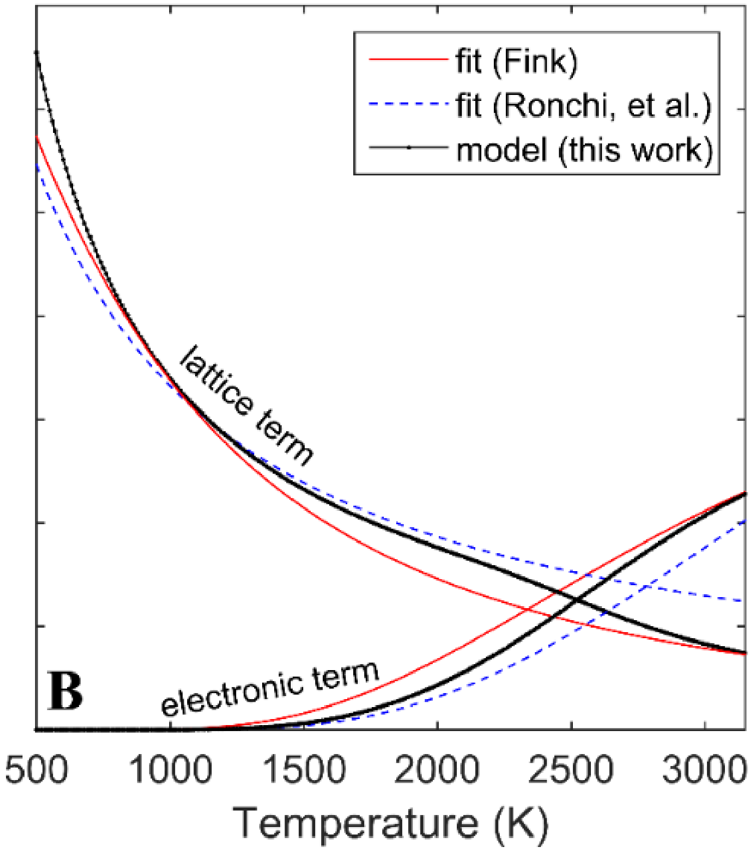


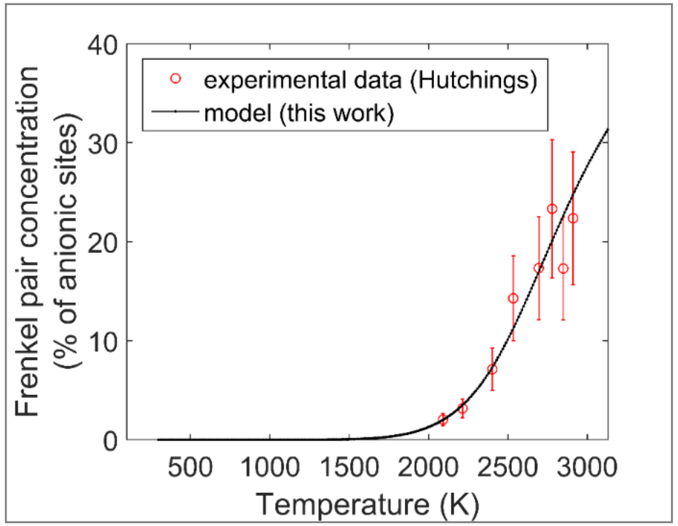


Source: Vlahovic et al. J. Nucl. Mater. 499 (2018) 504.

Nuclear Fuel: Uranium dioxide







Source: Pavlov et al. Acta Mater. 139 (2017) 138.

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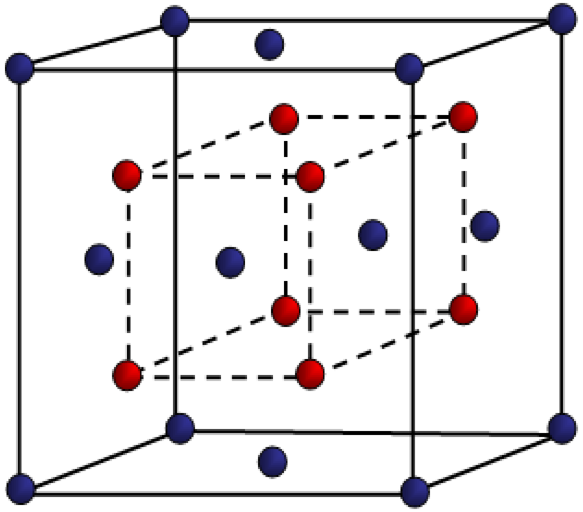
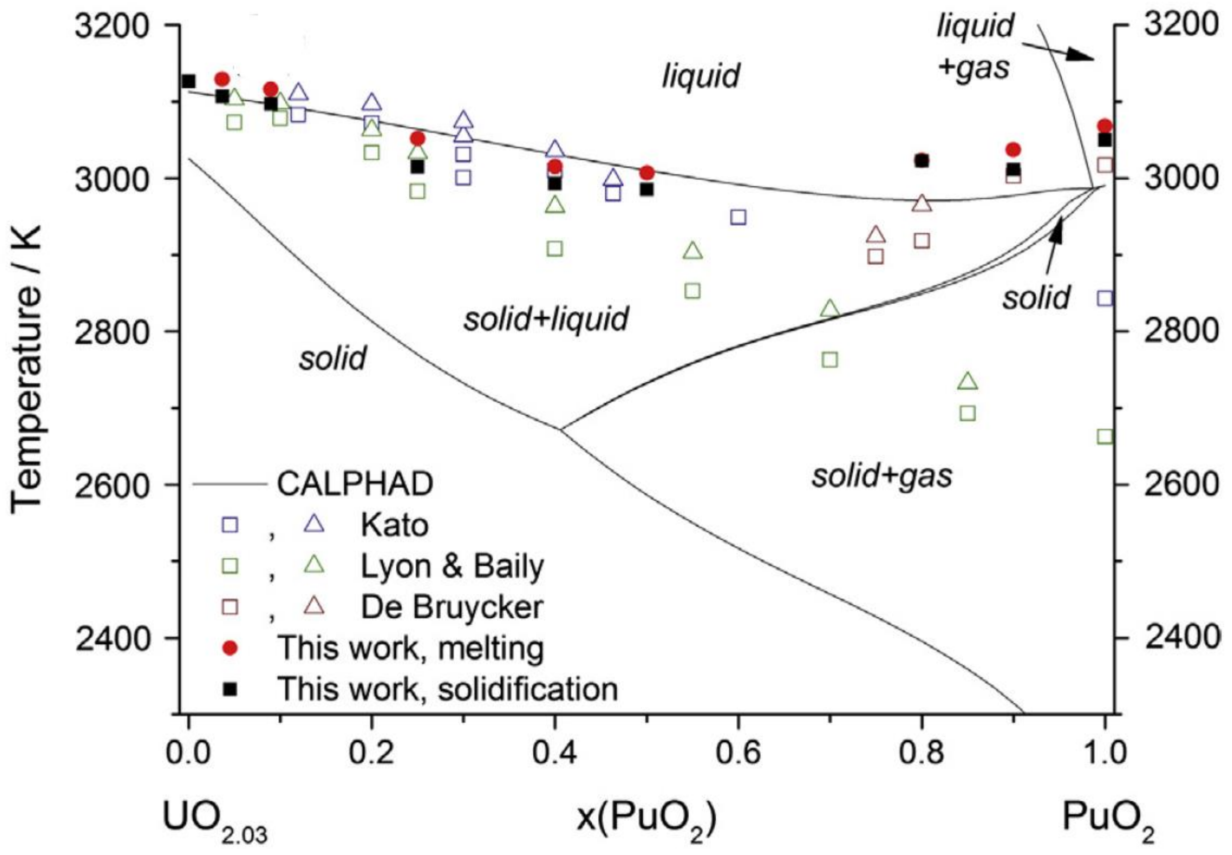
Nuclear Fuel:Mixed oxide

- Plutonium dioxide also has a fcc structure
- Pu^{4+} has a slightly smaller ionic radius
- Pu^{4+} has a different electronic structure
- PuO_2 is very poorly soluble in acids



Nuclear Fuel: Mixed Oxide

Böhler et al., J. Nucl. Mater. 448 (2014) 330



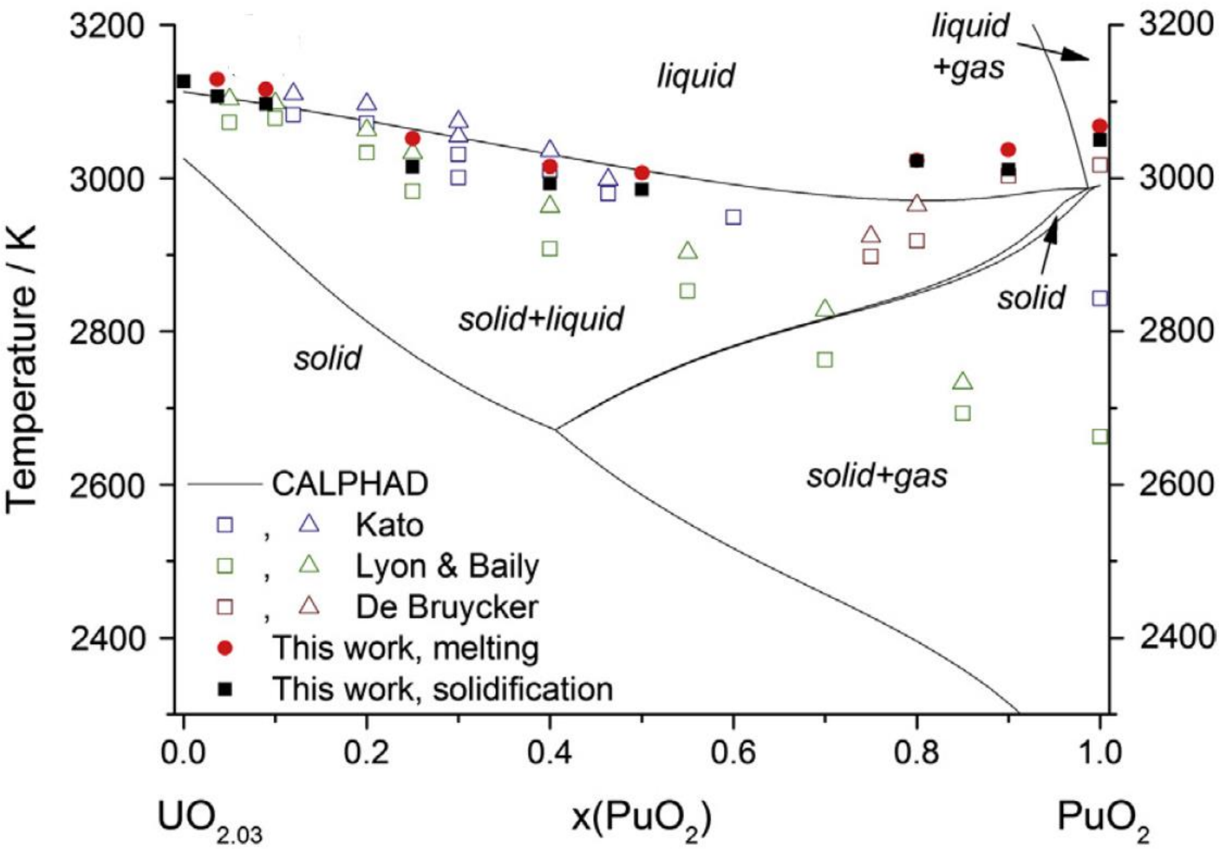
The revised UO_2-PuO_2 phase diagram experimentally determined by laser melting studies and modelled using the CALPHAD approach

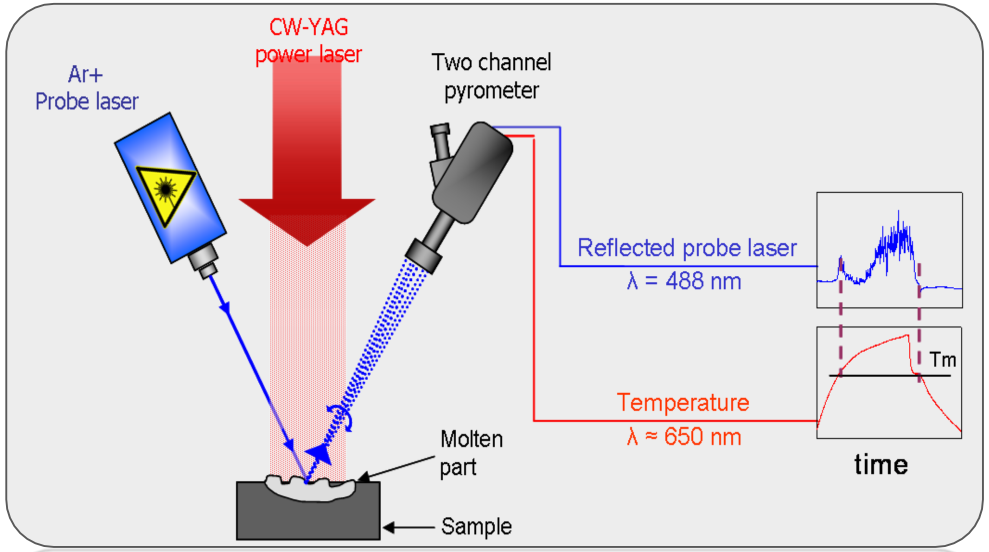
Melting temperature of PuO_2 was found to be about 300 K higher than accepted values from the 1960s

Source: Böhler et al. J. Nucl. Mater. 448 (2014) 330. .

Nuclear Fuel: Mixed Oxide

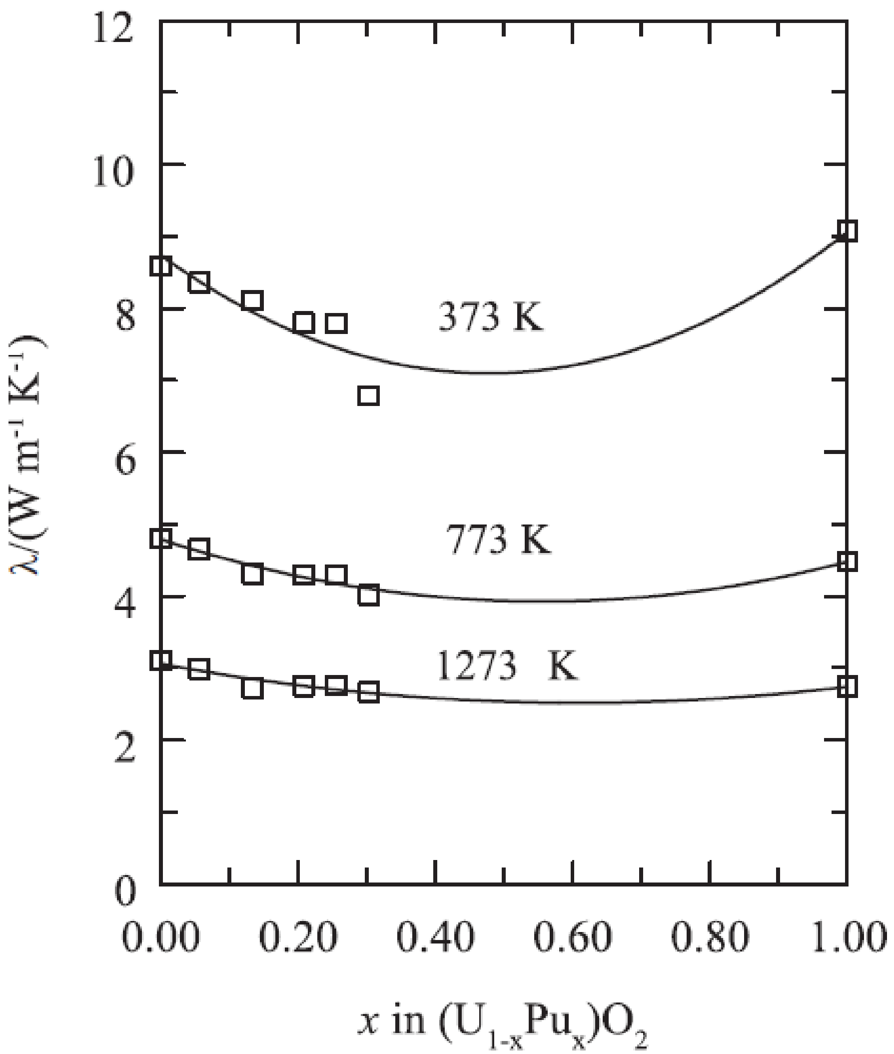
Böhler et al., J. Nucl. Mater. 448 (2014) 330





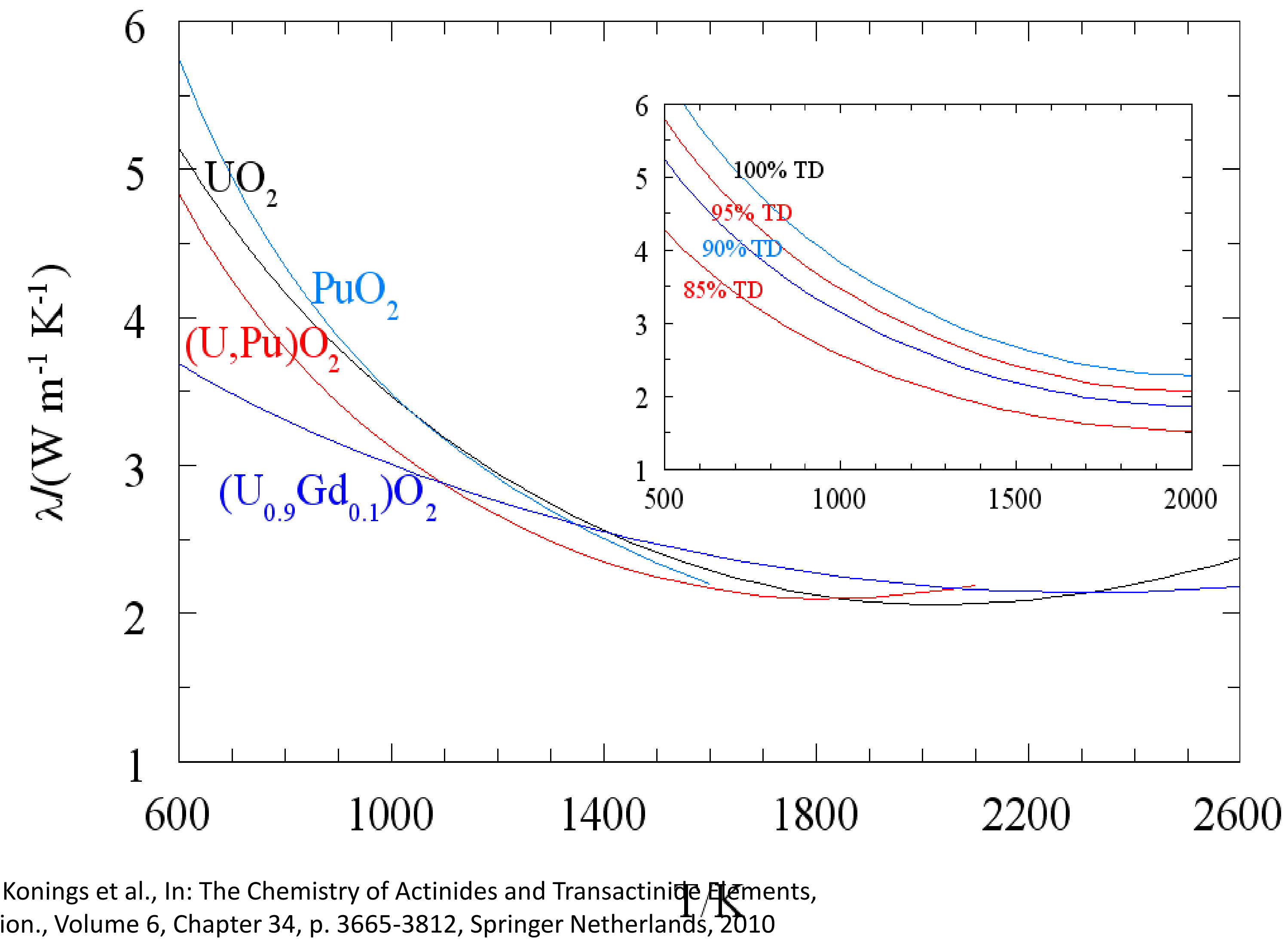
Source: Böhler et al. J. Nucl. Mater. 448 (2014) 330. .

Nuclear Fuel: Mixed Oxide



The thermal conductivity of $(U,Pu)O_2$ as a function of the Pu concentration

Nuclear Fuel: Mixed Oxide



Inset: The thermal conductivity of UO_2 is dependent on porosity

Maxwell-Euken correction:

$$\lambda = \lambda_0 \frac{1-P}{1+\beta P}$$

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

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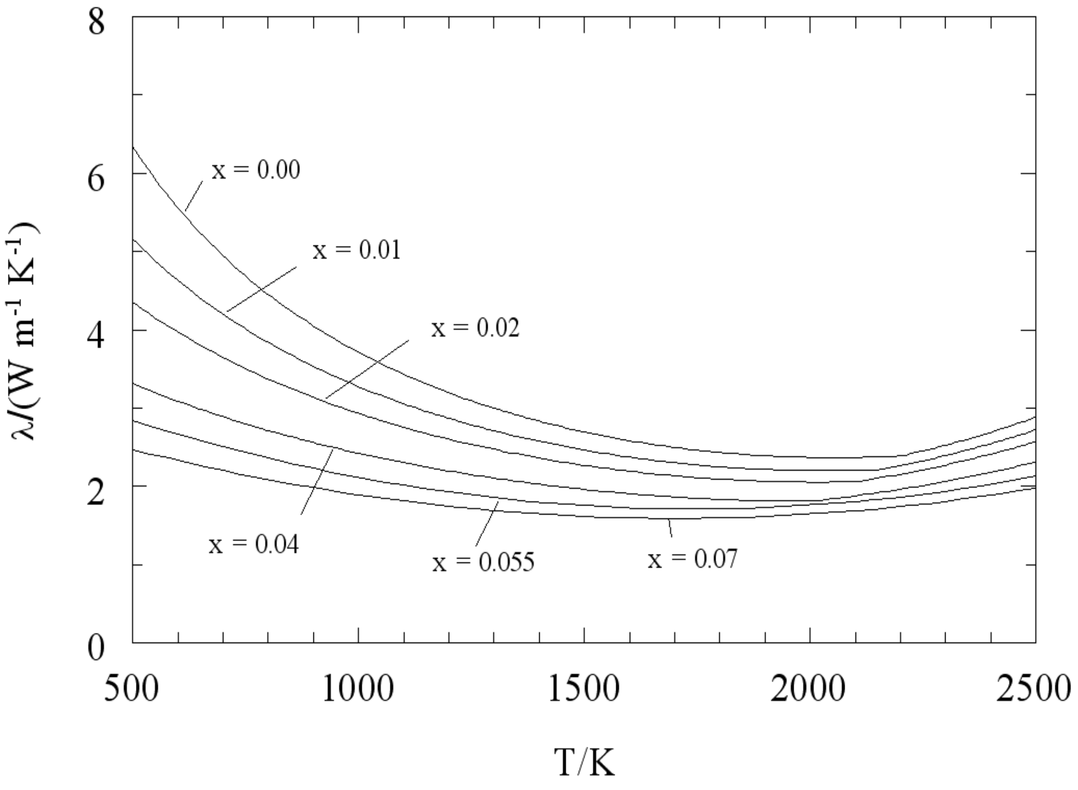
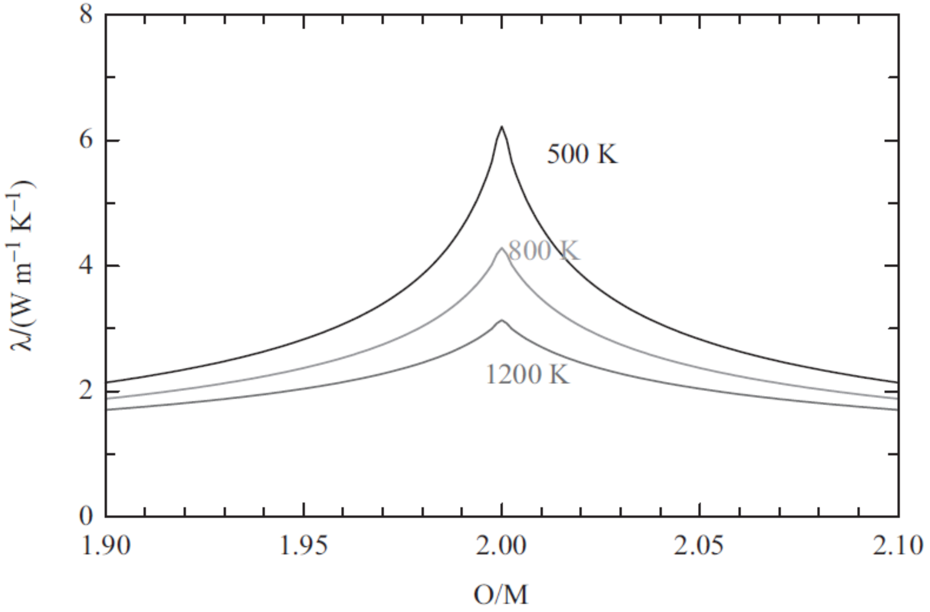
Nuclear Fuel: Mixed Oxide



The thermal conductivity as a function of *x* in $(U,Pu)O_{2-x}$



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Pros and cons of UO_2

Pro

- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO_2 …
- …

Con

- Low thermal conductivity
- Low fissile density
- …
- …
- …

3

Fuel fabrication

Fuel Fabrication

Provide a compact that :

- is mechanically stable
- has the required density
- has the right dimensions
- has the right microstructure
- is free from impurities

- no cracks, no chips
- > 95% theoretical density
- very small tolerance (µm‘s)
- grain size
- Neutron poisons, halides

Fuel Fabrication: Typical specification

		Unit	Target value	
Outer diameter		mm	8.05 ± 0.01	
Pellet height		mm	8 ± 1	
Visual aspect		-	No cracks, no chips	
Pellet density		%TD	96 ± 1	
Grain size		µm	5-25	
Pore size	mean	µm	100	
	limit	µm	500	
	condition		< 10% of the pores > 100 µm	
Stoichiometry		O/M	2.000 ± 0.002	
Thermal stability		-	< 1.5% density change	
Impurities		ppm	B, Cd, Gd < 1	Co, N < 75
			F < 10	C, Ni, Ca < 100
			Cl < 15	Most other elements < 200
			Th < 60	
H_2O content		ppm	< 100	

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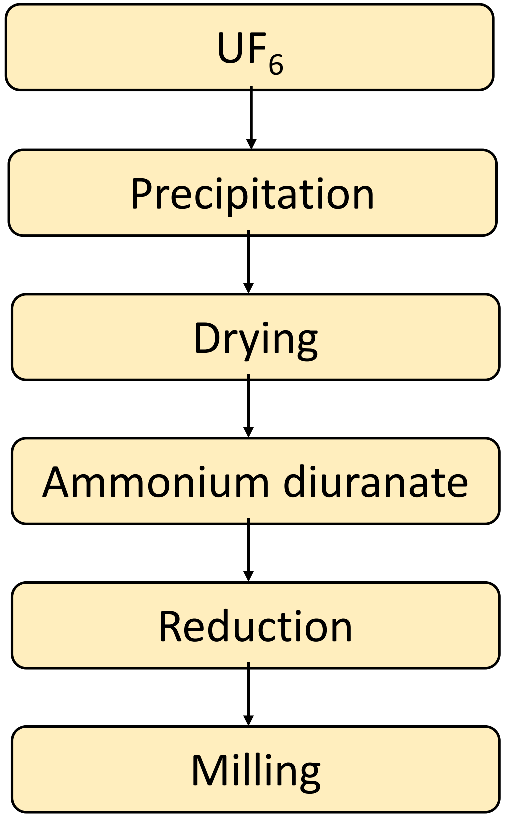
Fuel Fabrication: Powder preparation

The ADU process: aqueous process



$$2UF_6 + 7H_2O + 2NH_3 \rightarrow (NH_4)_2U_2O_7 + 12HF$$

$$(NH_4)_2U_2O_7$$

$$(NH_4)_2U_2O_7 \rightarrow U_3O_8 \rightarrow UO_2$$

Fuel Fabrication: Powder preparation

The AUC process: aqueous process

UF_6 → Precipitation → Drying → Ammonium uranyl carbonate → Reduction → Milling

$$UF_6 + 5H_2O + 10NH_3 + 3\,CO_2 \rightarrow (NH_4)_4UO_2(CO_3)_3 + 6NH_4F$$

$$(NH_4)_4UO_2(CO_3)_3$$

$$(NH_4)_4UO_2(CO_3)_3 + H_2 \rightarrow UO_2 + 3CO_2 + 4NH_3 + 3H_2O$$

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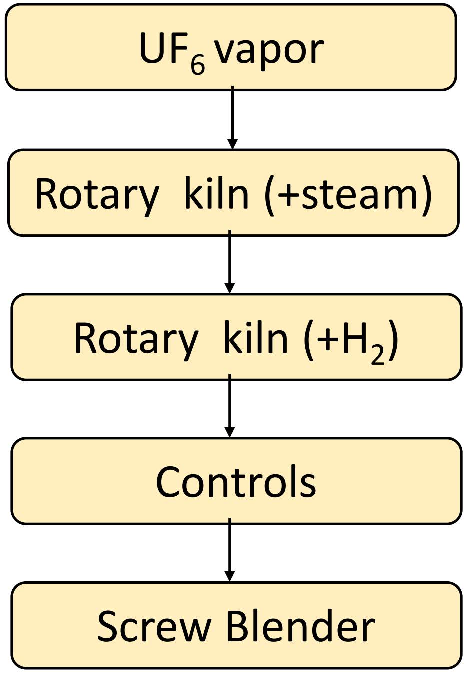
Fuel Fabrication: Powder preparation

The IDR process: integral dry route



$$UF_6 + 2H_2O\ (vap) \rightarrow UO_2F_2(sol) + 4HF$$

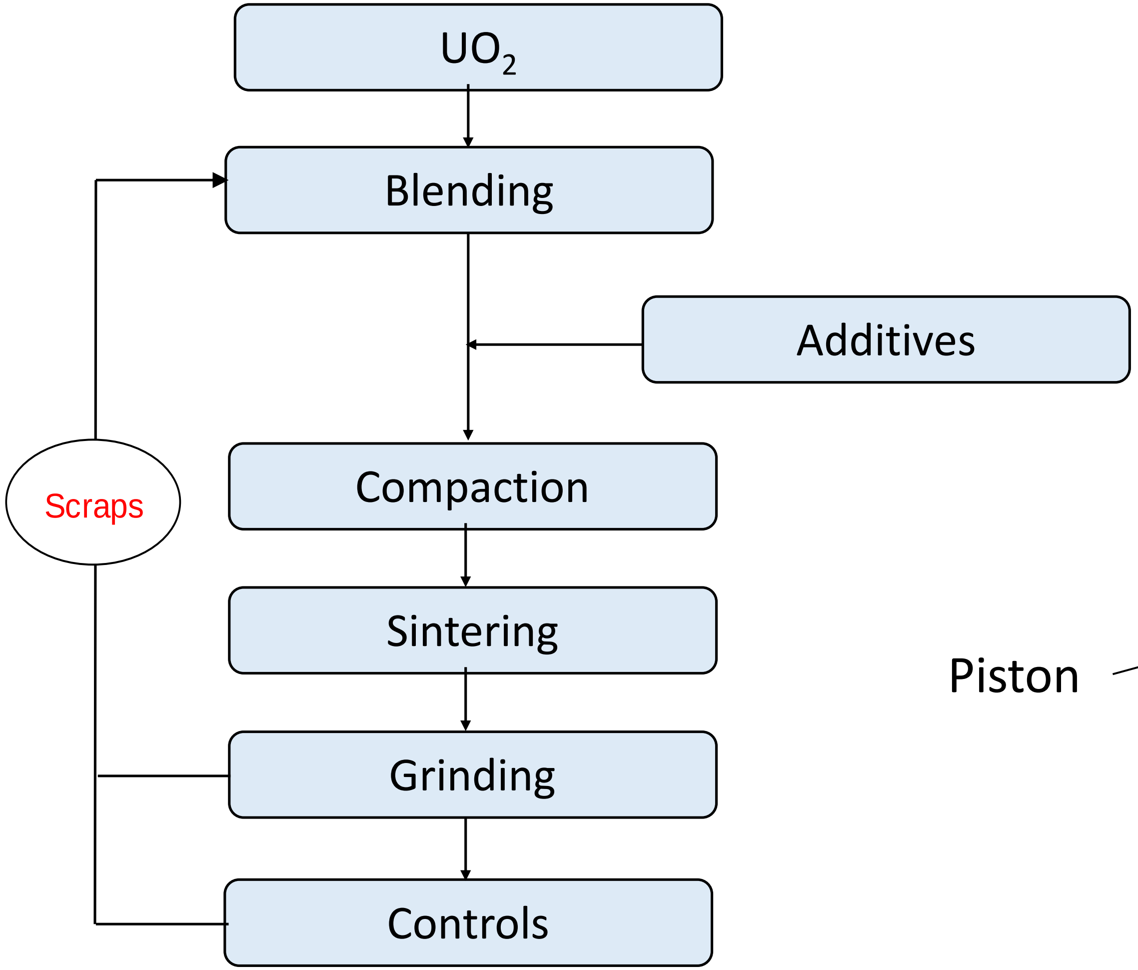
$$UO_2F_2(sol) + H_2O \rightarrow UO_3 + 2HF$$

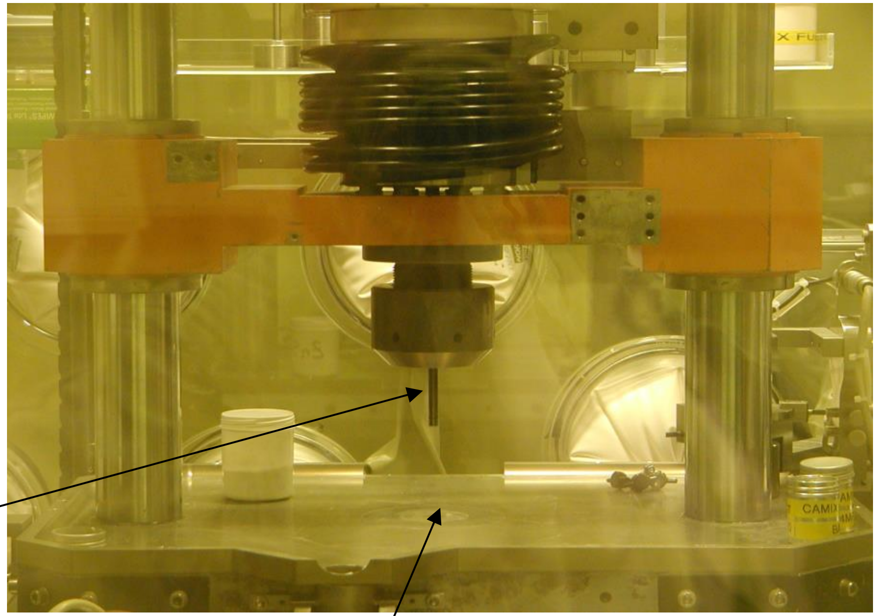
$$UO_3 + H_2 \rightarrow UO_2 + H_2O$$

Fuel Fabrication: Powder preparation

	ADU	AUC	IDR
Specific surface (m^2/g)	2.8-3.2	5.0-6.0	2.5-3.0
Raw density (g/cm^3)	1.5	2.0-2.3	0.7
Tap denisty (g/cm^3)	2.4-2.8	2.6-3.0	1.65
Mean size (microns)	0.4-1.0	8	2.4
Morphology	spheroids	Porous aggl.	dendrites
O/U ratio	2.03-2.17	2.06	2.05
Fluor (ppm)	30-50	30-70	<25
Carbon (ppm)	40-200	120	20
Iron (ppm)	70	10-20	10
Boron (ppm)	0.2	0.1	<0.05

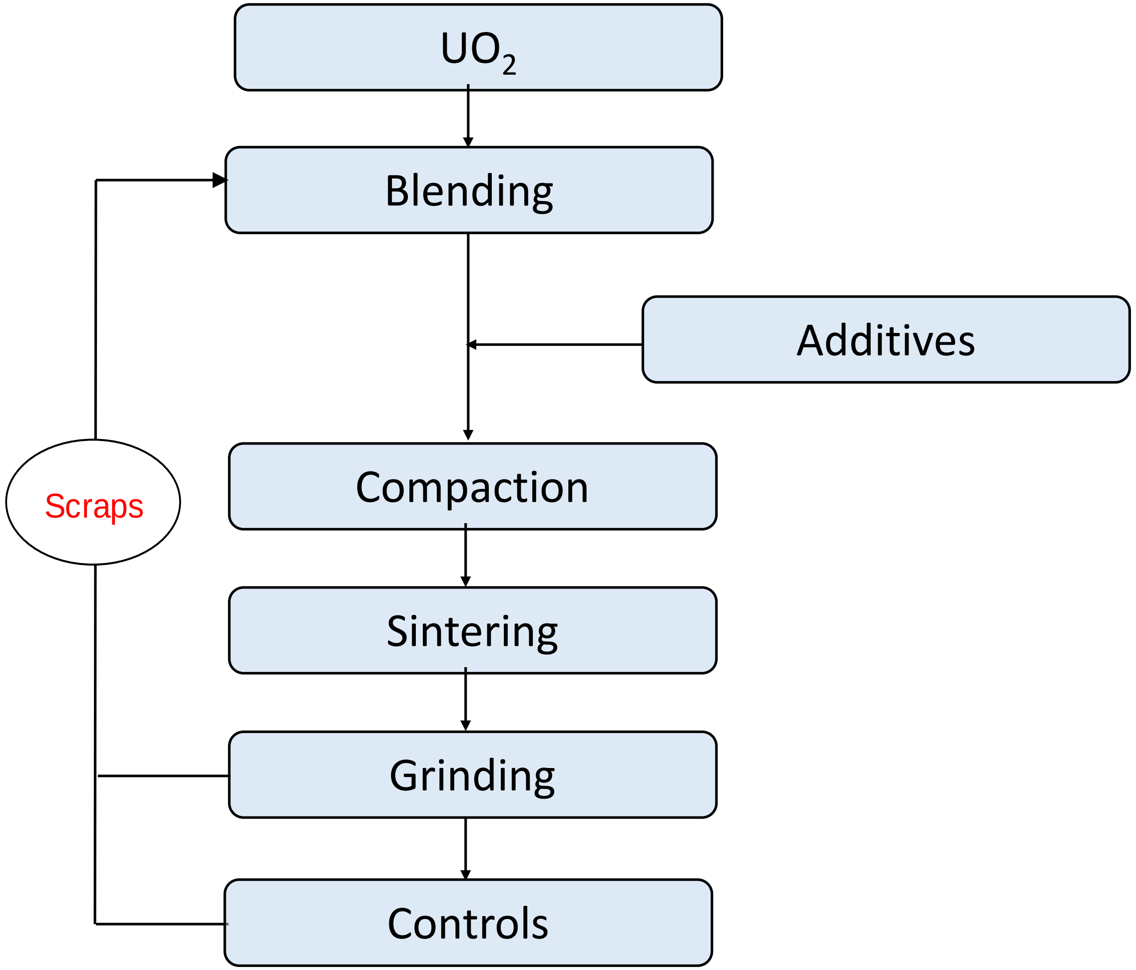
Fuel Fabrication: Powder preparation

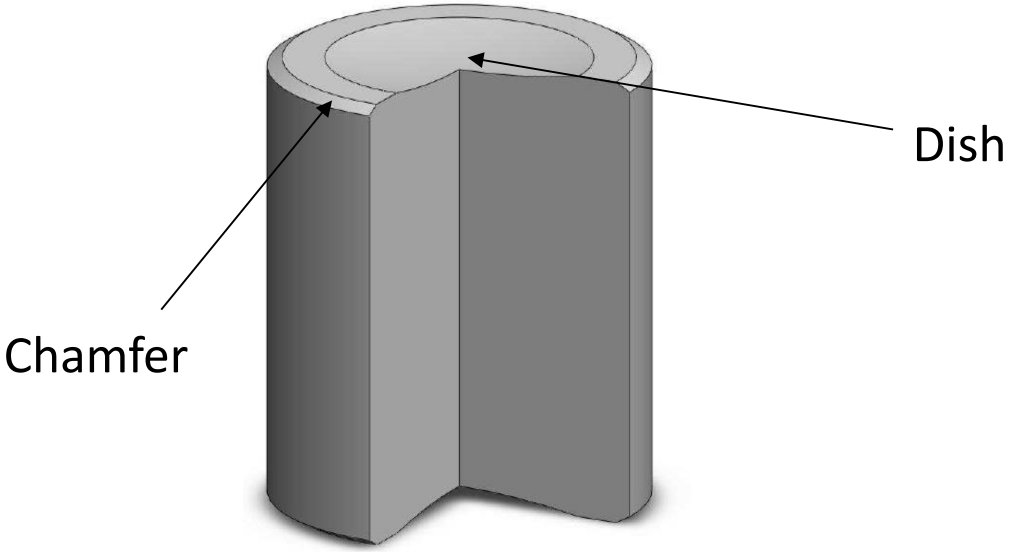




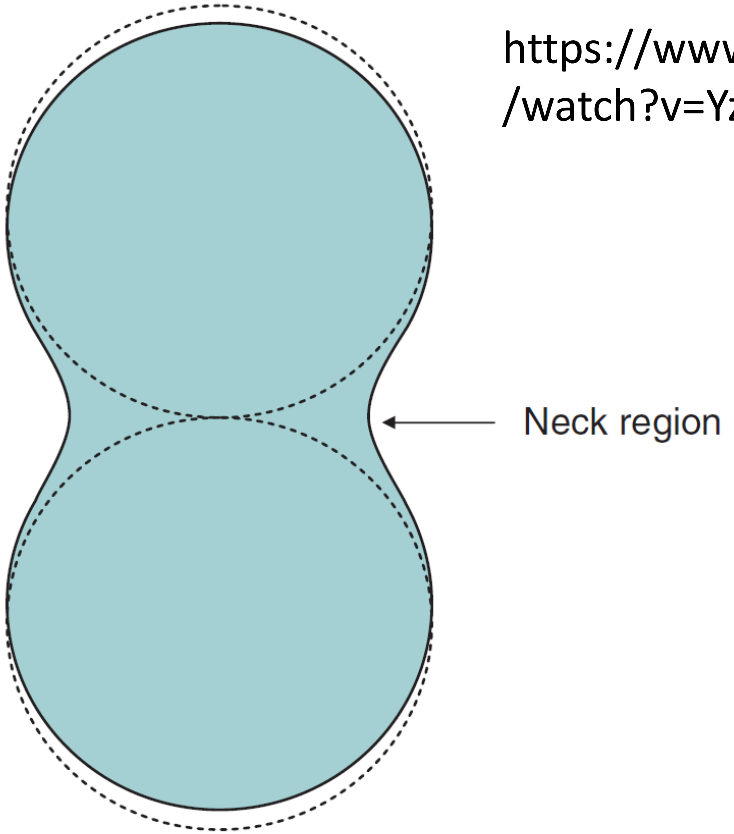
Source: European Communities

Fuel Fabrication: Powder preparation





Fuel Fabrication: Sintering

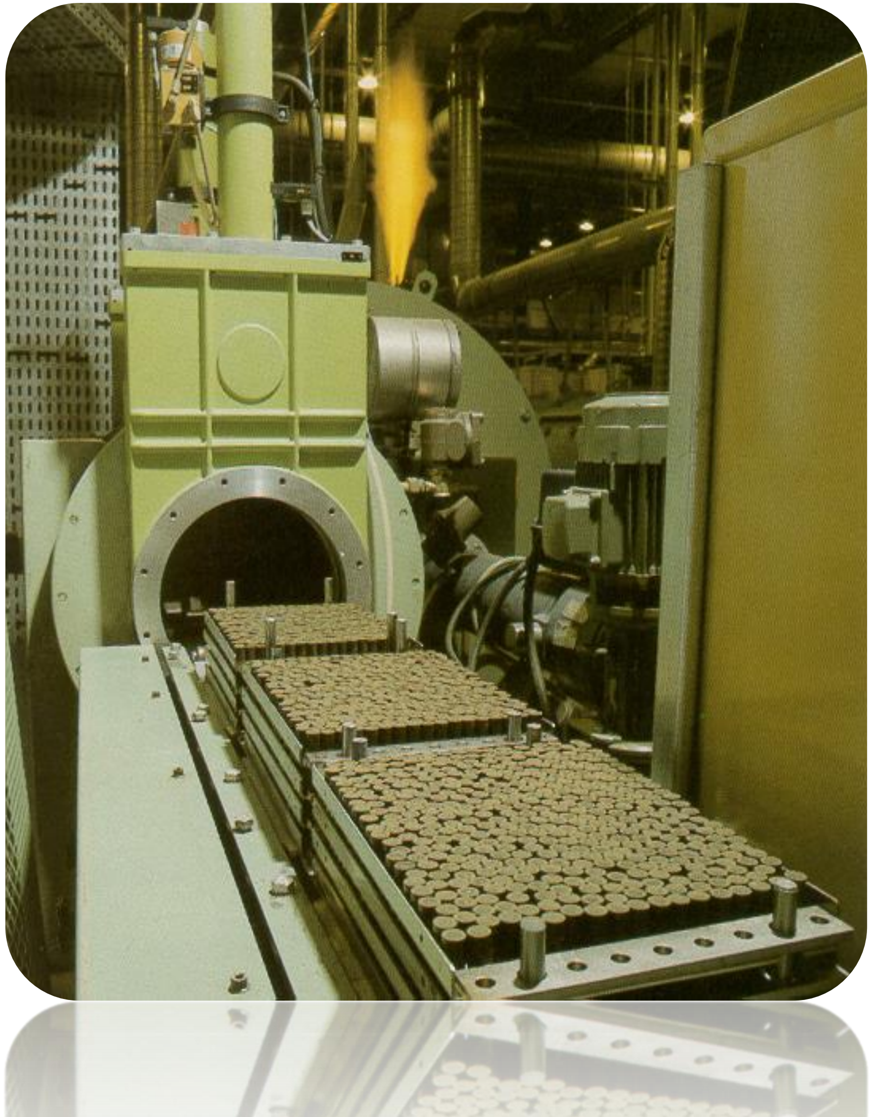


https://www.youtube.com/watch?v=Yz2FgkjFjHY

Schematic representation of sintering. The dotted lines indicated the as-fabricated particles, the solid lines the necking as a result of mass transport.

- Transforms a compacted powder to a dense object
- Homogenizes composition (partially)
- Based on thermally activated diffusion
- Principally surface diffusion

Fuel Fabrication: Sintering

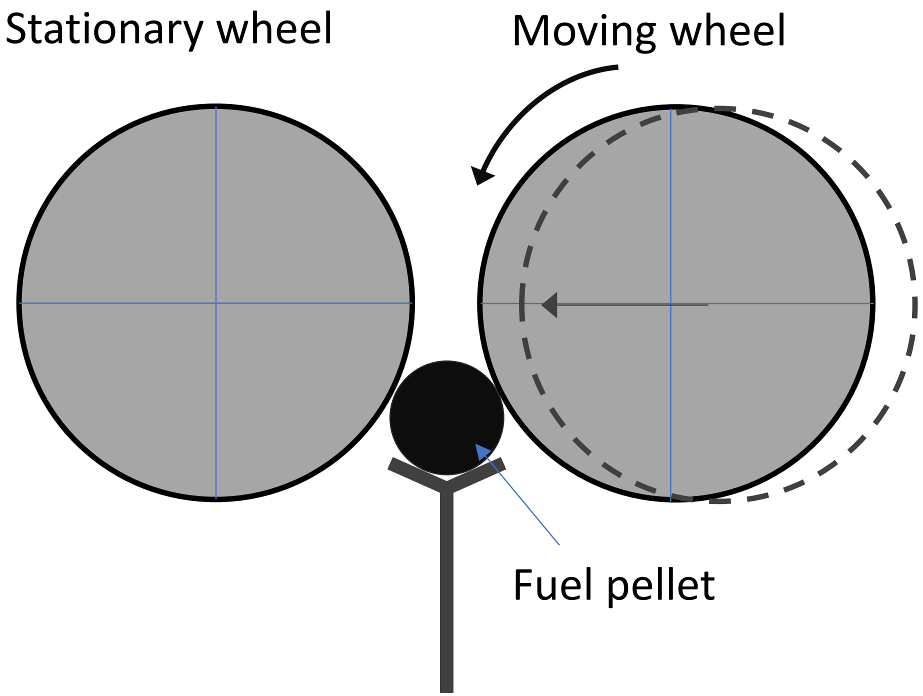


- **Atmosphere:** **Ar+H_2**
- **Temperature:** **1600-1700 °C**
- **Duration:** **6 hours at max**

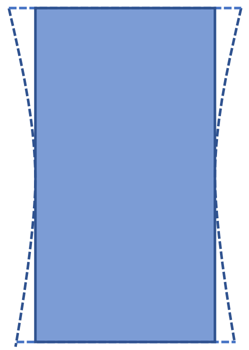
Density changes from
$\rho_{green} \approx 50$ % TD to
$\rho_{final} \approx 95$ % TD

TD = theoretical density

Fuel Fabrication: Grinding



- Pressed pellets have an hours glass shape → Needs to be corrected
- To meet the specification and tolerances

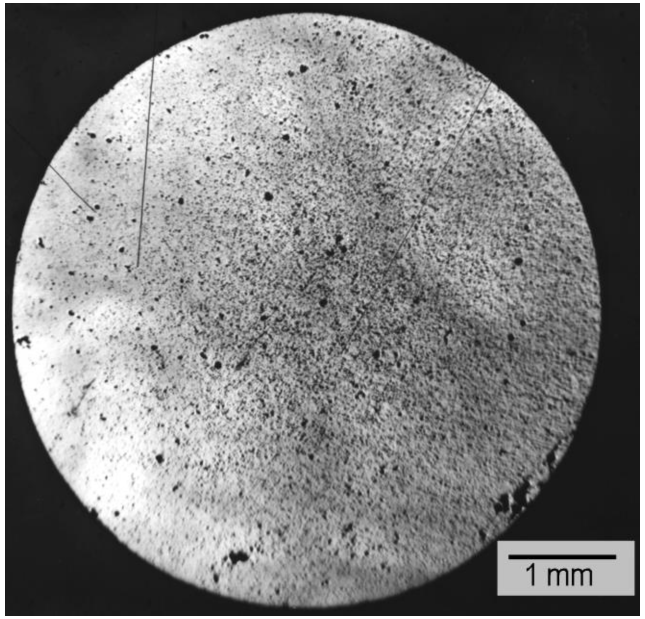


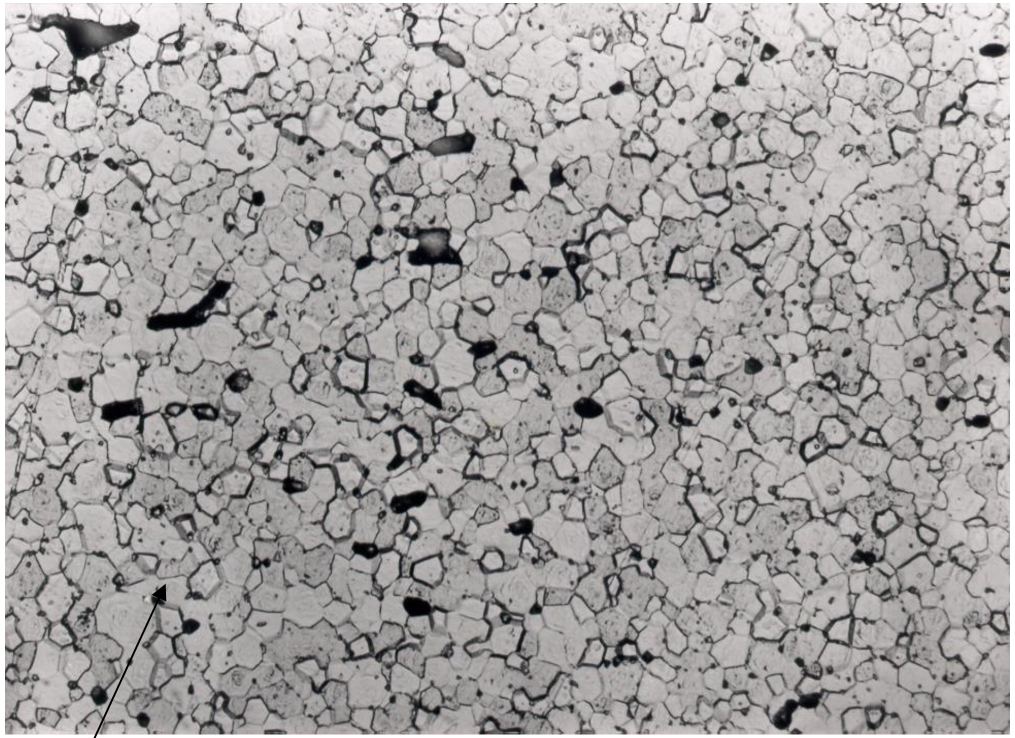
Fuel Fabrication: Controls

- Dimensions
- Density
- O/U
- Microstructure
- Impurities
- Stability

- Visual, Optical
- Archimedes, geometric
- X-ray, thermal gravimetric analysis
- Ceramography
- Chemical analysis (ICP-MS)
- Re-sintering test

Fuel Fabrication: Microstructure

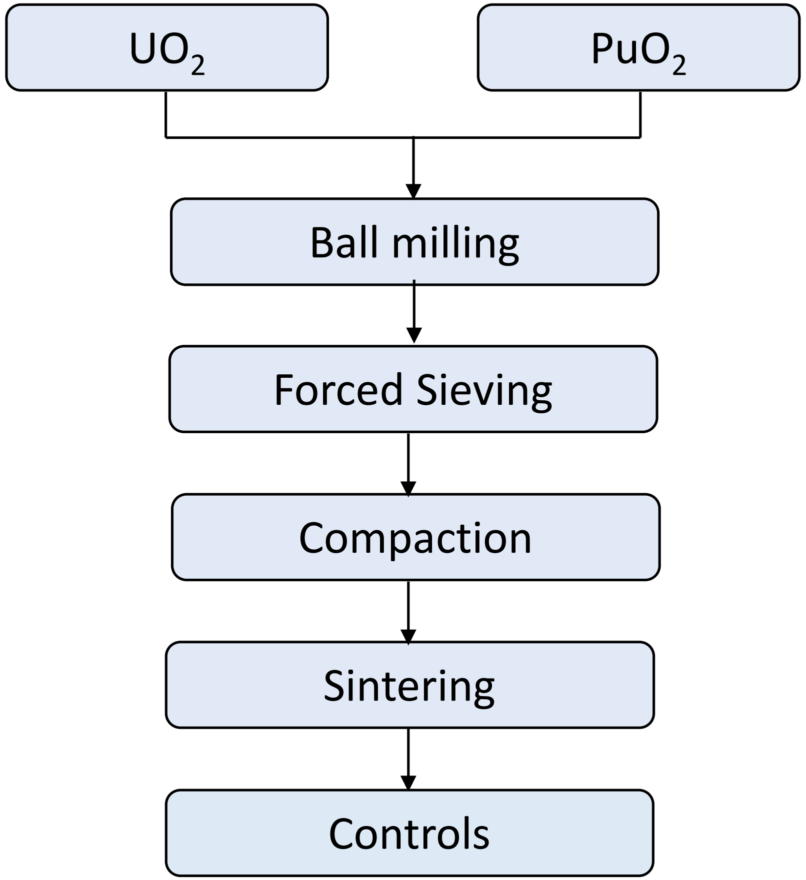


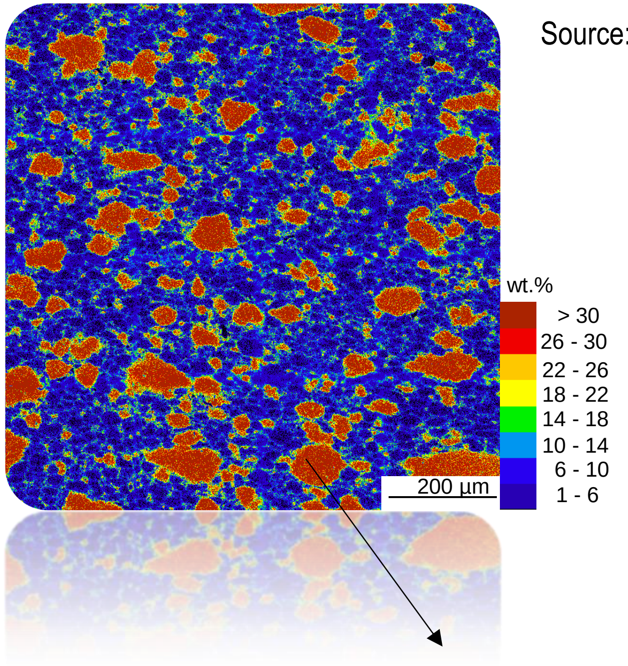


Grain size UO_2: 8 micro-meter

Source: European Communities

MOX Fabrication: Traditional



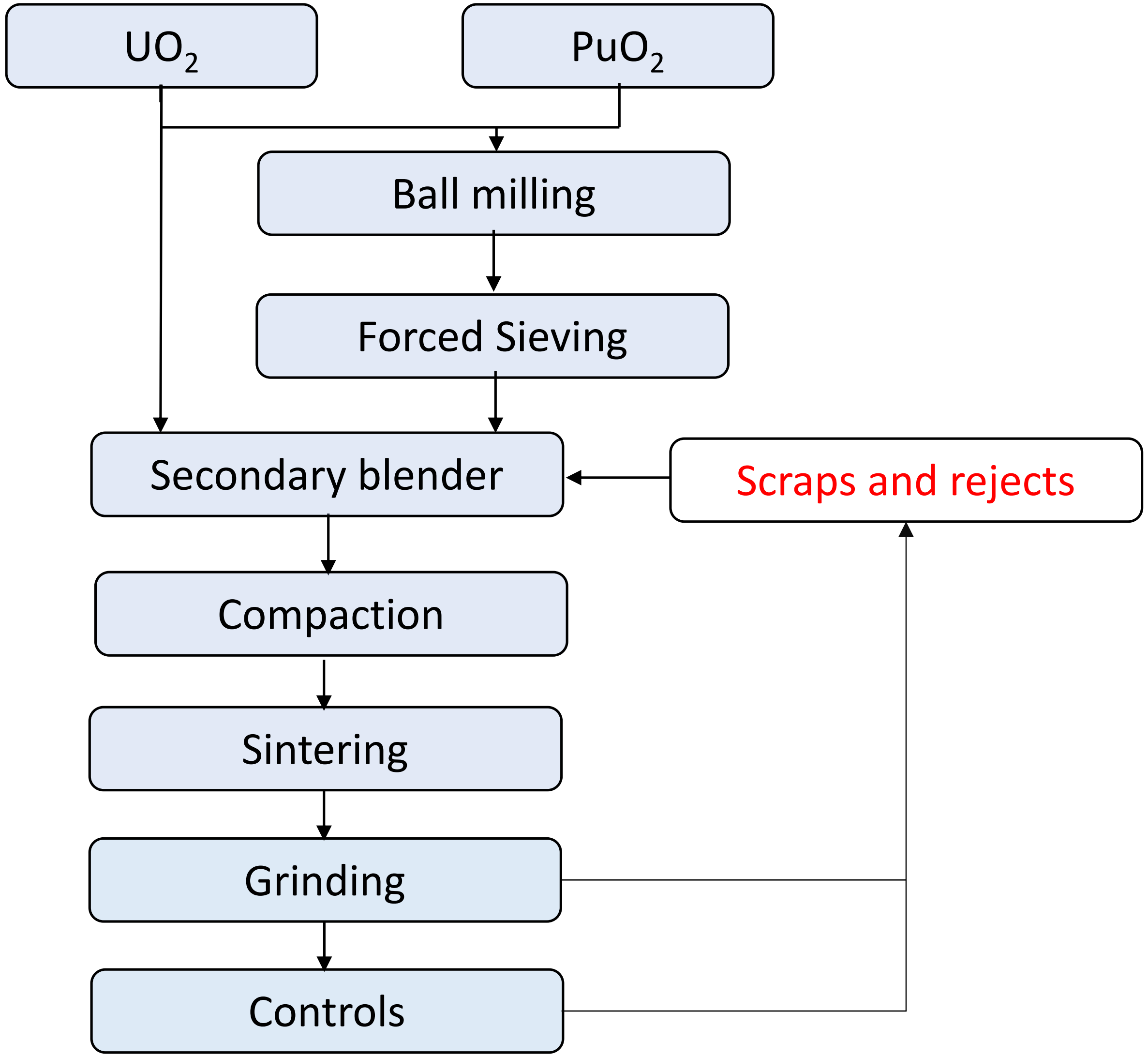


Source: European Communities

Pu-rich islands

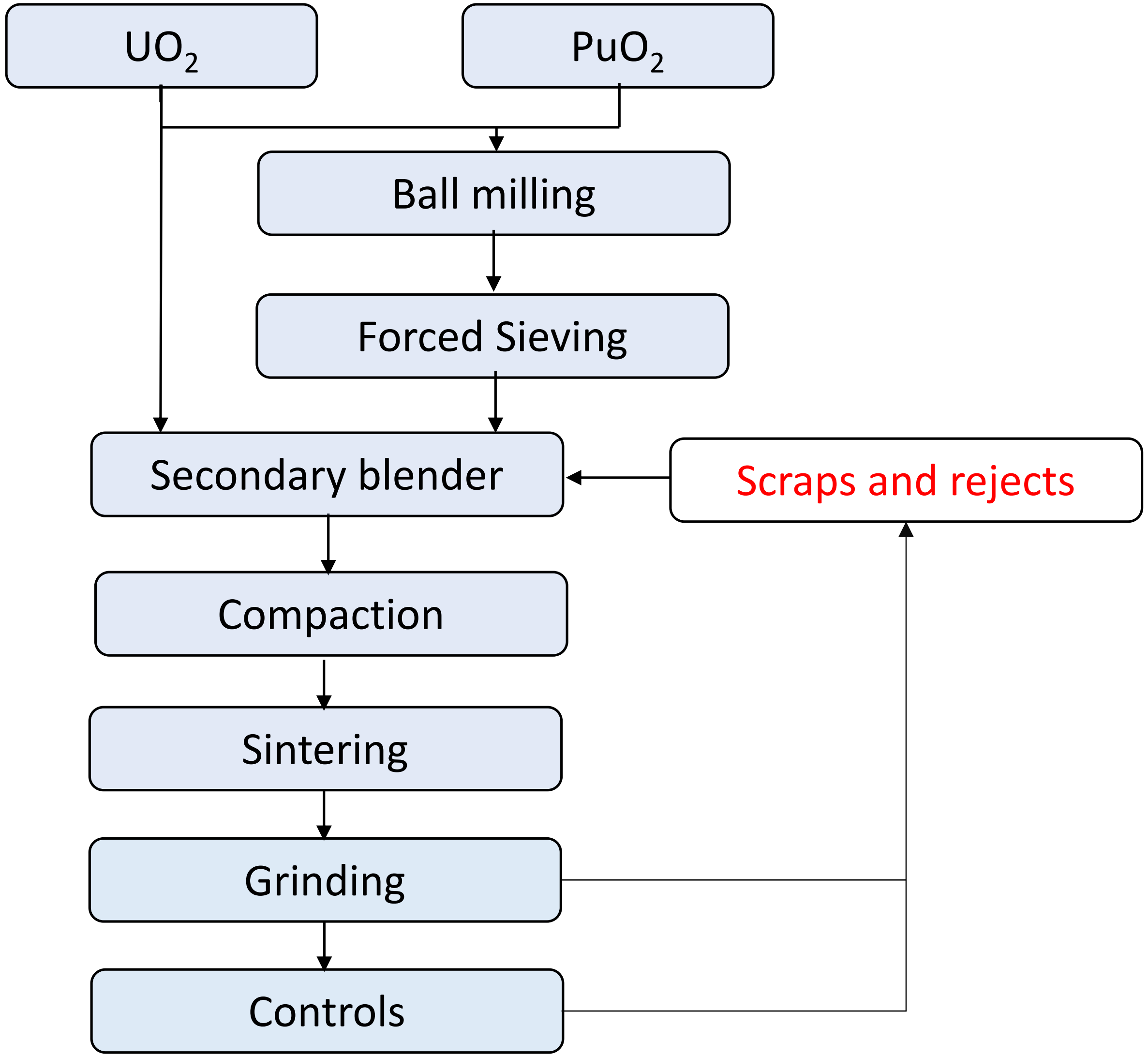
- Hot spots
- Local high burnup
- Insoluble

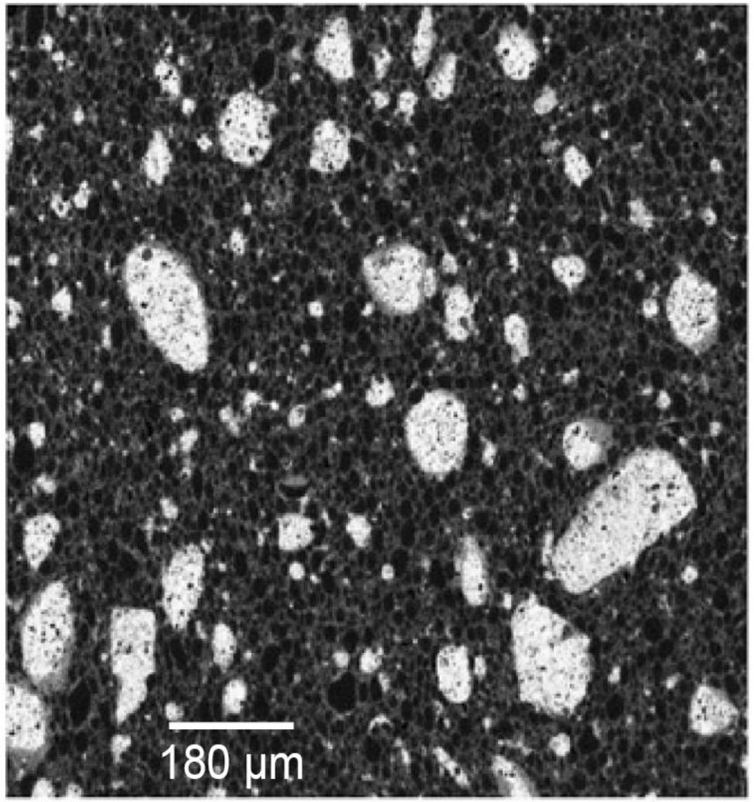
MOX Fabrication: MIMAS - industrial



Source: ORANO

MOX Fabrication: MIMAS - industrial

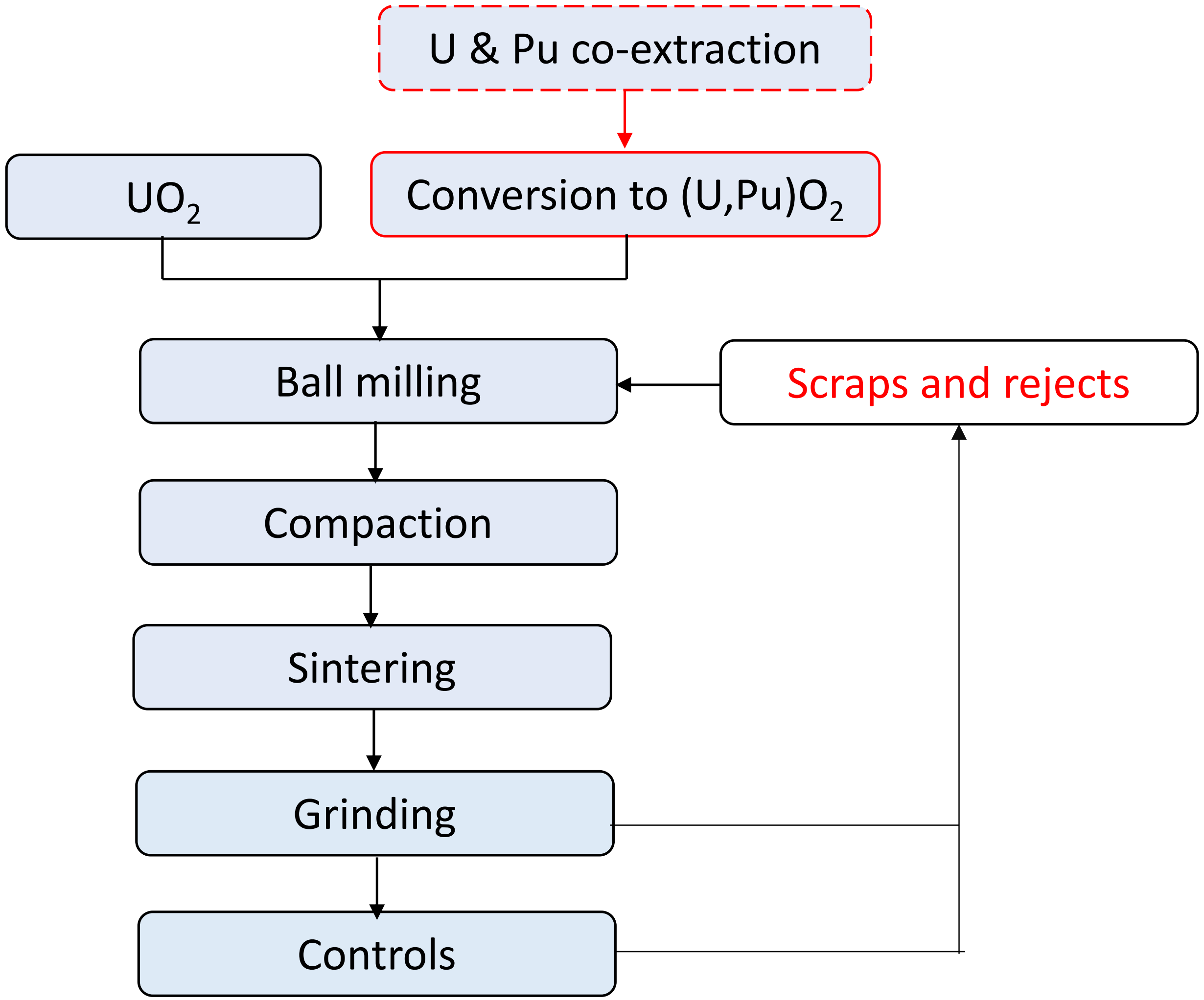




Source: Oudinet et al. J. Nucl. Mater., 375 (2008) 86

European Commission

MOX Fabrication: JMOX



Pros and cons of UO_2

Pro

- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO_2
- Straightforward fabrication
- …

Con

- Low thermal conductivity
- Low fissile density
- Dirty process (dust)
- Many steps (MOX)
- Proliferation risk (MOX)

4

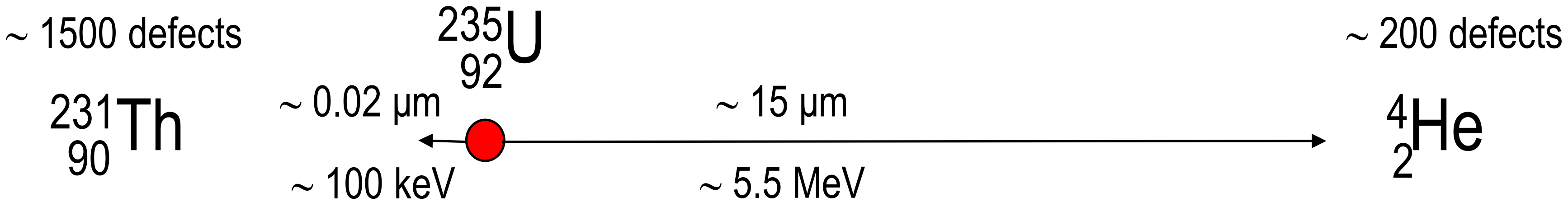
Radiation effects

European Commission

Radiation effects: Alpha decay

$^{235}U \longrightarrow {}^{231}Th + {}^{4}He$

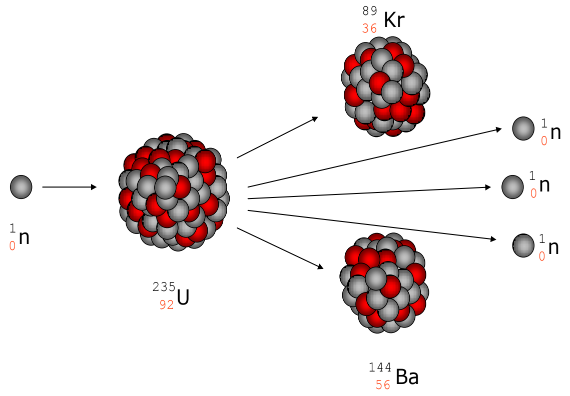
$E \approx 5.5\ MeV$

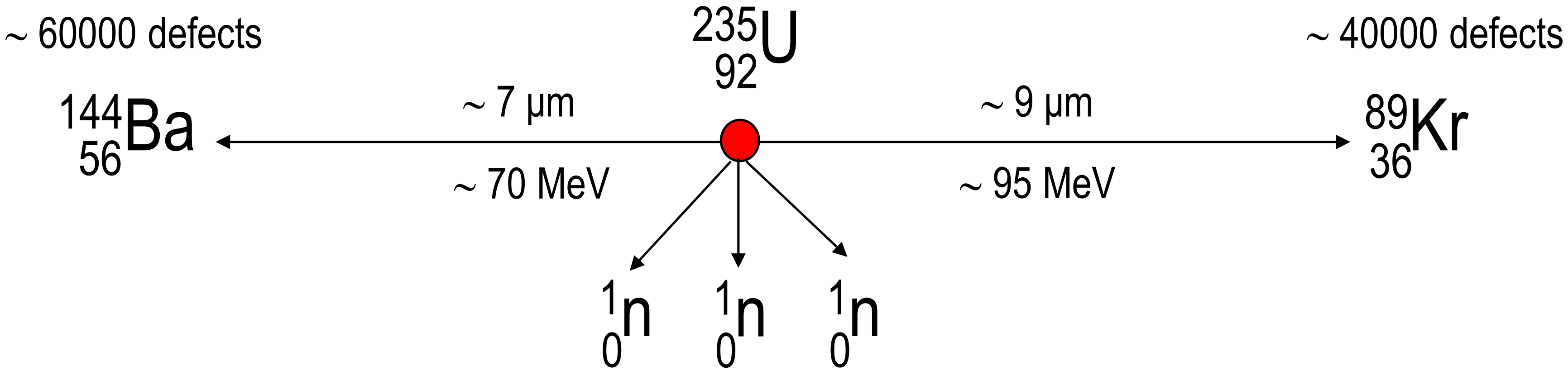


Radiation effects: Fission

$^{235}U + n \longrightarrow X_1 + X_2 + 2\tfrac{1}{2}n$

$E \approx 200$ MeV





Radiation effects: Energy loss

Particles passing through matter loose energy via two interaction processes:

- **Nuclear Collisions** (Rutherford scattering): direct collision with atoms (nucleus)

- **Electronic Collisions** (Born scattering): dissipating the energy to the electrons

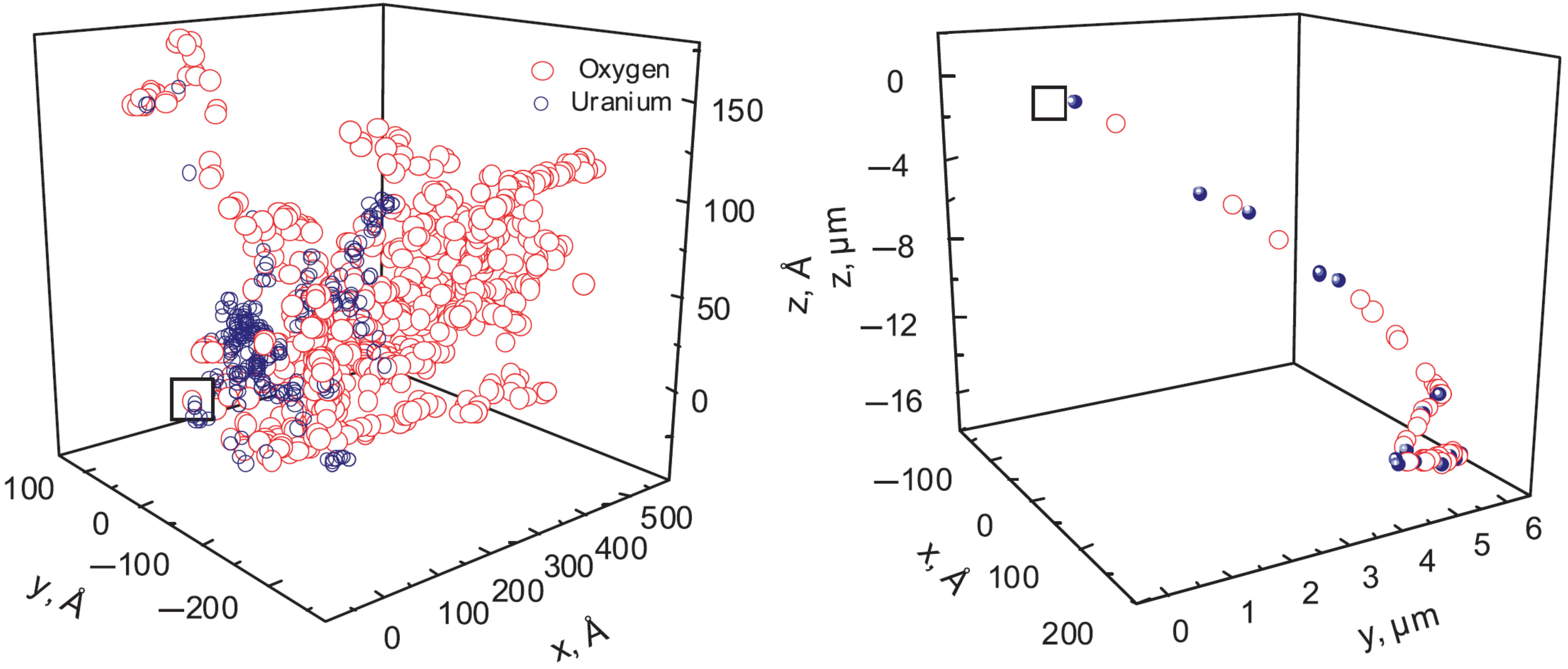
Radiation effects: Energy loss

Radiation sources to be considered

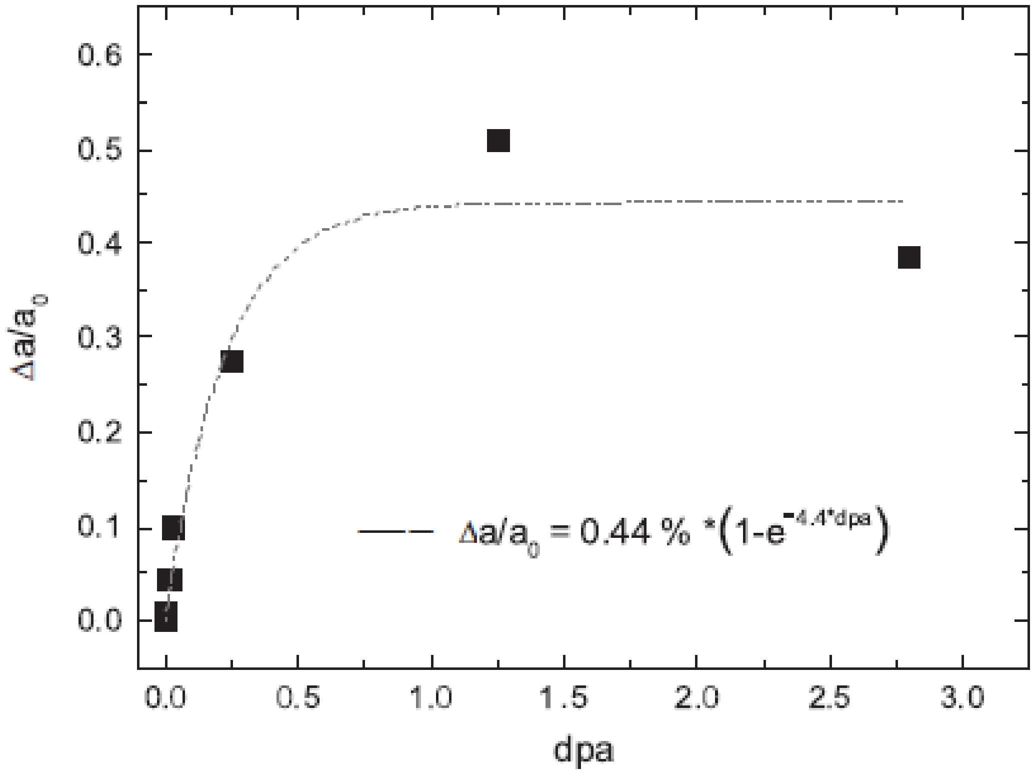
- Neutrons → *minimum displacement energy ~ 100 eV*
  in LWR's neutrons are moderated (< 1eV)
- β-decay → *electronic scattering*
- α-decay
- Fission fragments

→ *Atomic displacements*

Radiation effects: Alpha decay



Radiation effects: Alpha decay



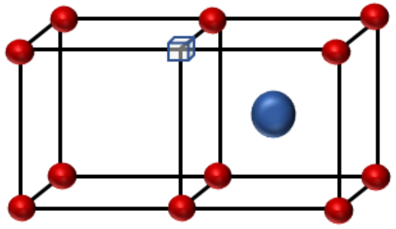
- Radiation defects lead to expansion of the lattice
- Equilibrium between creation and annihilation occurs after about 1 dpa

Source: Wiss & Konings (2013), In: Properties of Fluorite Structure Materials. (P. Vajda and M. Costandini, Eds.). Nova Publishers 2013

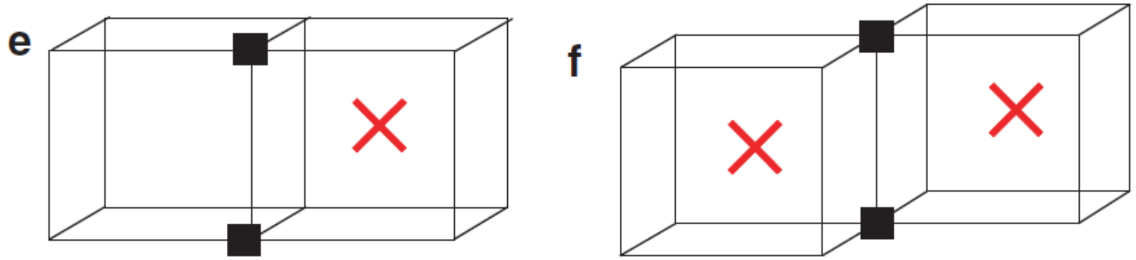
European Commission

Radiation effects: Lattice defects

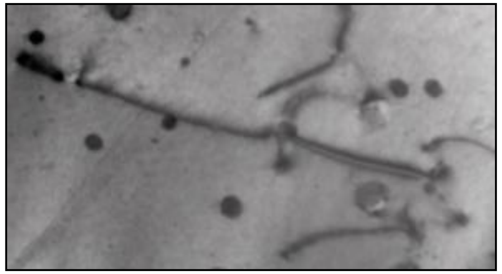
- Point defects

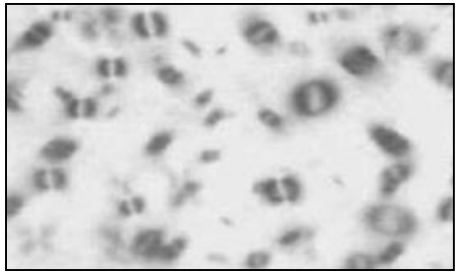


- Extended defects

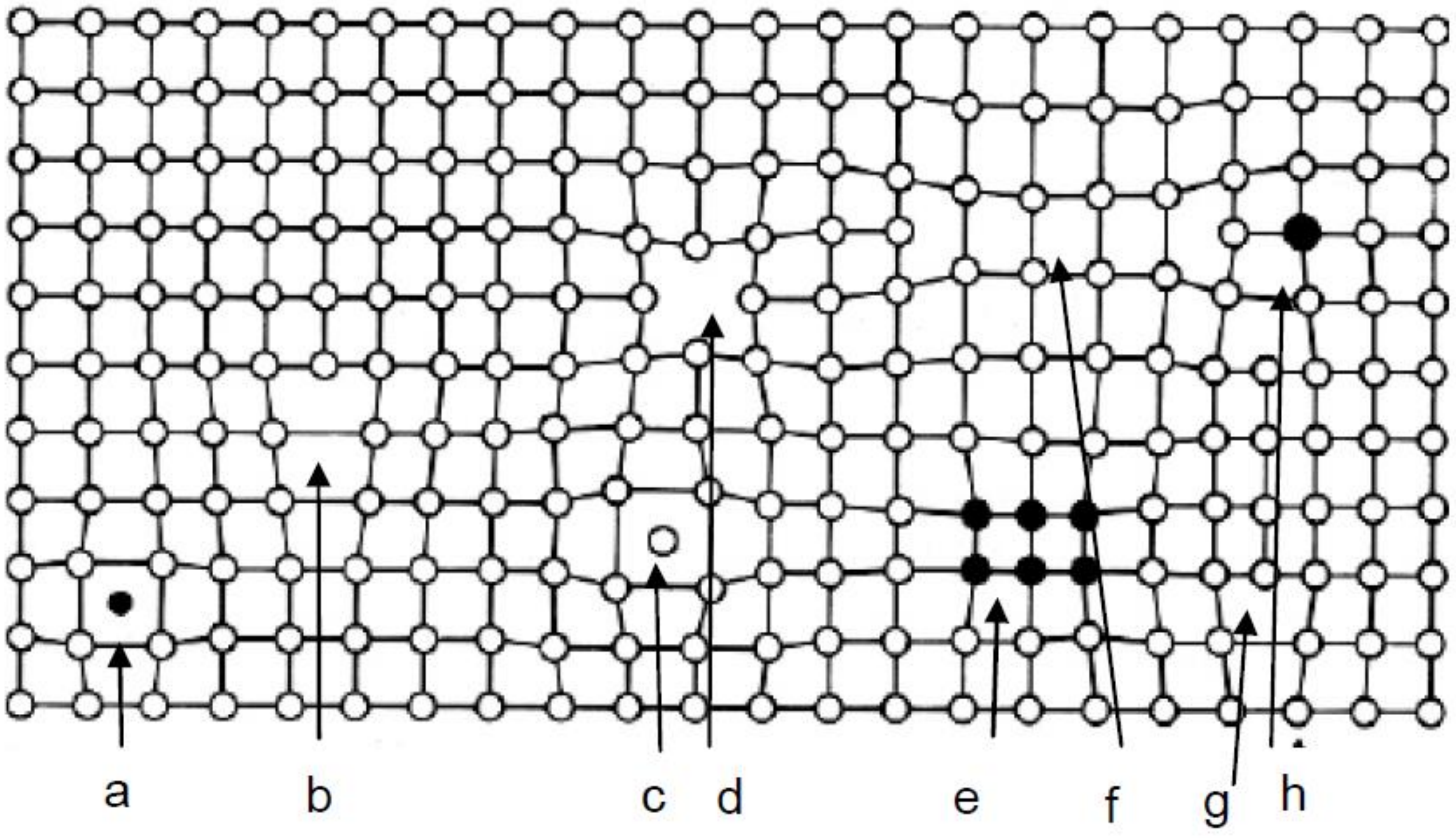


- Dislocations (line, loop)



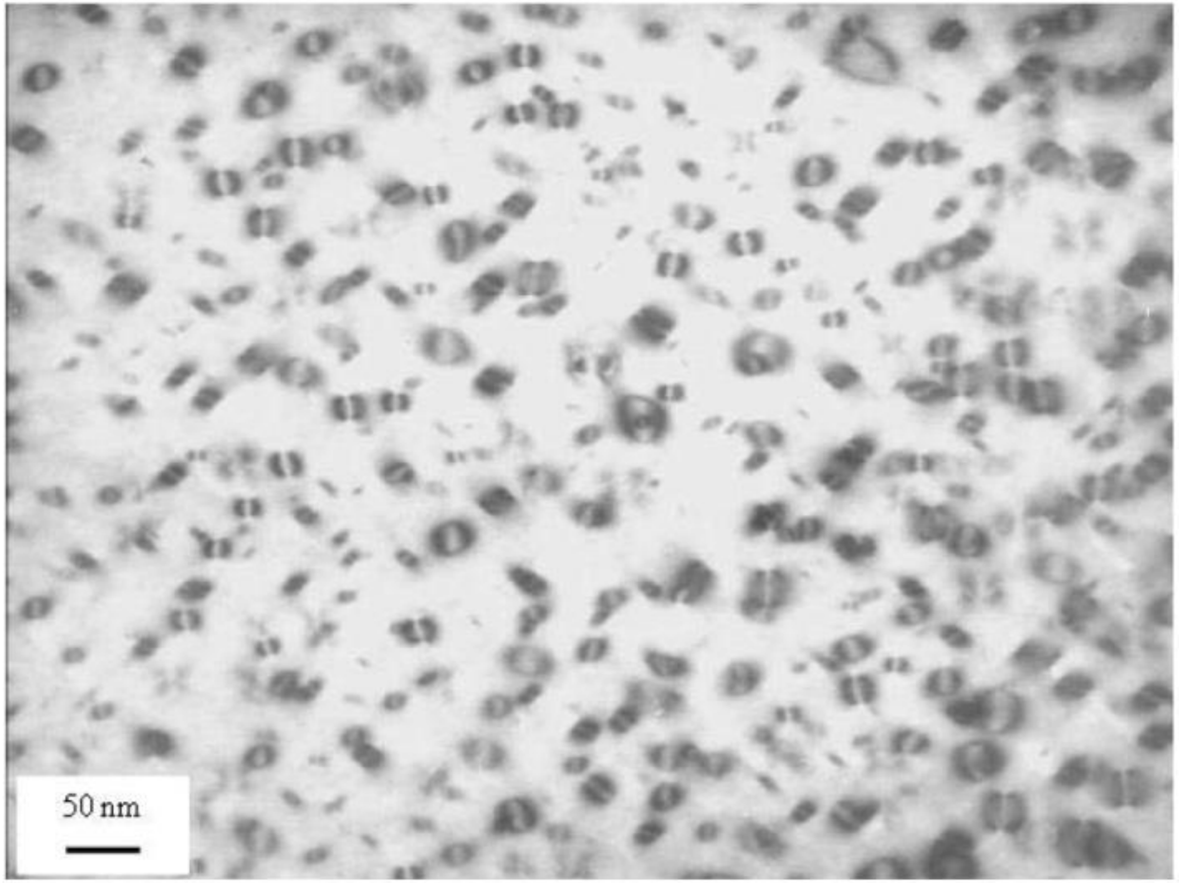


Radiation effects: Lattice defects



(a) interstitial impurity atom
(b) edge dislocation
(c) Self-interstitial atom
(d) Vacancy
(e) precipitate of impurity atoms
(f) vacancy-type dislocation loop
(g) interstitial-type dislocation loop
(h) Substitutional impurity atom

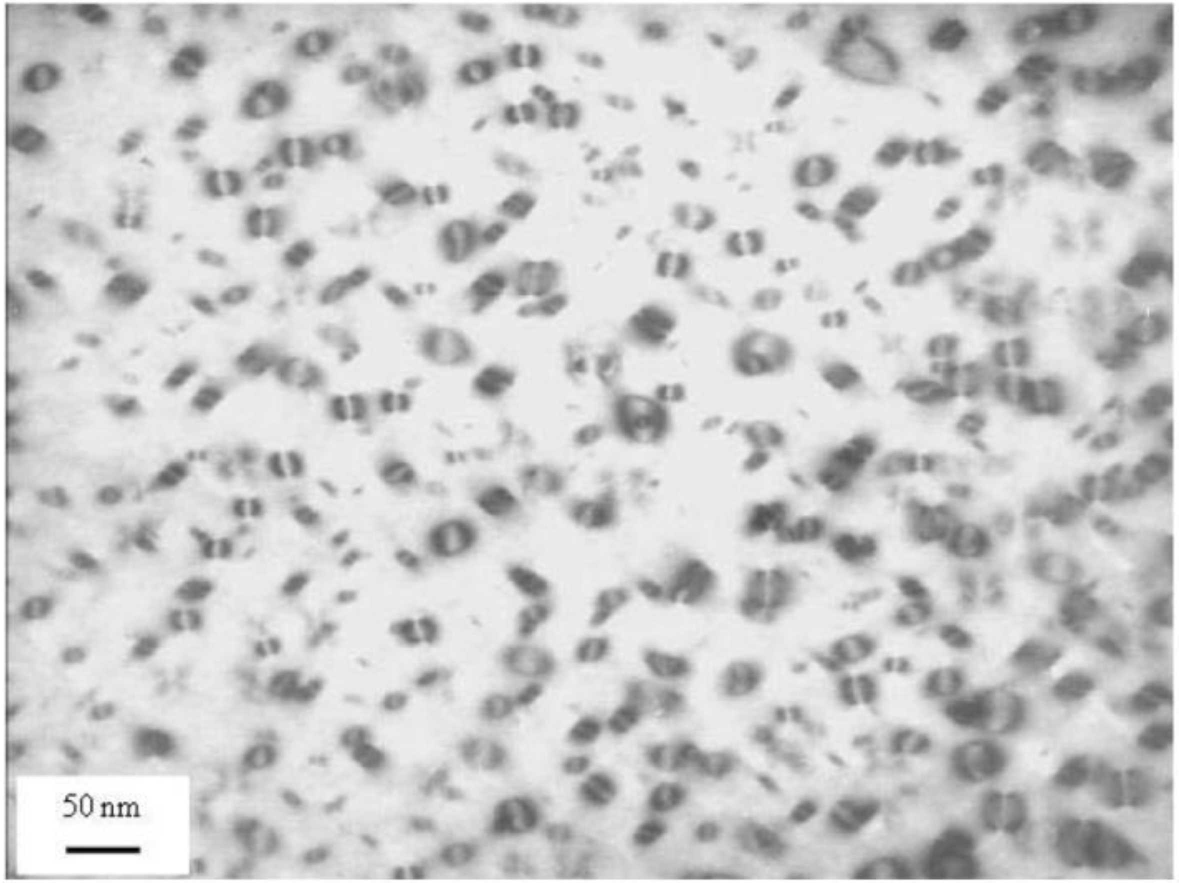
Radiation effects: Alpha decay



Source: European Communities

TEM micrograph of 10 wt% ^{233}U-doped UO_2 showing the presence of prismatic loops resulting from the alpha-damage

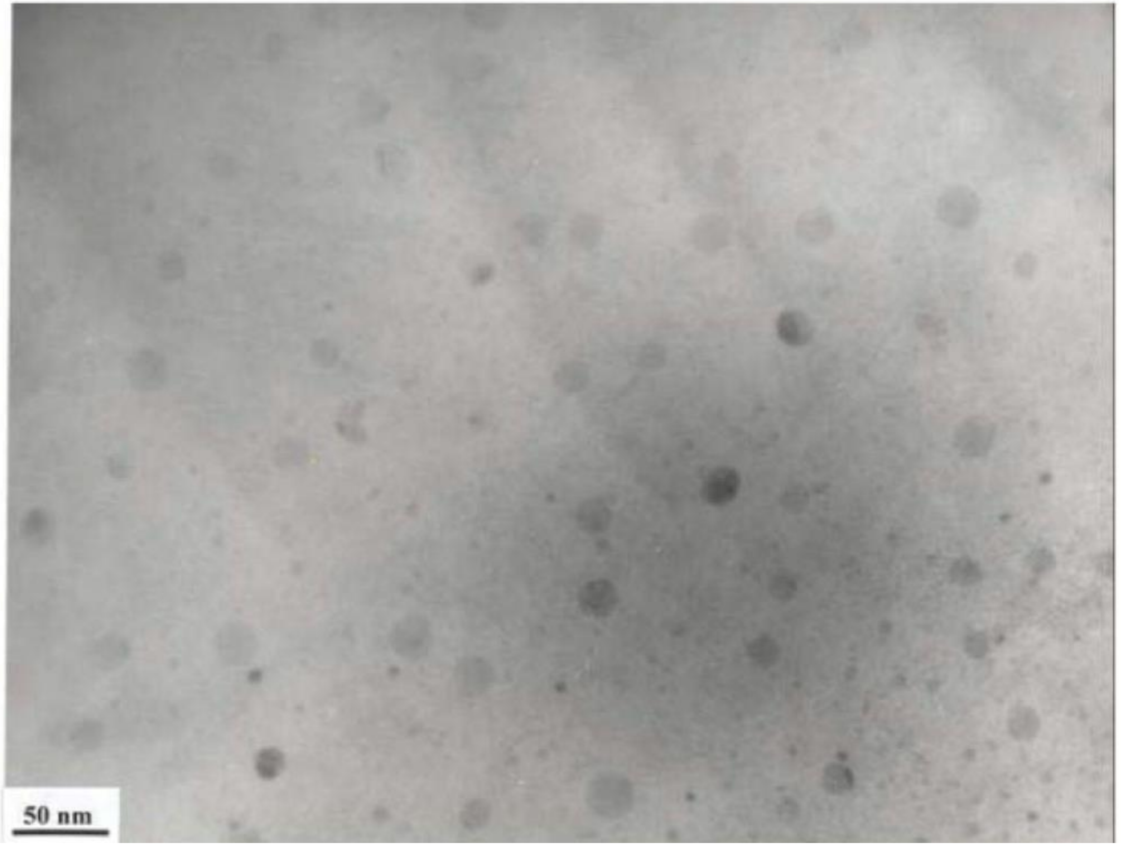
Radiation effects: Alpha decay



Source: European Communities

European Commission

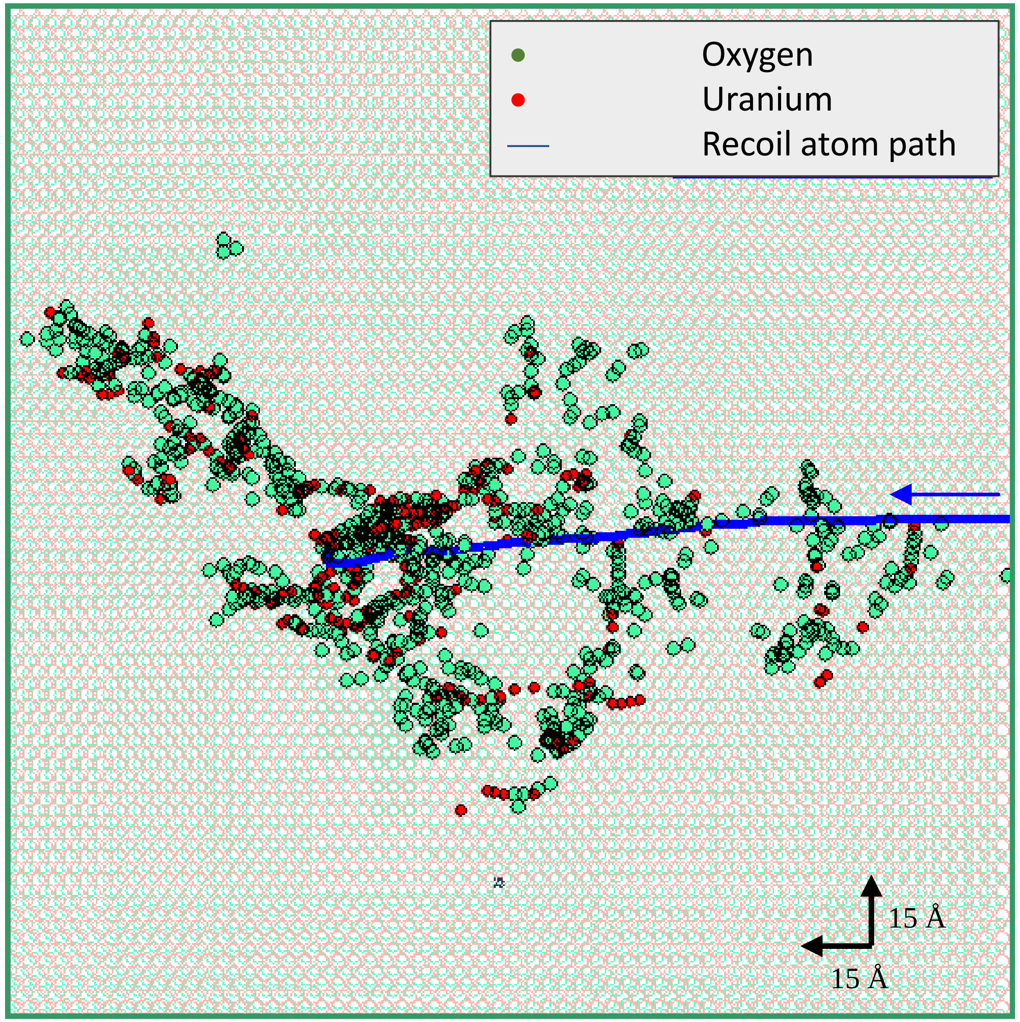
Radiation effects: Alpha decay



Source: European Communities

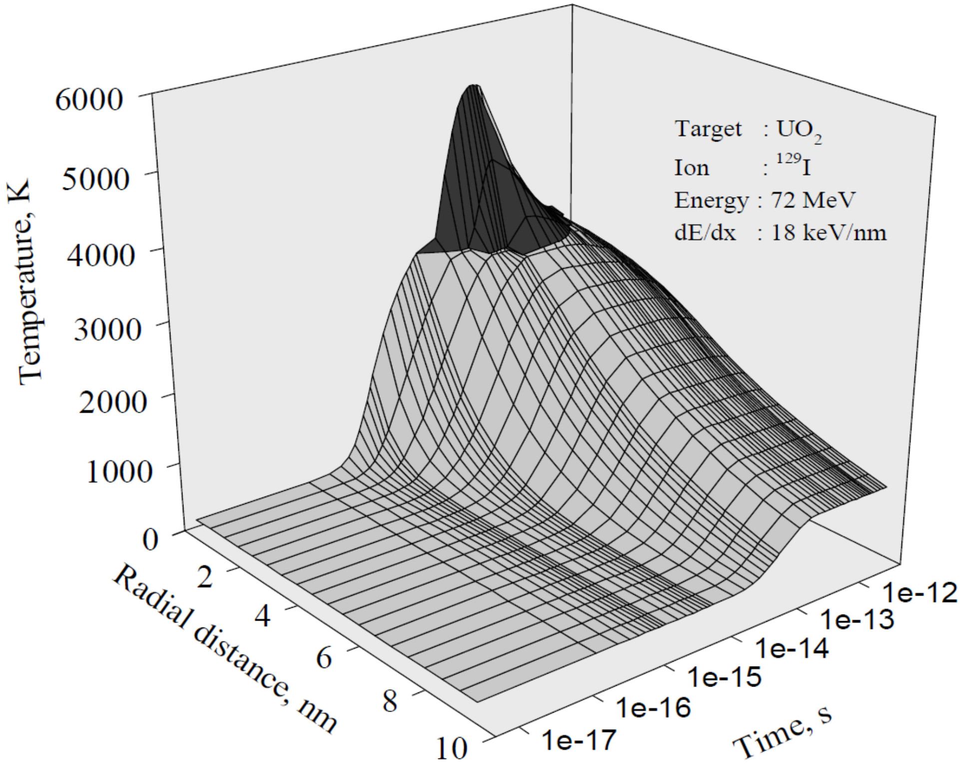
TEM micrograph of 10 wt% ^{233}U-doped UO_2 after annealing at 1100 K showing the presence of Helium gas bubbles

Radiation effects: Fission product recoil



European Commission

Radiation effects: Fisson product recoil

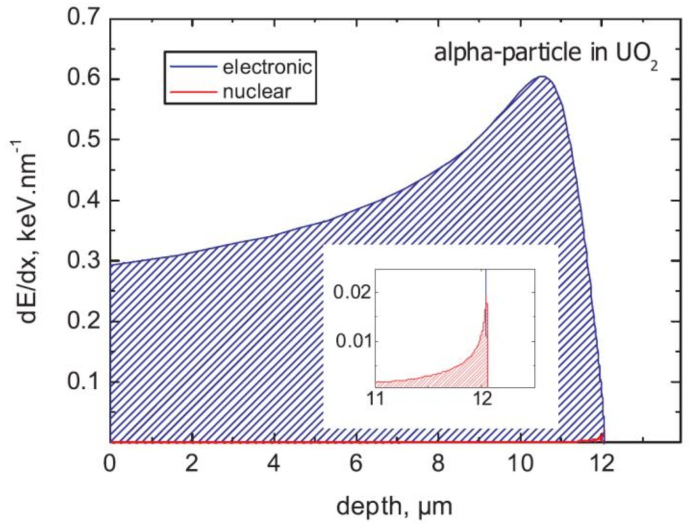


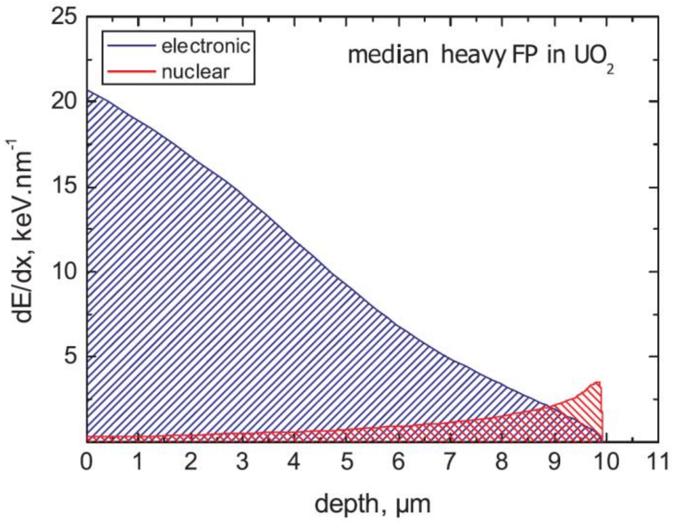
Calculated radial temperature distribution of a fission track in UO_2 as a function of time. Calculations were made for the first nm of the material.

Source: Wiss & Konings (2013), In: Properties of Fluorite Structure Materials. (P. Vajda and M. Costandini, Eds.). Nova Publishers 2013

European Commission

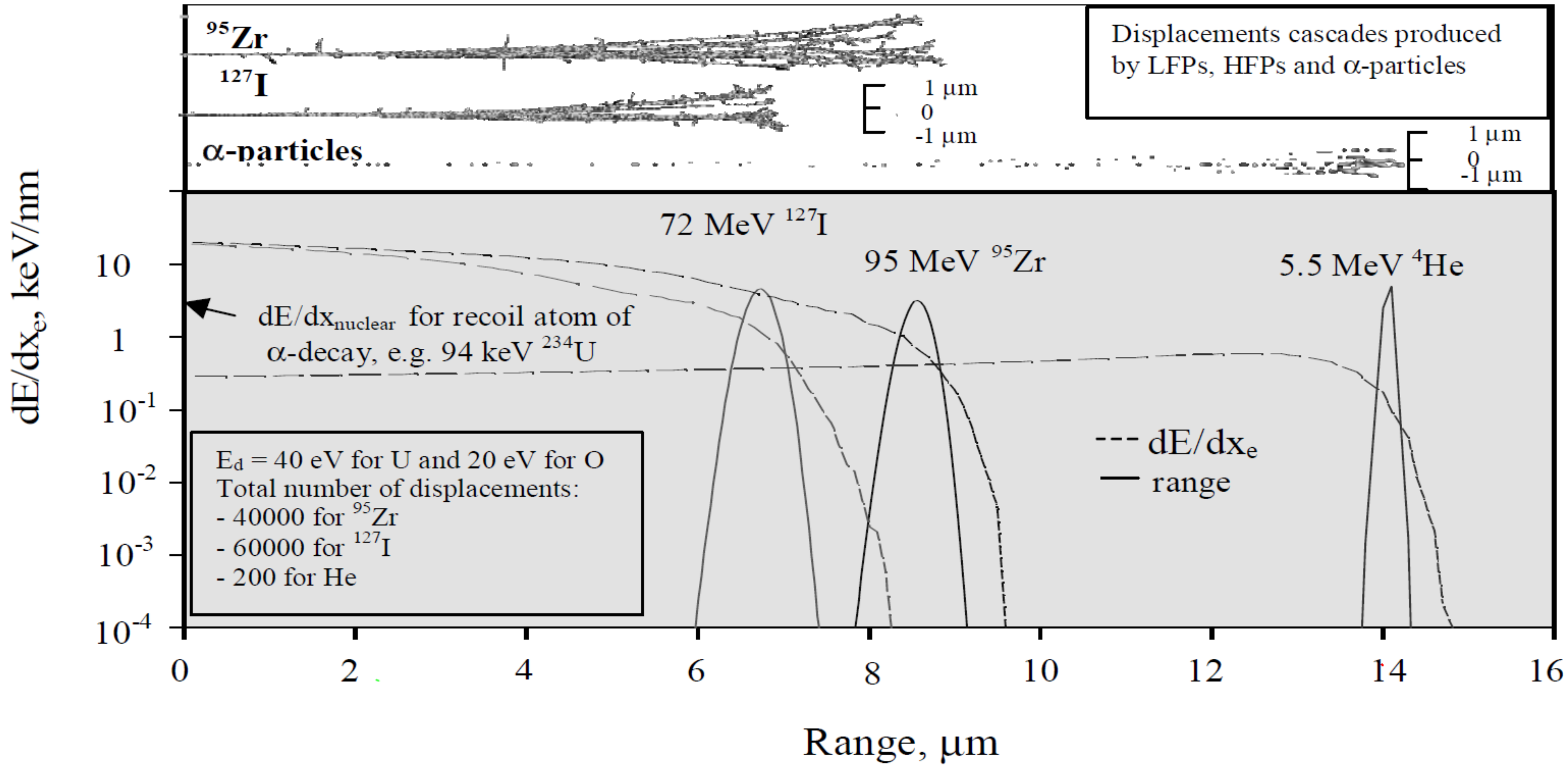
Radiation effects: Fisson product recoil





	Energy (keV)	Range (µm)	$(de/dx)_n$	$(de/dx)_e$	No. of defects
Light fission product	95,000	9	0.03	0.97	40,000
Heavy fission product	70,000	7	0.06	0.94	60,000
α particle	5,500	15	0.01	0.99	200
α recoil atom	95	0.02	0.90	0.10	1,500

Radiation effects : Fisson product recoil



Source: Wiss & Konings (2013), In: Properties of Fluorite Structure Materials. (P. Vajda and M. Costandini, Eds.). Nova Publishers 2013

Pros and cons of UO_2

Pro

- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO_2
- Straightforward fabrication
- Stable against irradiation

Con

- Low thermal conductivity
- Low fissile density
- Dirty process (dust)
- Many steps (MOX)
- Proliferation risk (MOX)

5

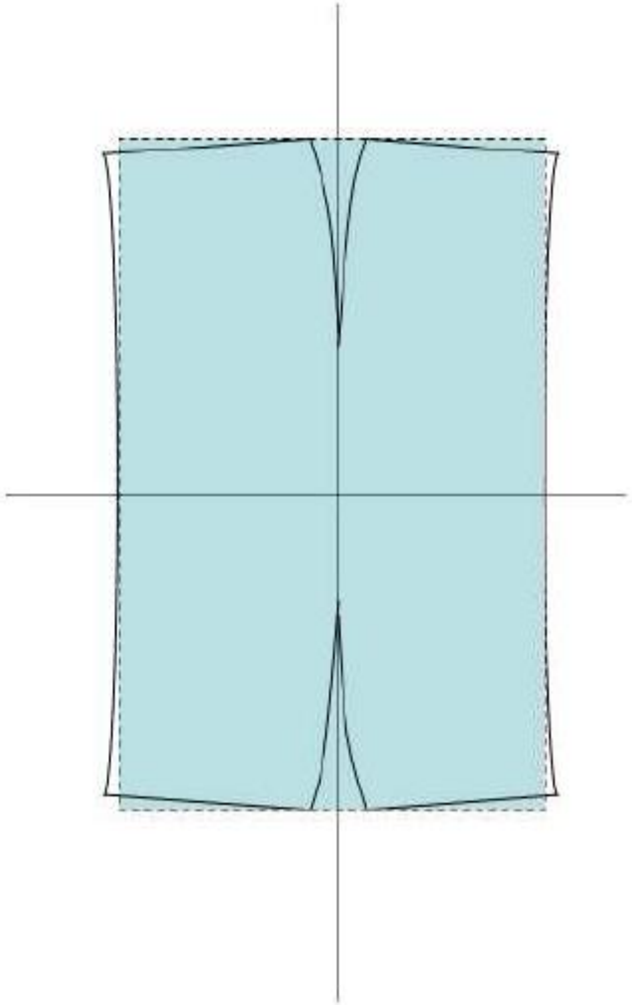
Behaviour under irradiation

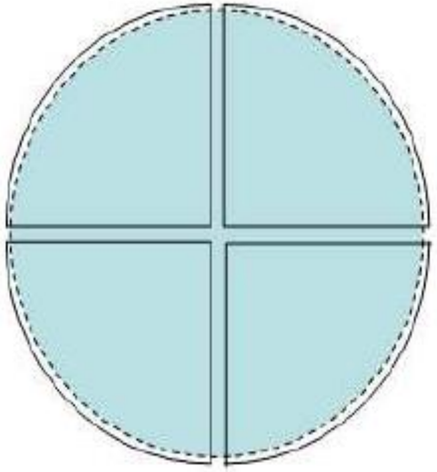
European Commission

Irradiation behaviour: Swelling & Cracking

- The UO_2 expands with temperature (dilatation)
- During irradiation UO_2 swells about 1 vol% per 10 MWd/kgU as results of fission product accumulation
- Volume increase partially compensated by
  - closing of porosity
  - closing of pellet “dishes”
  - closing of gap

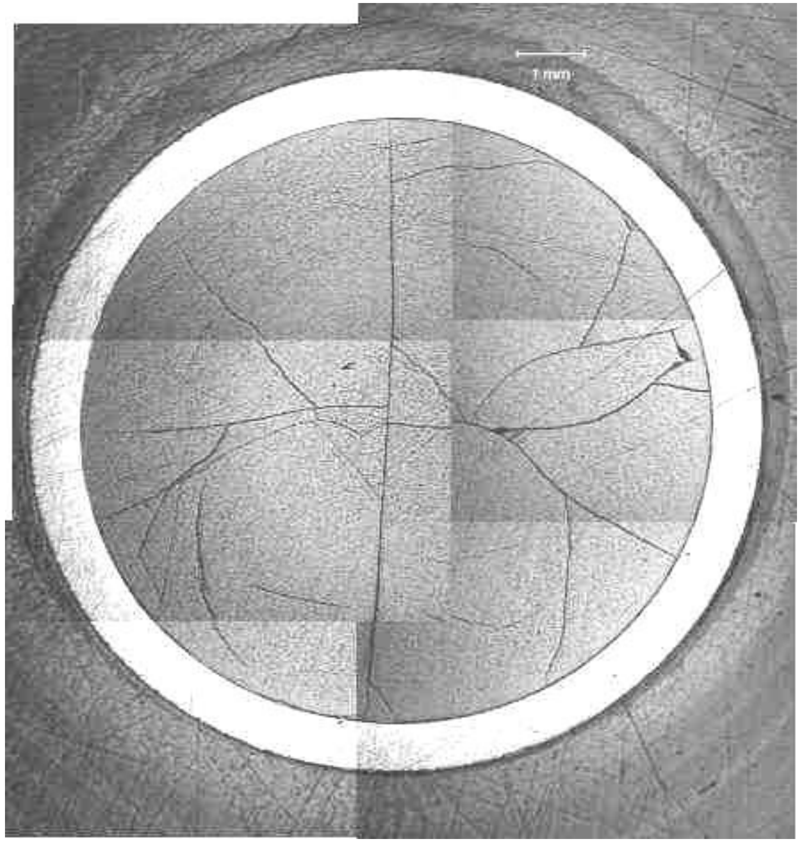
Irradiation behaviour: Swelling & Cracking



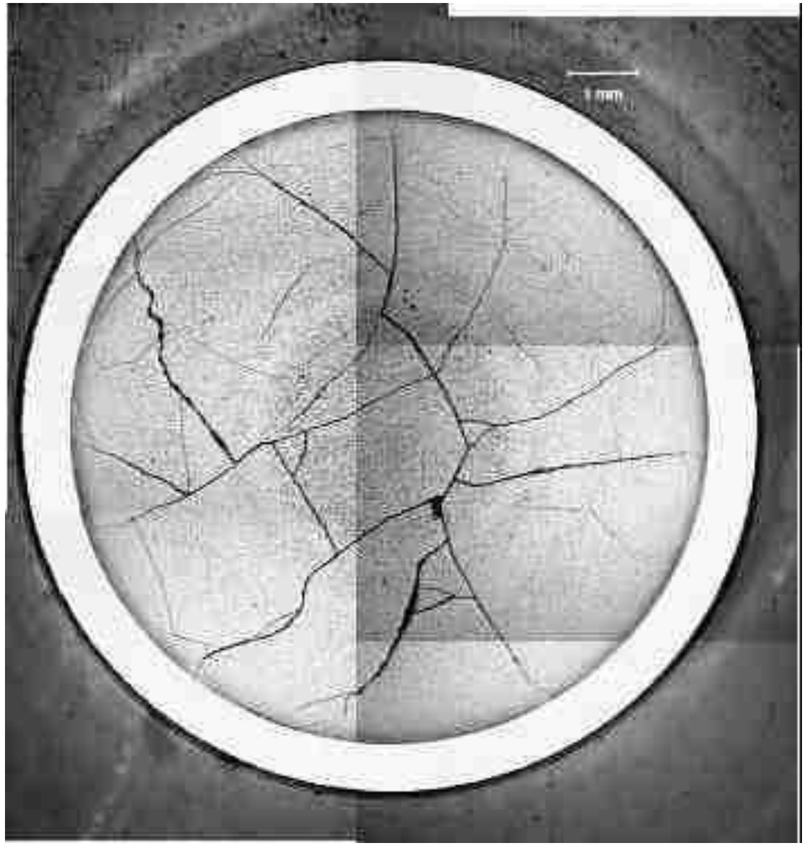


Irradiation behaviour: Swelling & Cracking

UO_2, 53 GWd/t



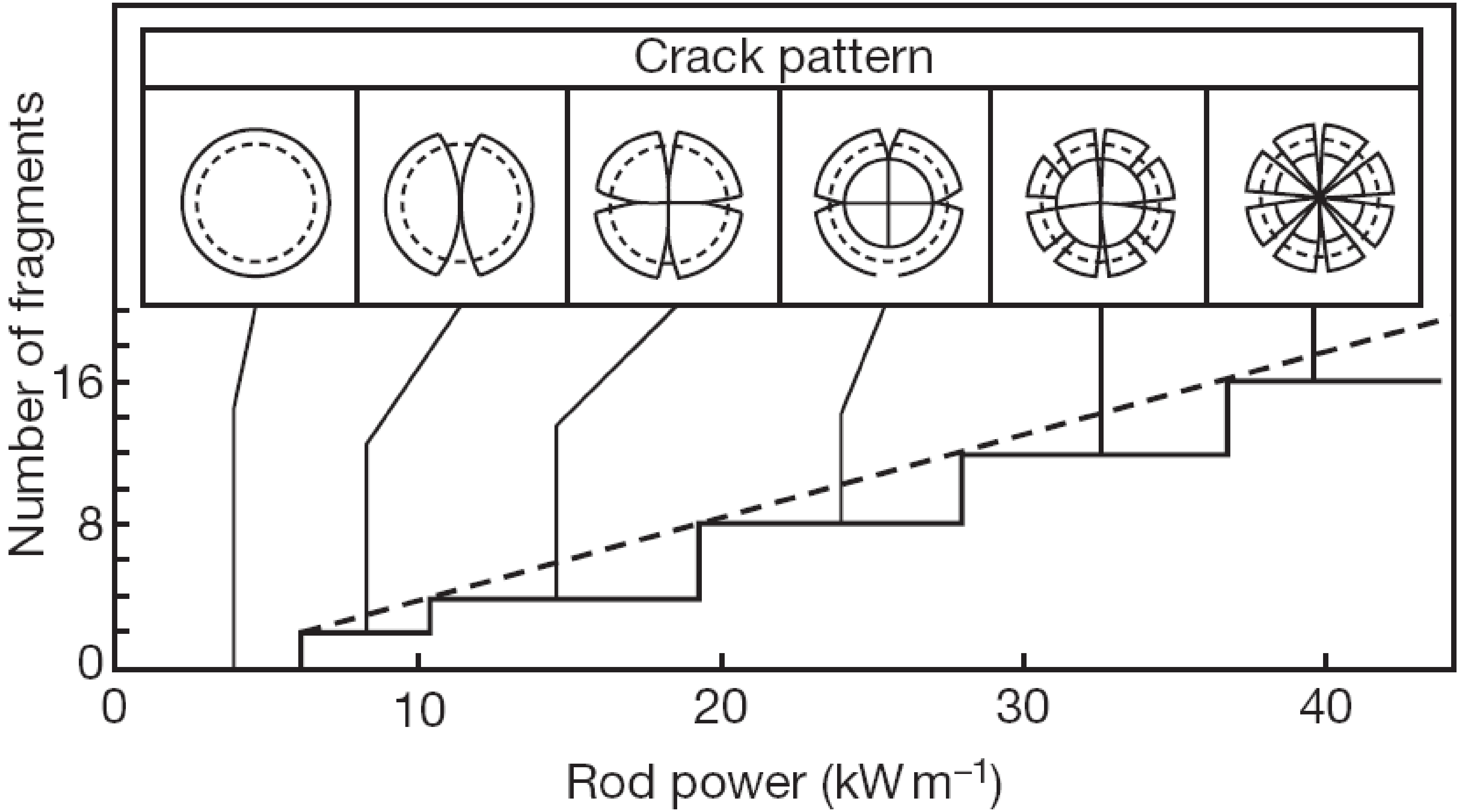
UO_2, 66 GWd/t



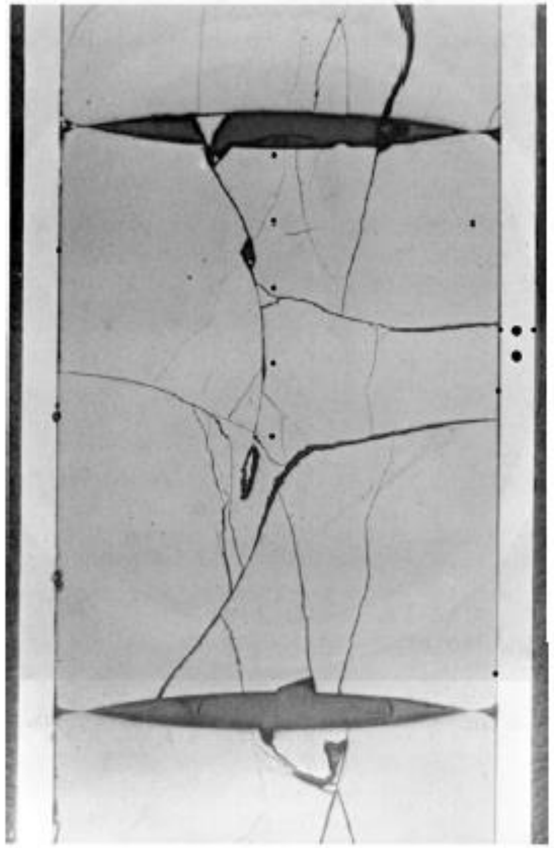
Source: European Communities

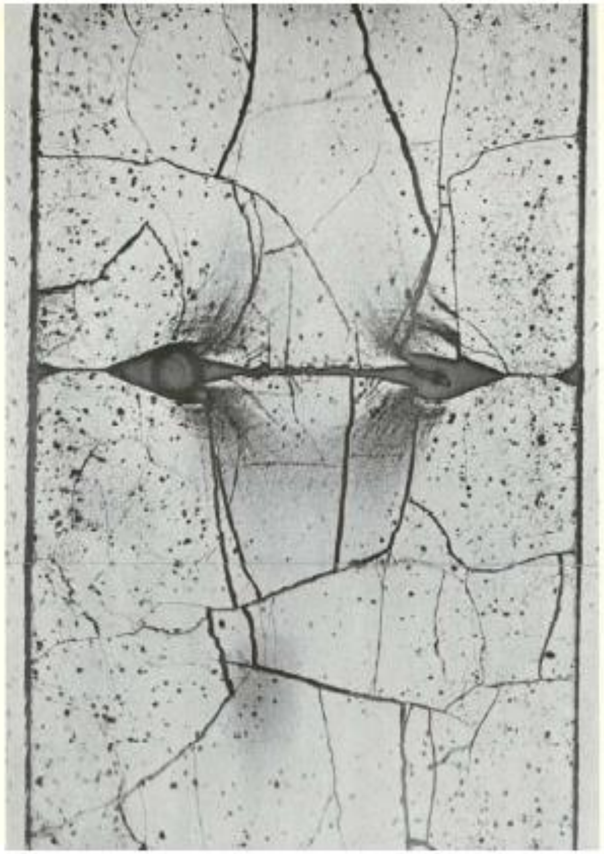
European Commission

Irradiation behaviour: Cracking & Cracking



Irradiation behaviour: Swelling & Cracking

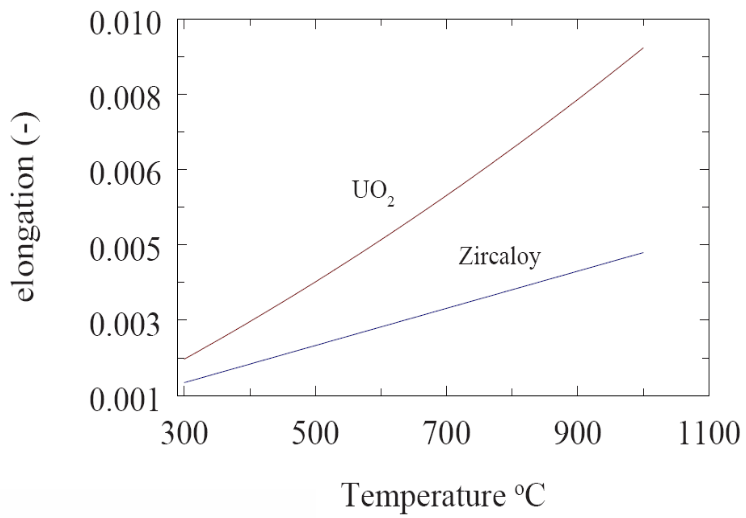


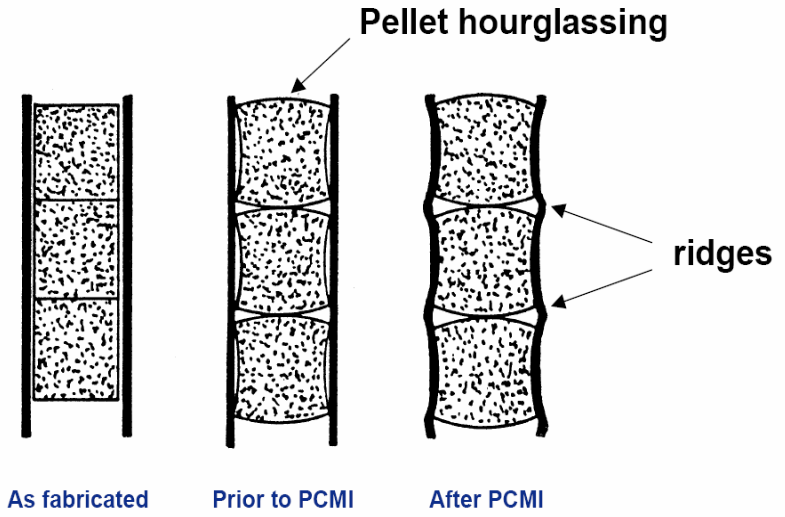


Source: AREVA S.A., Reproduced with permission

Irradiation behaviour: Swelling & Cracking

- Difference in thermal expansion UO_2 and cladding
- The cladding creeps down (shrinks) due to the accumulation of radiation damage
- Once the gap is closed "Pellet Cladding Mechanical Interaction" occurs





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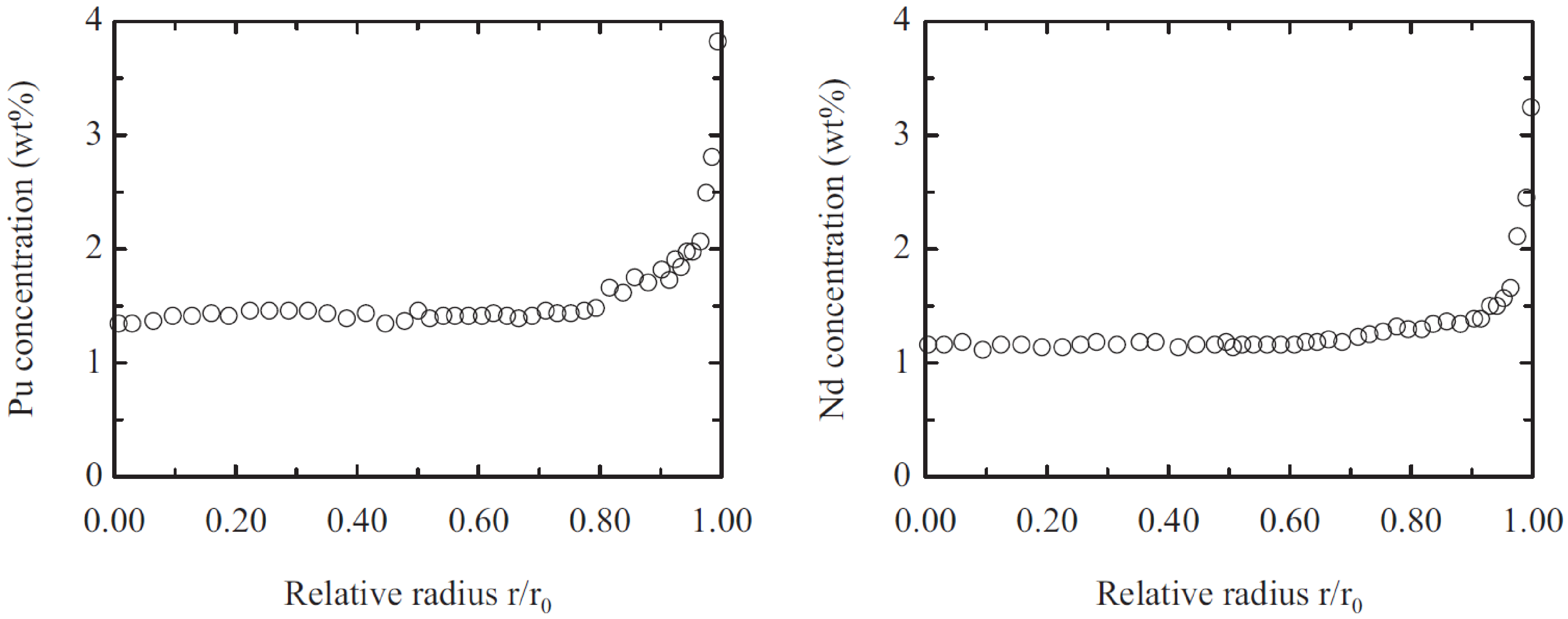
Irradiation behaviour: Plutonium distribution

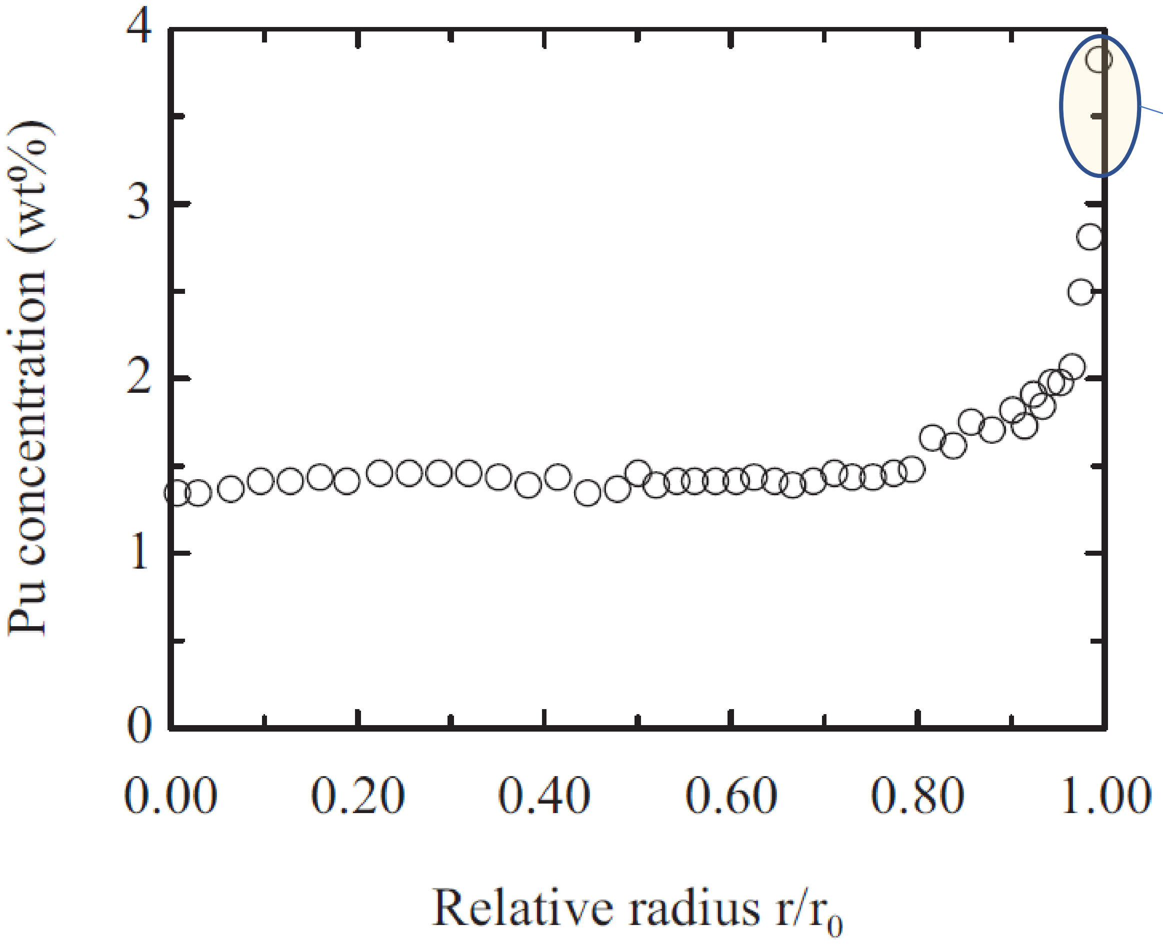


The Pu and Nd concentration profiles for irradiated UO_2 of 97.8 MWd/kgU pellet average burnup measured by EPMA.

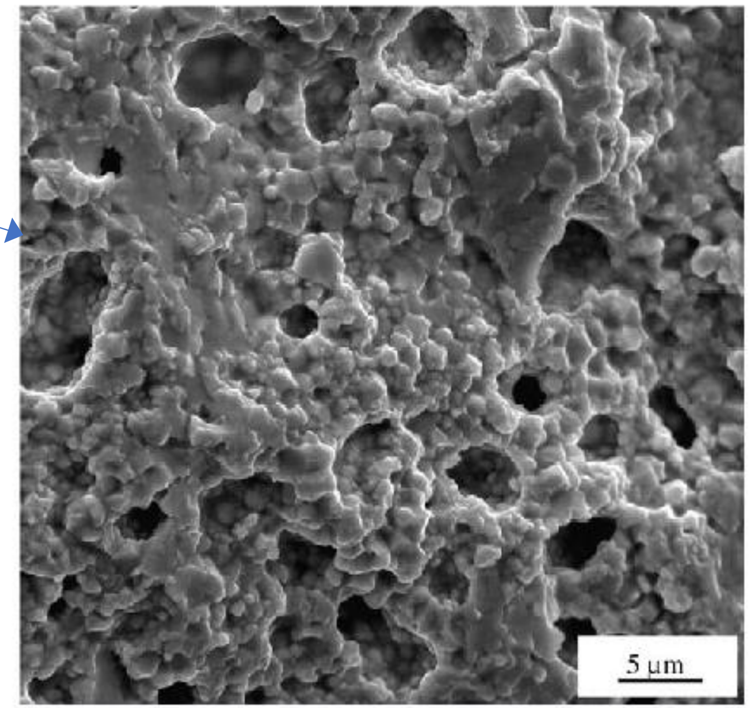
Source: Manzel and Walker, J. Nucl. Mater., 301 (2002) 170).

European Commission

Irradiation behaviour: High Burnup Structure

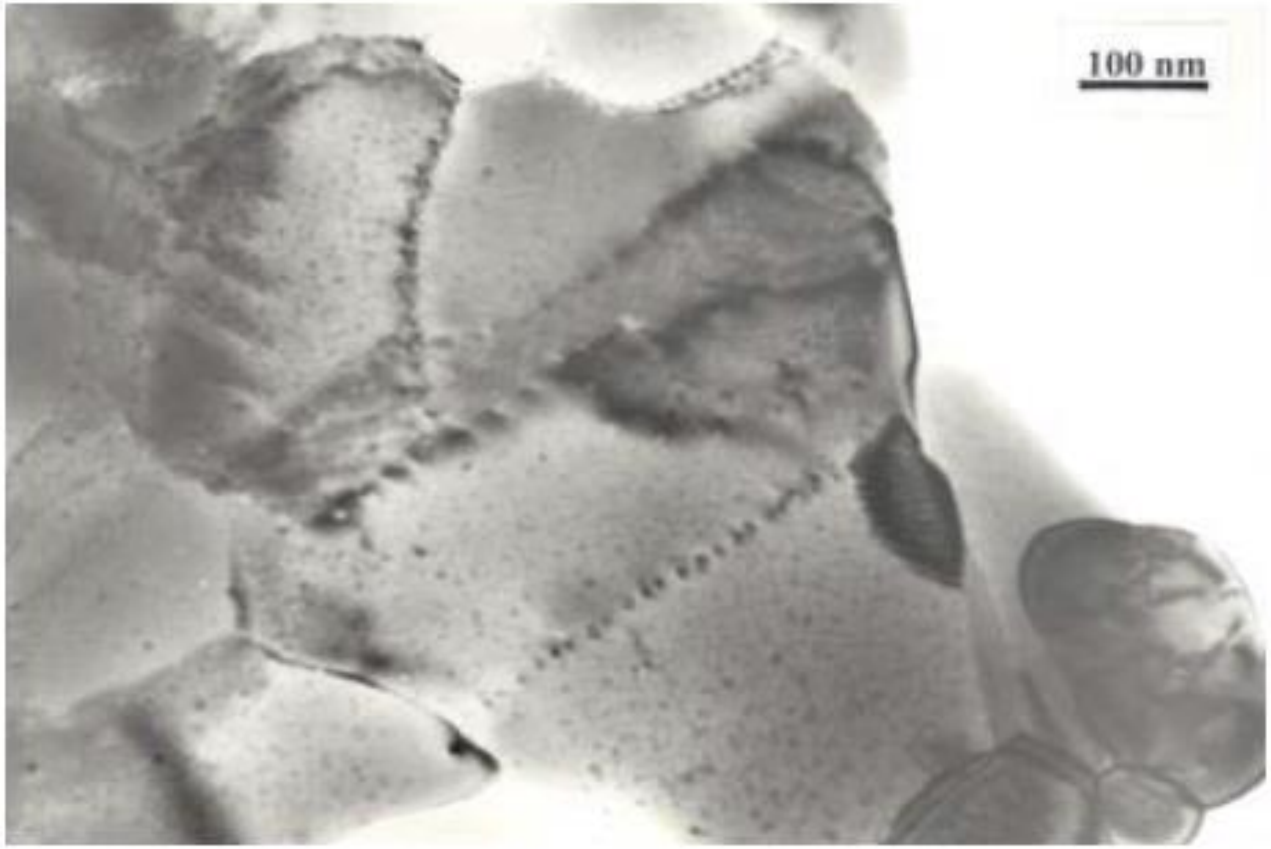


Source: Manzel and Walker, J. Nucl. Mater., 301 (2002) 170).



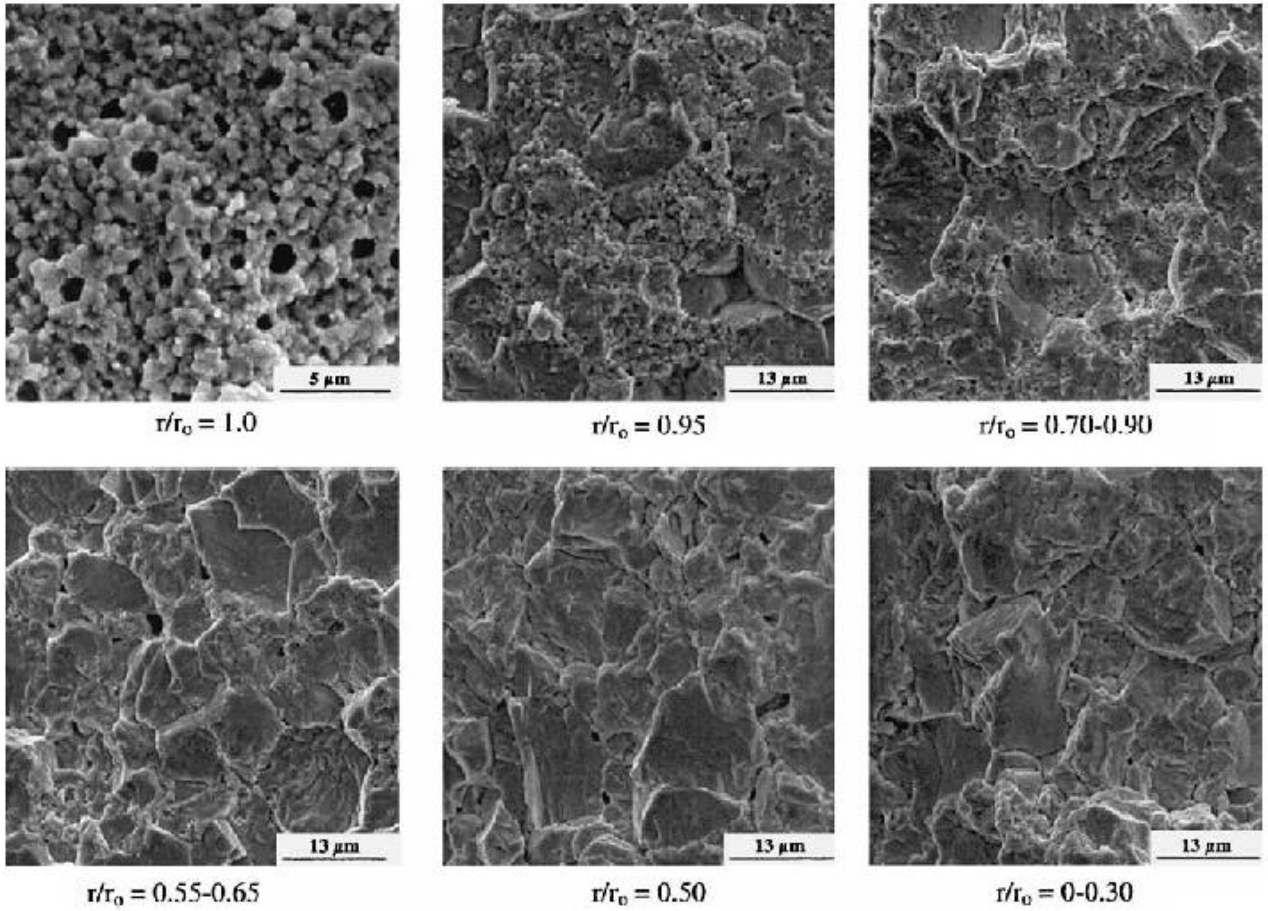
HBS at r/r_0 =0.99. Local burnup is about 210 MWd/kgHM

Irradiation behaviour: High Burnup Structure



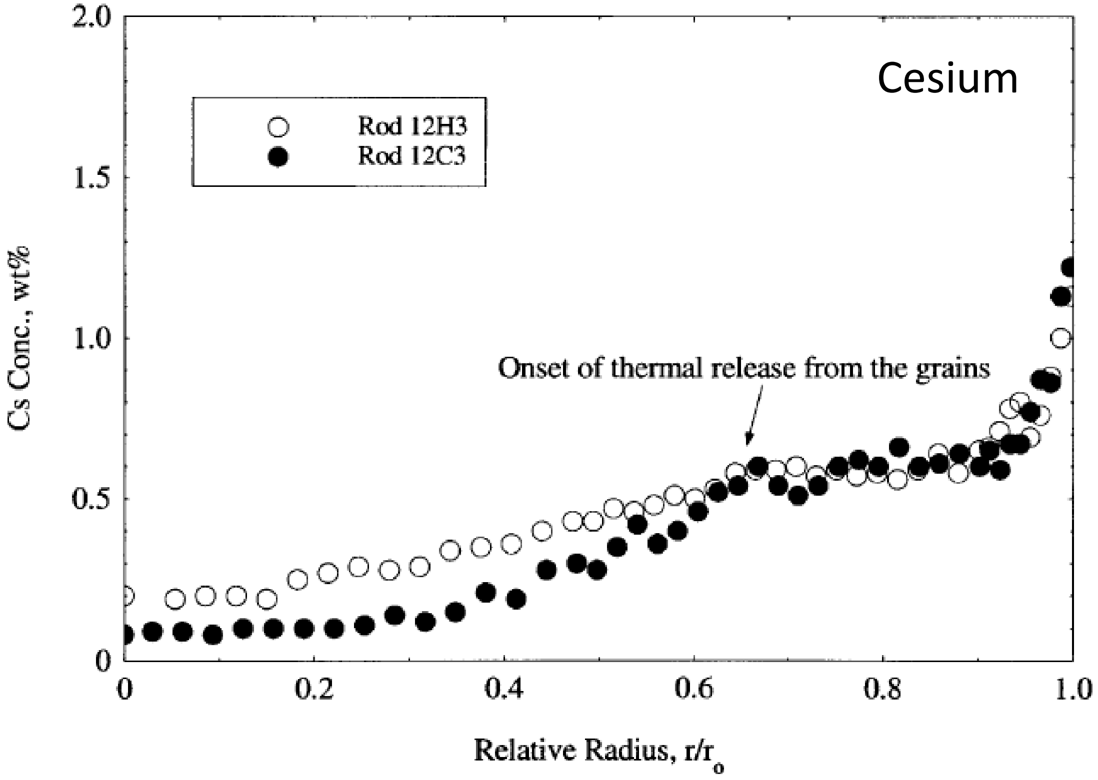
Source: Wiss & Konings (2013), In: Properties of Fluorite Structure Materials. (P. Vajda and M. Costandini, Eds.). Nova Publishers 2013

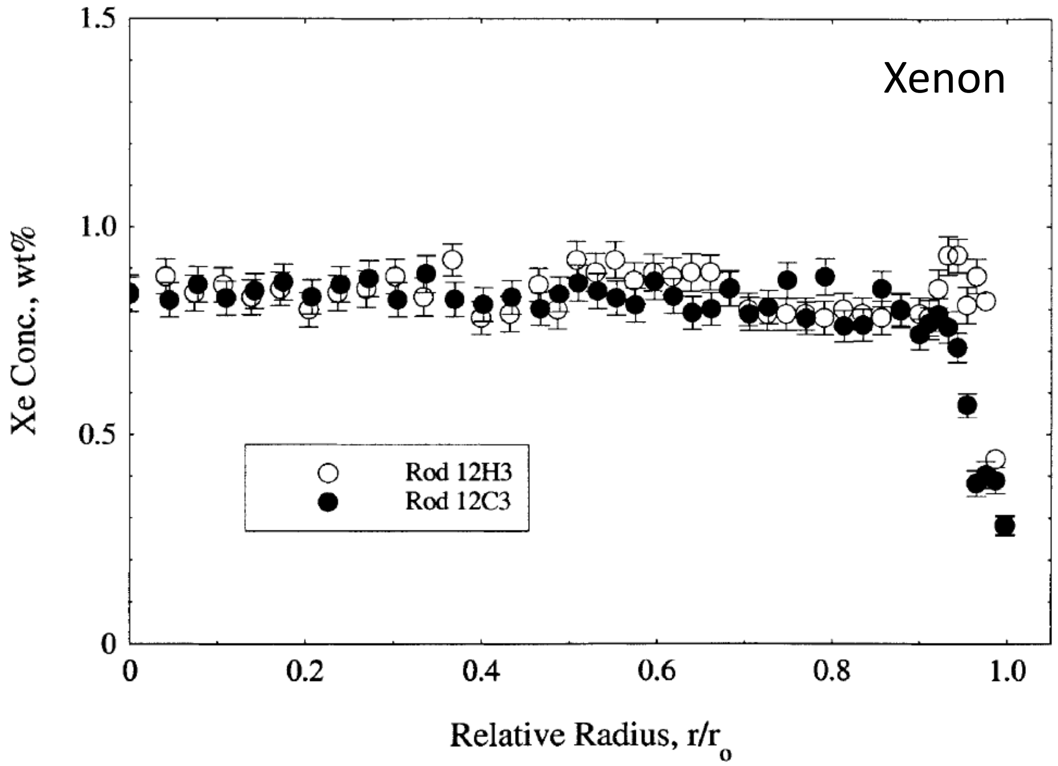
Irradiation behaviour: High Burnup Structure



Source: Manzel and Walker, J. Nucl. Mater., 301 (2002) 170).

Irradiation behaviour: High Burnup Structure



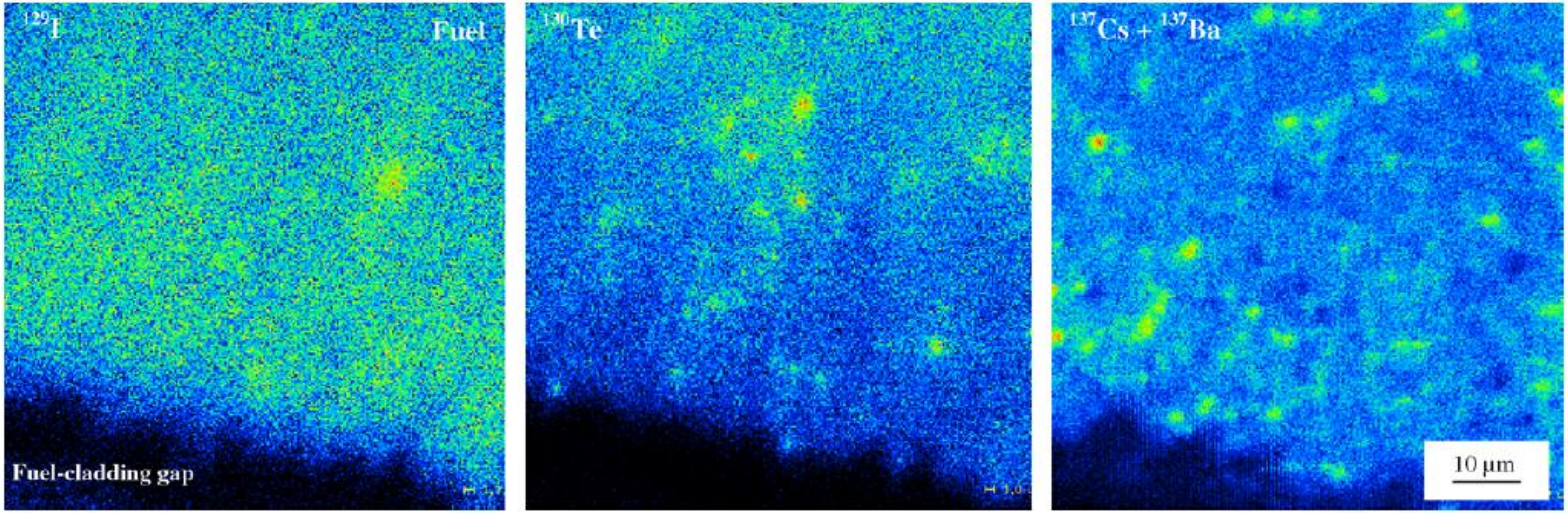


The Cs and Xe concentration profiles for irradiated UO_2 of 97.8 MWd/kgU pellet average burnup measured by EPMA. (After Manzel and Walker, 2002).

European Commission

Irradiation behaviour: High Burnup Structure

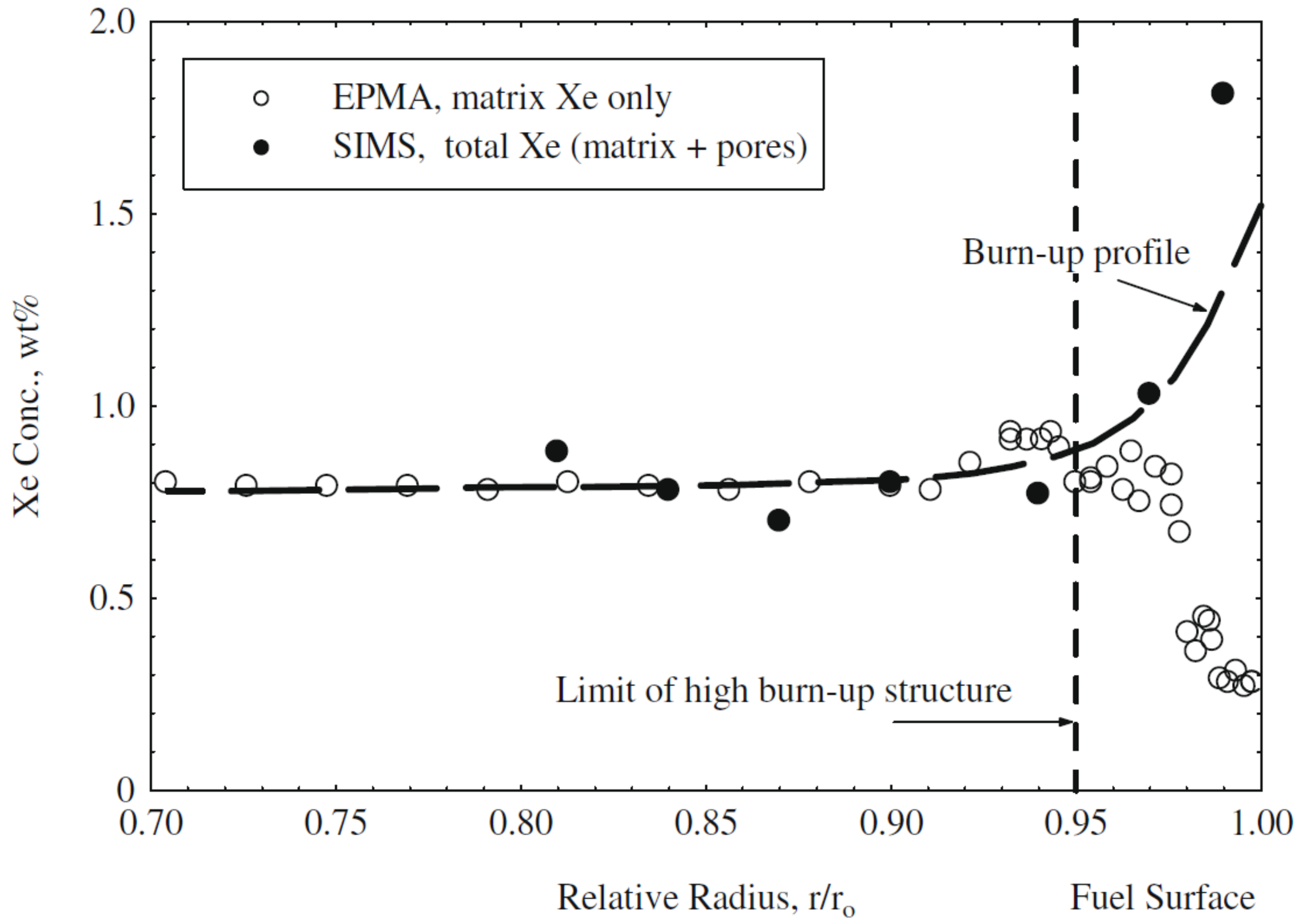
^{129}I ^{130}Te $^{137}Cs + ^{137}Ba$



Elemental distribution maps of fission products in the High Burnup Structure near pellet rim.

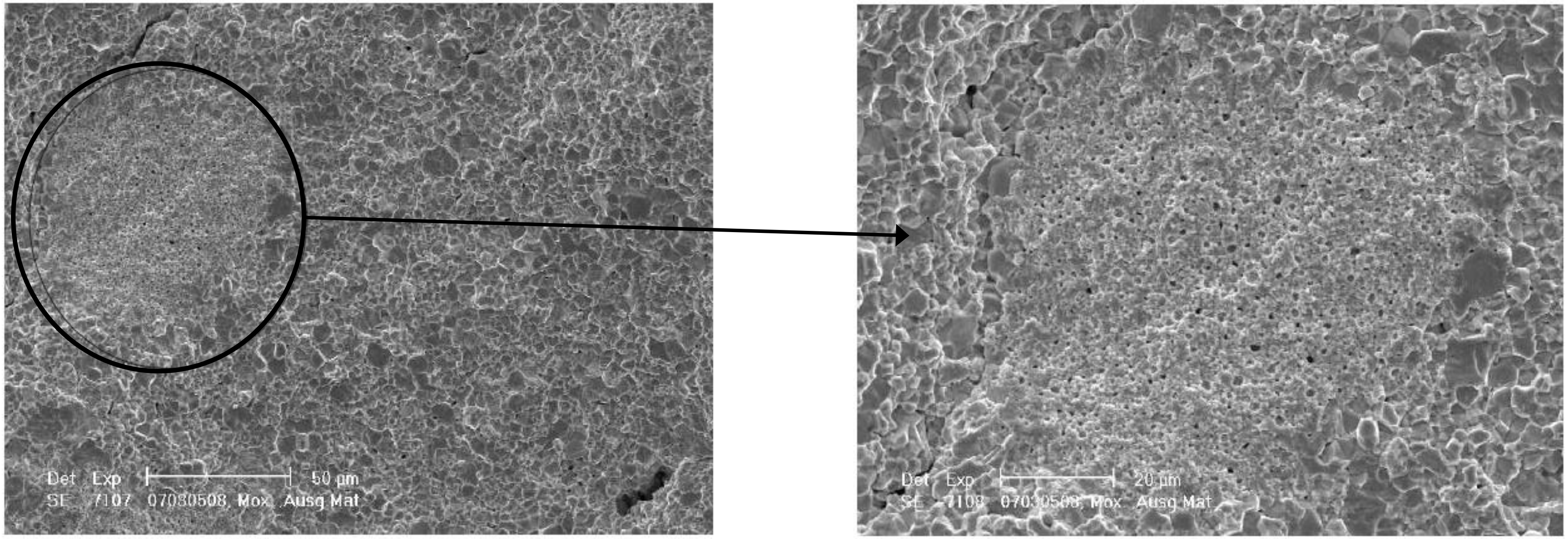
Source: Walker et al. (2009), J. Nucl. Mat.393:212

Irradiation behaviour: High Burnup Structure



Source: Walker et al. (2009), J. Nucl. Mat.393:212.

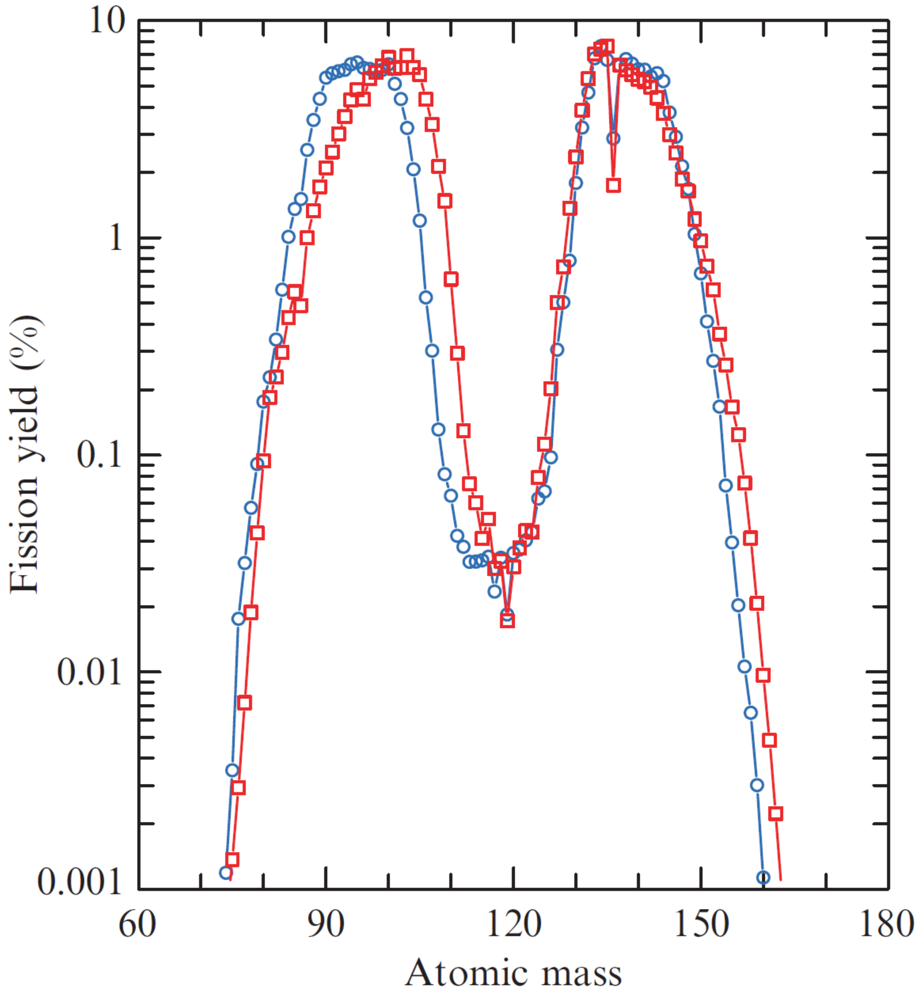
Irradiation behaviour: High Burnup Structure



High Burnup Structure formation in Pu-rich island in irradiated Mixed Oxide fuel

European Commission

Irradiation behaviour: Fission products

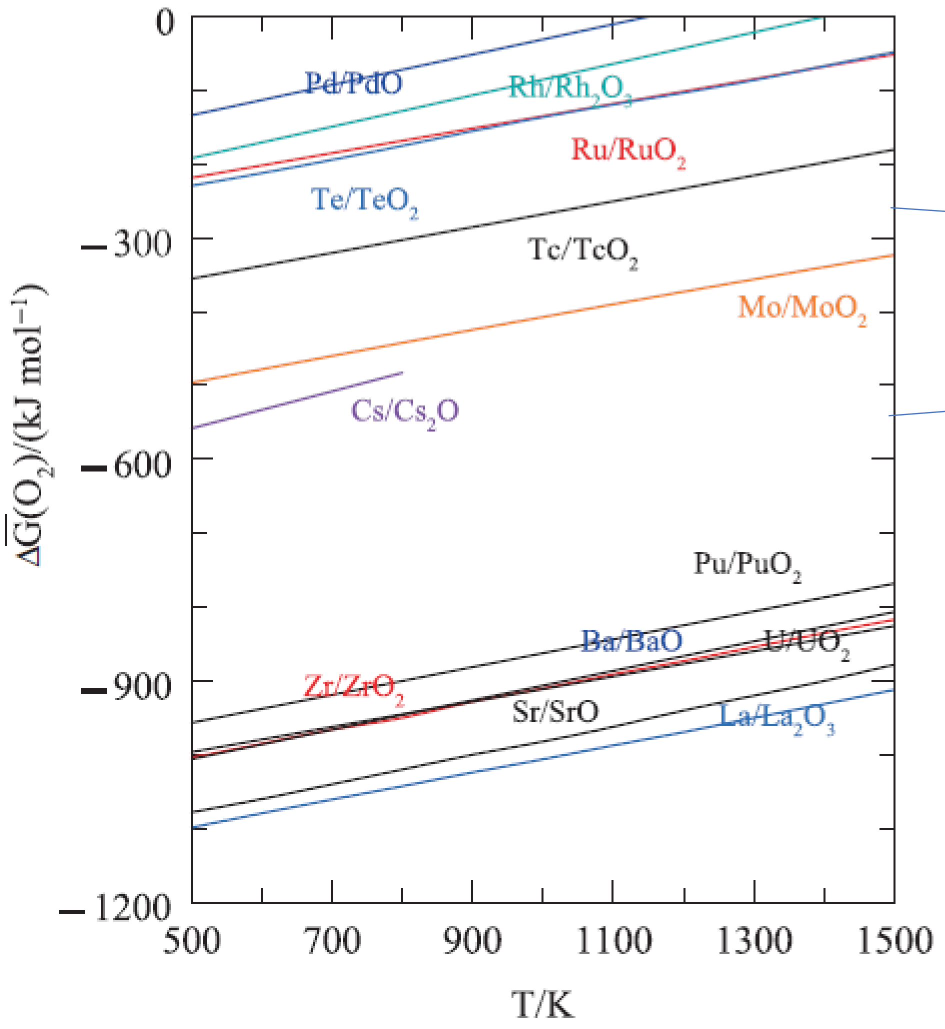


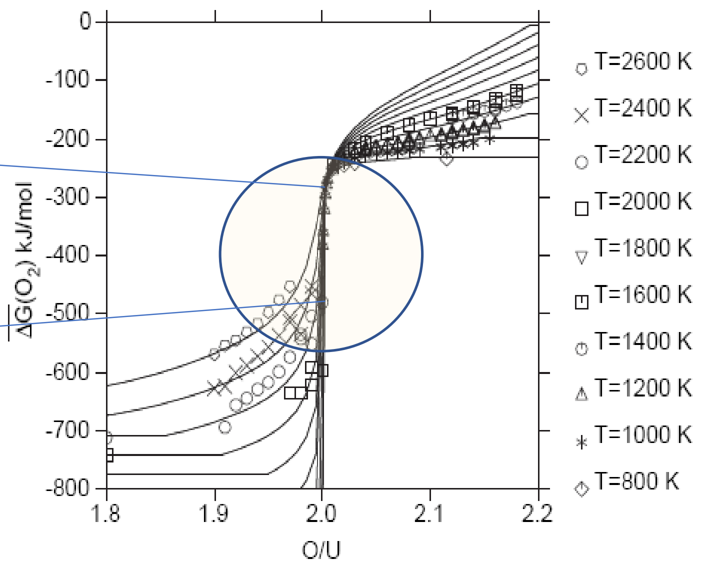
Fissions yields of ^{235}U (blue) in a thermal neutron spectrum and ^{239}Pu (red) in a fast neutron spectrum.

Irradiation behaviour: Fission products

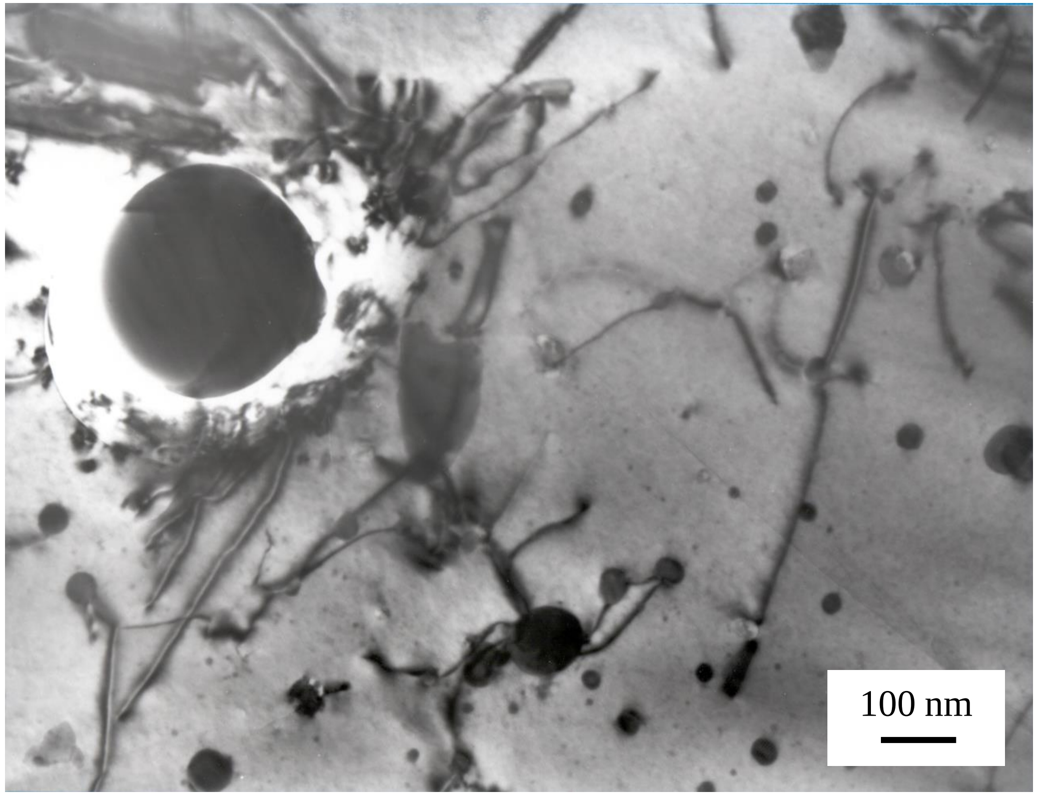
1) Dissolved in the matrix: Rb, Sr, Y, Zr, Nb, Te, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu
2) Oxide precipitates at grain boundaries: Rb, Sr, Zr, Nb, Mo, Se, Te, Cs, Ba
3) Metallic precipitates: Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Se, Te
4) Gases: Kr, Xe
5) Volatiles: Br, Rb, I, Cs, Te

Irradiation behaviour: Fission products





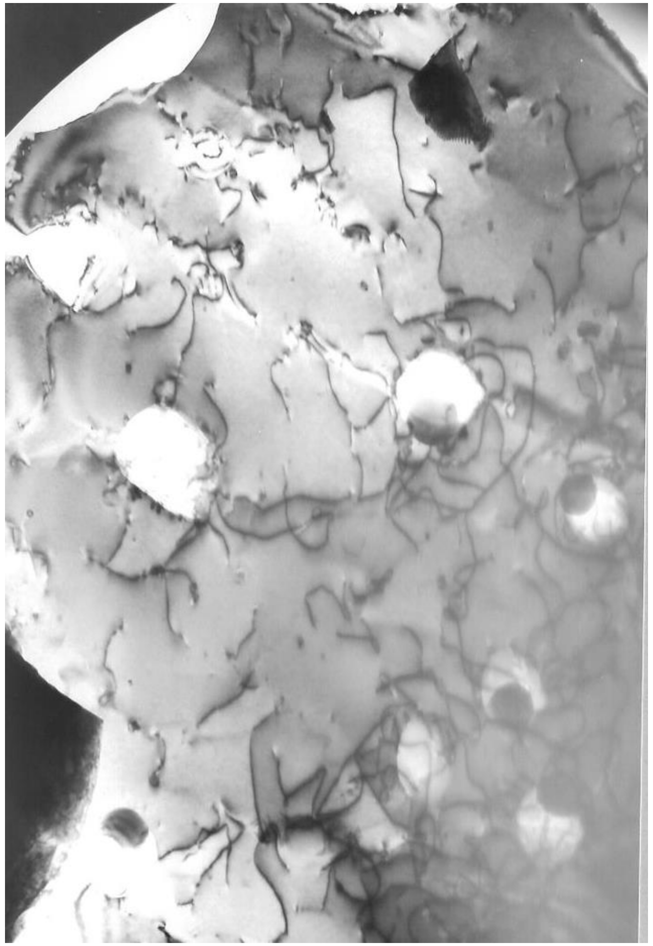
Irradiation behaviour: Fission products



Source: European Communities ©

TEM micrograph of a UO_2 fuel irradiated to high burnup showing a large fission product inclusion, dislocation loops, fission product precipitates sometimes pinning dislocation lines.

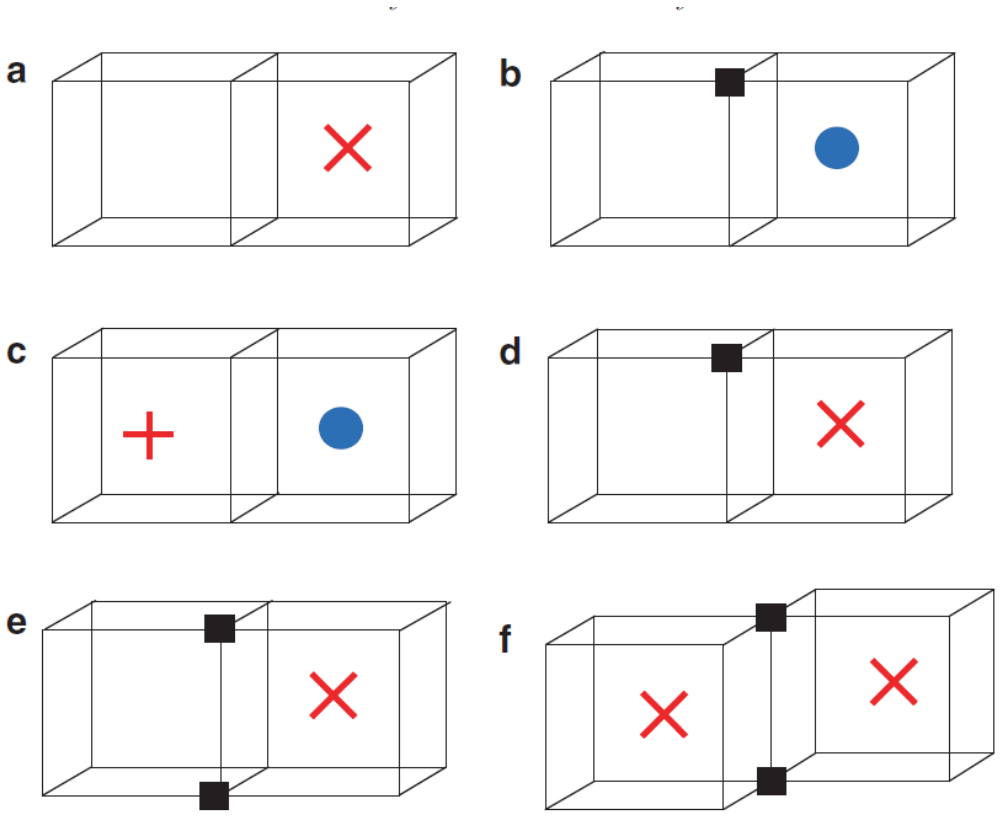
Irradiation behaviour: Fission products

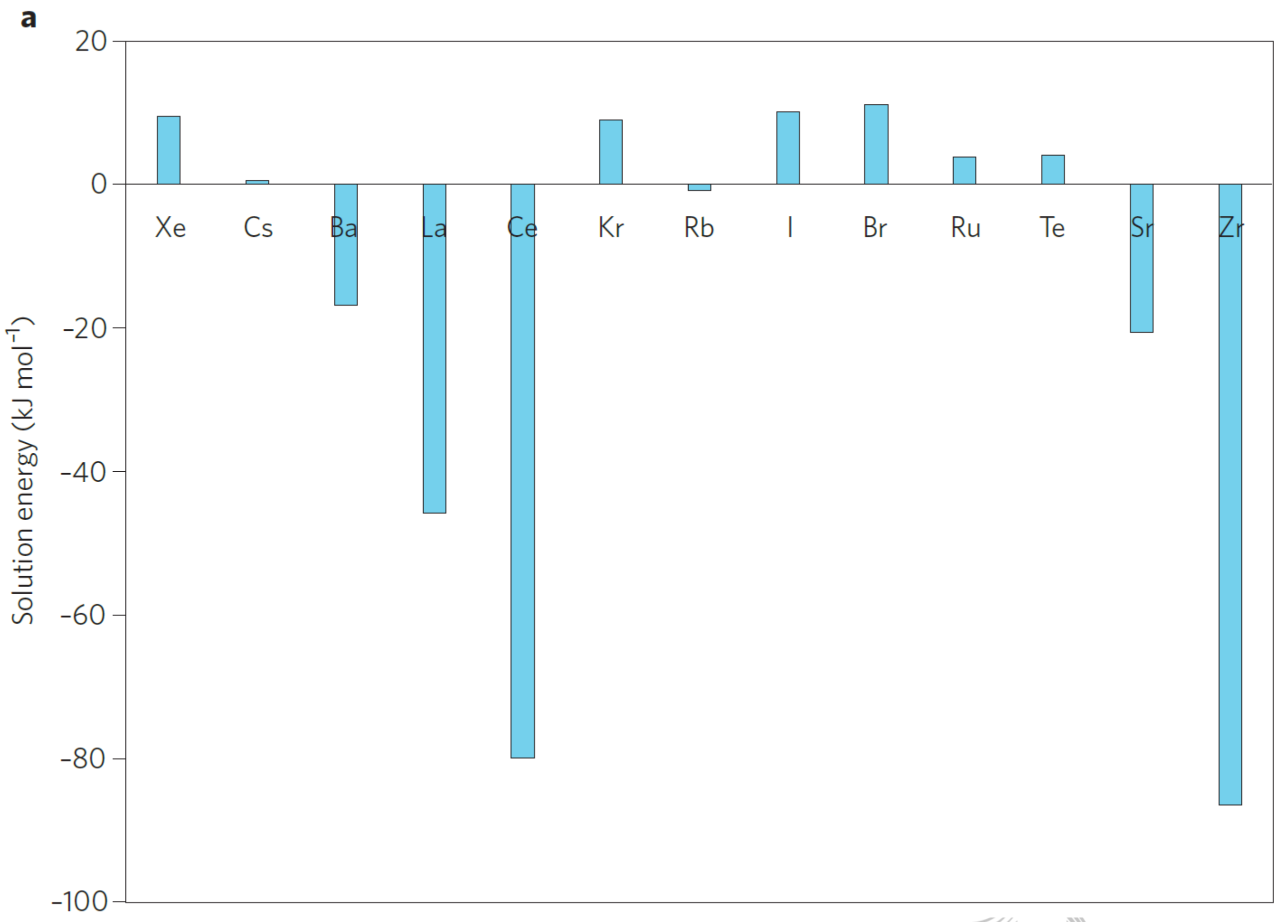


Source: European Communities ©

TEM micrograph of a UO_2 fuel irradiated to high burnup showing a large fission product inclusion, dislocation loops, fission product precipitates sometimes pinning dislocation lines.

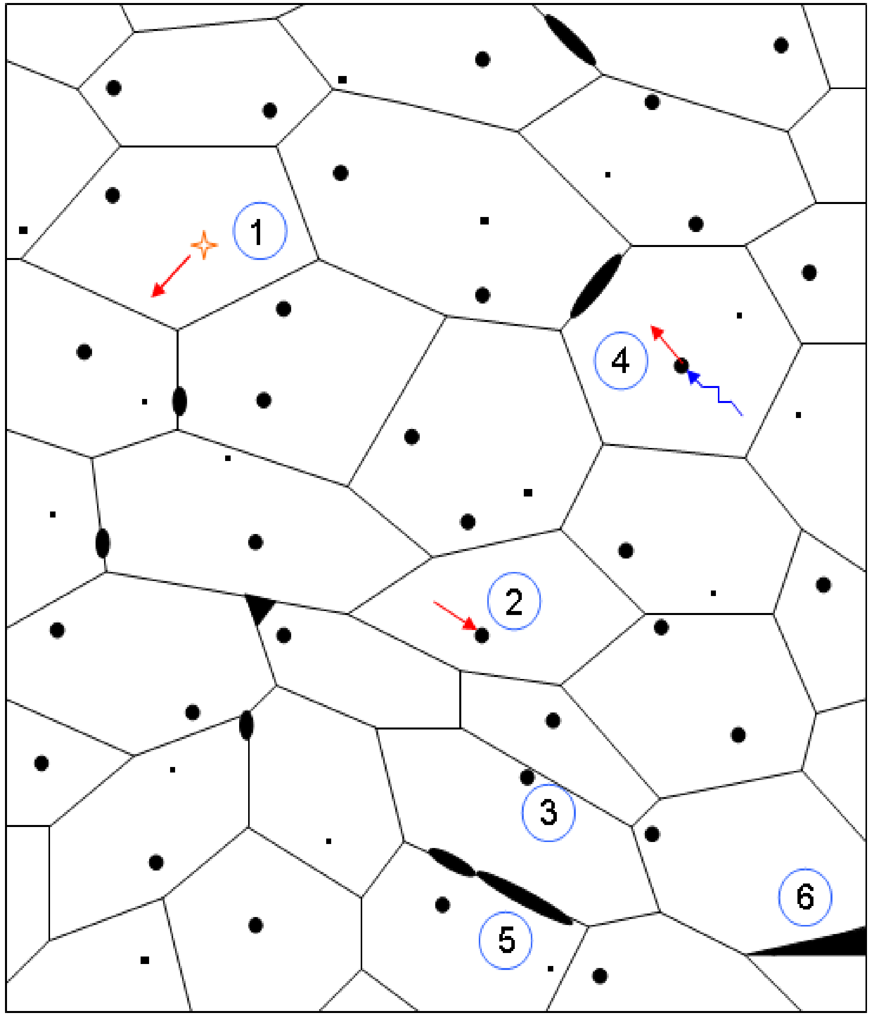
Irradiation behaviour: Fission products





After Grimes and Catlow, Phil. Trans. R. Soc. London A, 335 (1991) 609

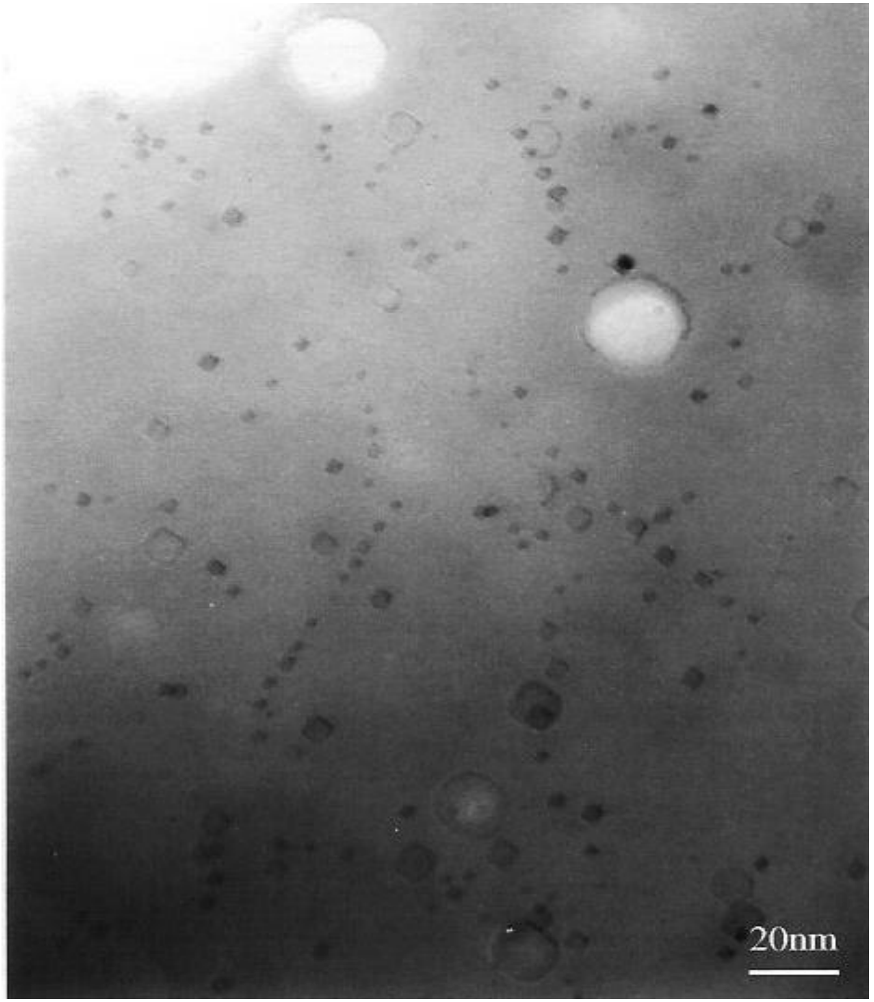
Irradiation behaviour: Fission gases



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

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

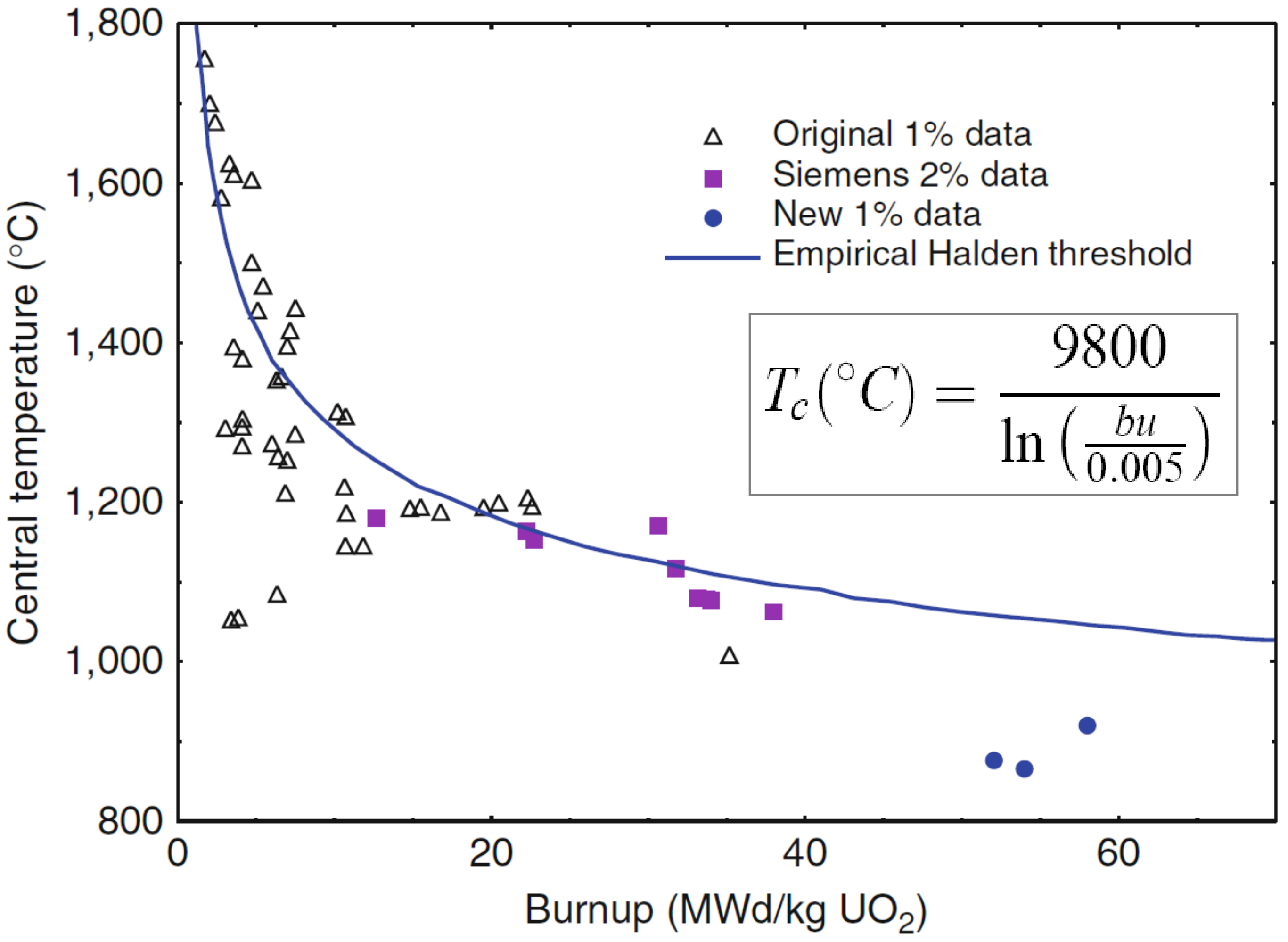
Irradiation behaviour: Fission gases



Source: European Communities ©

1. Small nano-meter sized bubbles
2. Density 10^{23}-10^{24} m^{-3}
3. Reduce D_{eff} by reducing the amount of gas available for migration
4. Nucleated in lines in the wake of fission fragments

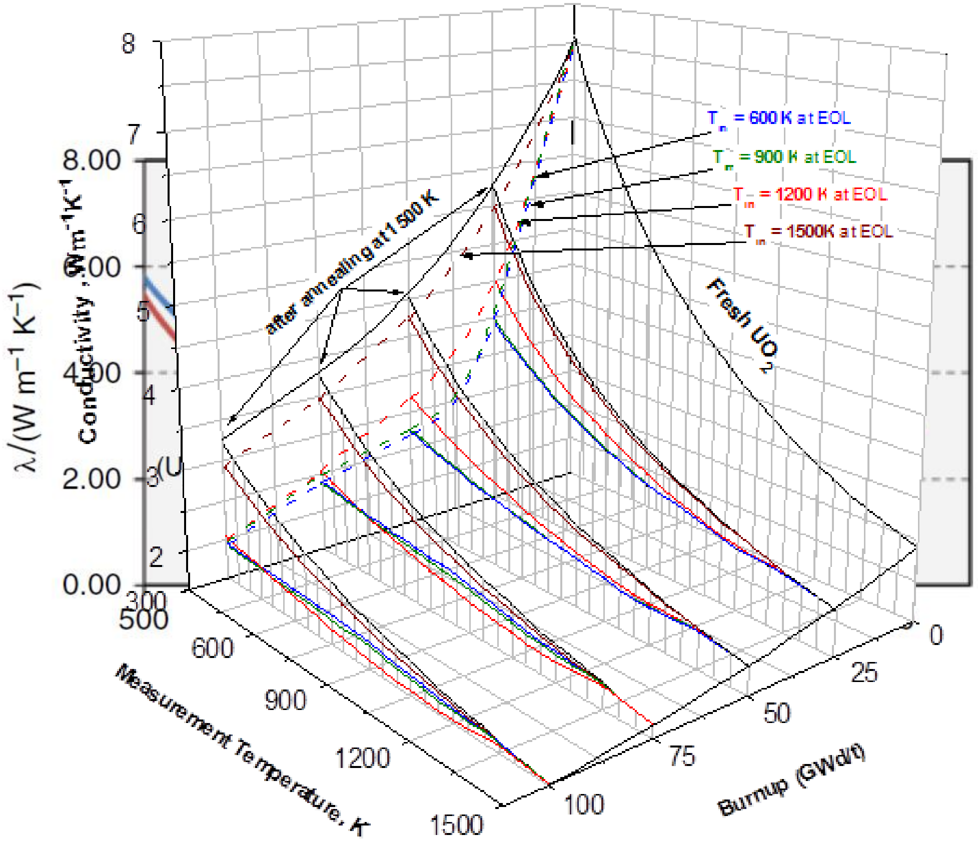
Irradiation behaviour: Fission gases



Halden treshold: the relation between the fuel centerline temperature and burnup for a (arbitrary) 1% fission gas release, based on the in-pile experiments performed in the Halden test reactor (Norway).

European Commission

Irradiation behaviour: Thermal conductivity

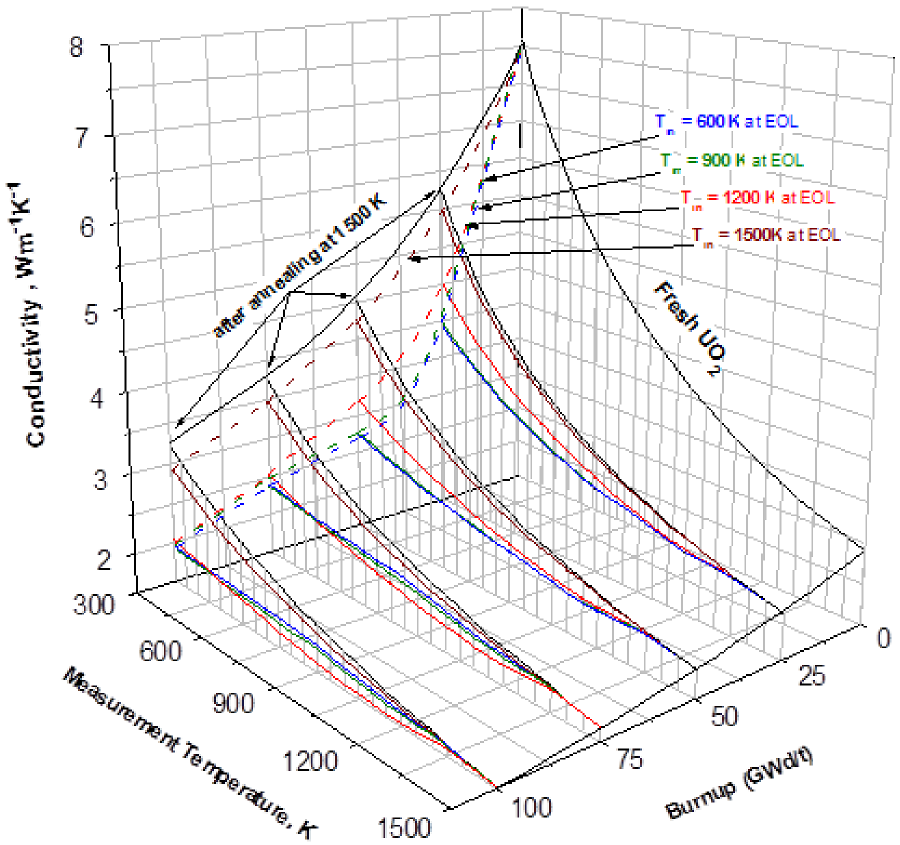


$$\lambda = \frac{1}{A(T_{irr}, T_{an}, bu) + B(T_{irr}, T_{an}, bu)T}$$

- T_{ann}: maximum temperature (700-1450 K) reached during annealing
- *bu*: burn-up (0 to 100 GWd/t)
- Adaptation to in-pile thermal conductivity: $T_{irr}=T_{ann}=T$

Source: Ronchi et al. J. Nucl. Mater. 327 (2004) 58.

Irradiation behaviour: Thermal conductivity



Source: Ronchi et al. J. Nucl. Mater. 327 (2004) 58.

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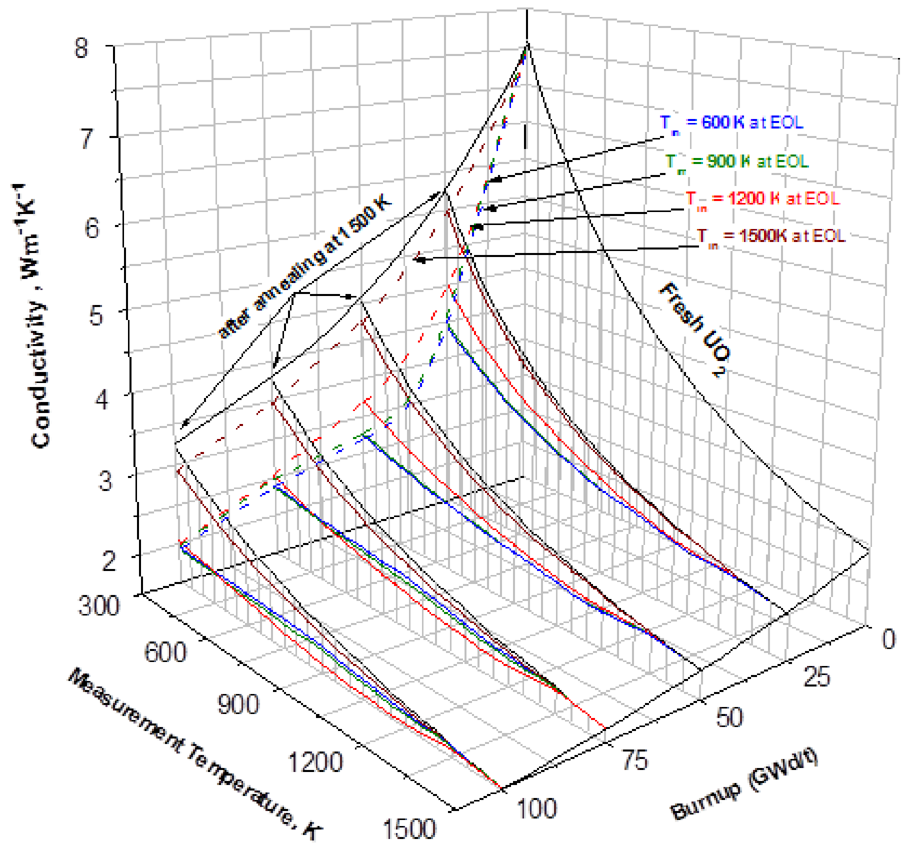
Irradiation behaviour: Thermal conductivity

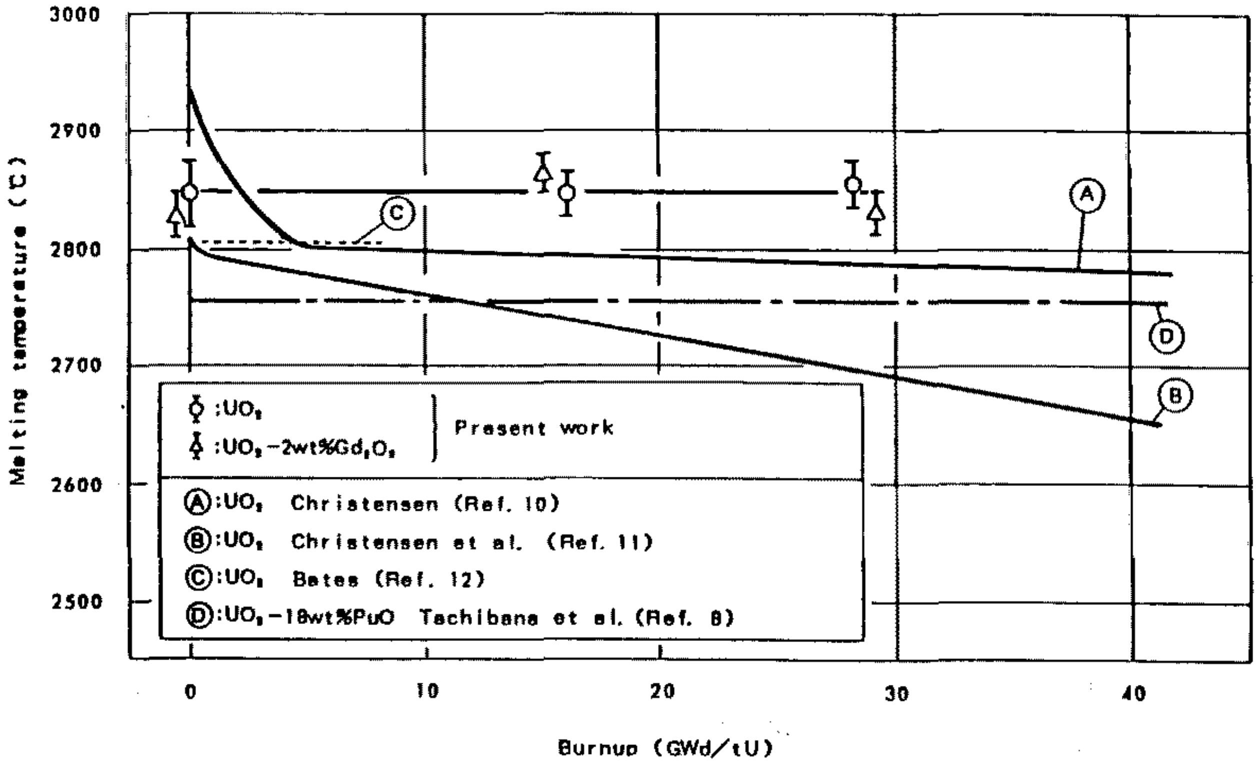


Dominant effects (Ronchi et al., 2004b):

1. Soluble, non-volatile fission products
2. Fission gas and Cs content and its state
3. Irradiation defects (both present at end-of-life and created during subsequent storage by self-irradiation)
4. Precipitation of the fission gasses
5. Annihilation of irradiation defects for thermal recovery conditions

Source: Ronchi et al. J. Nucl. Mater. 327 (2004) 58.

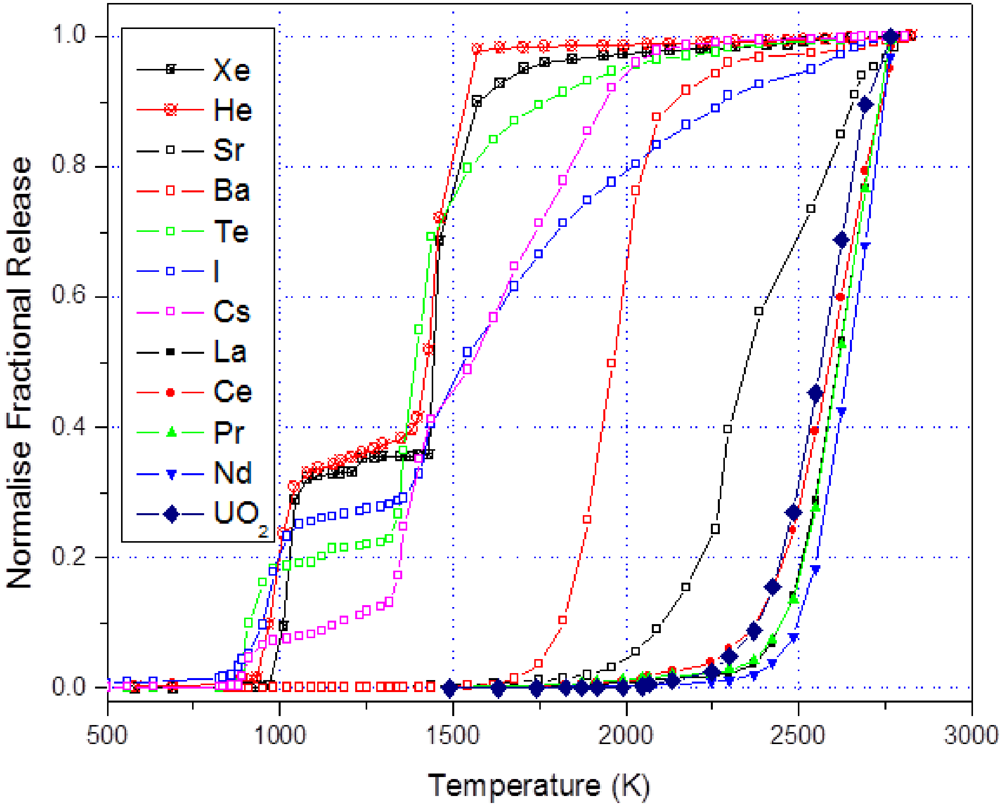
Irradiation behaviour: Melting point



Change in melting temperture of irradiated fuel as a function of burnup (Yamanouchi et al.)

Source: Yamanouchi et al., J. Nucl. Sci. Technol. 25 (1988) 528.

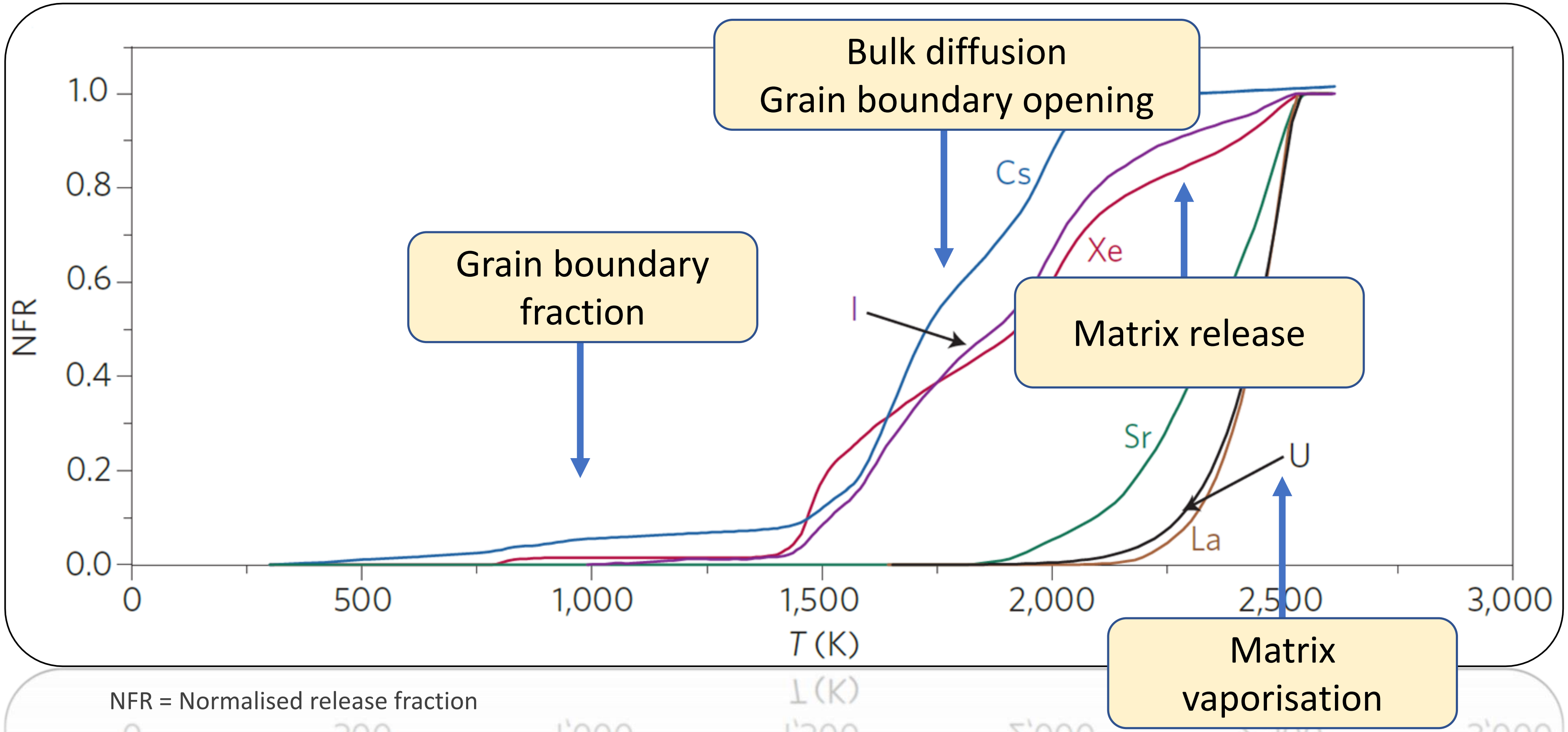
Irradiation behaviour: Fission gas release



Source: European Communities ©

Irradiation behaviour: Fission Product Release

Konings et al., Nature Materials 14 (2015) 247



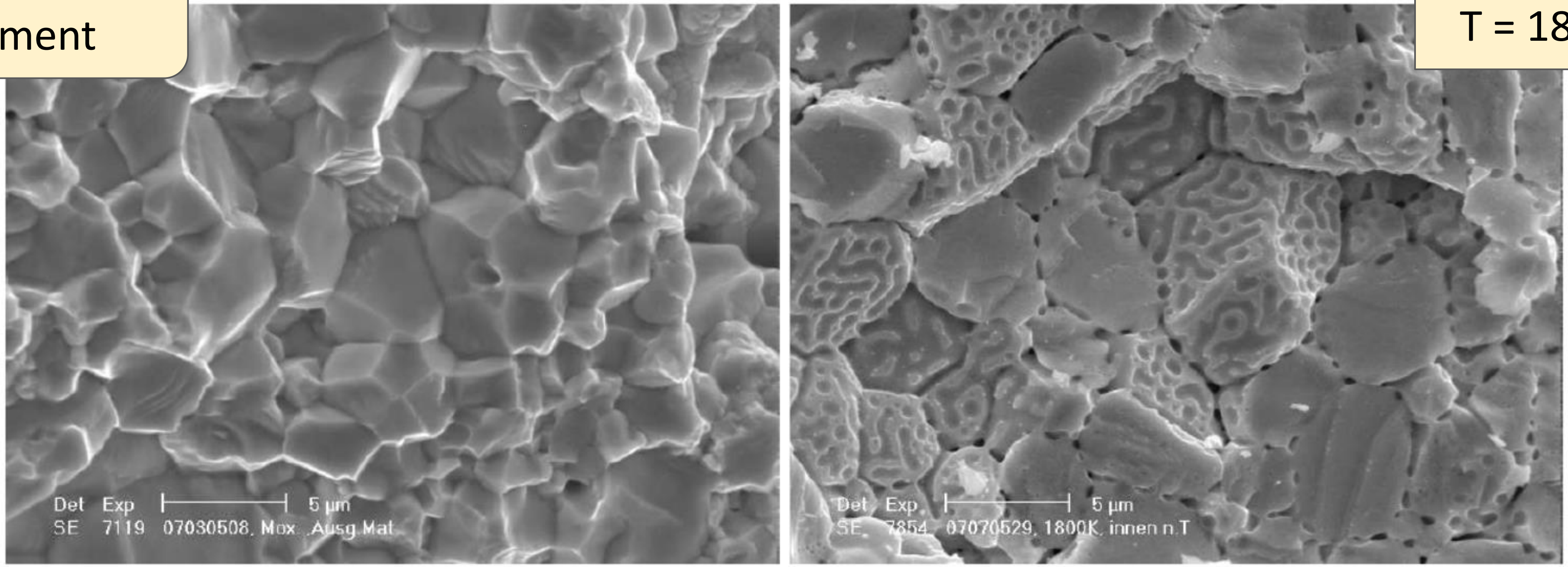
NFR = Normalised release fraction

Source: Konings et al. Nature Materials 14 (2015) 247

Irradiation behaviour: Fission Product Release

SEM of before KEMS measurement

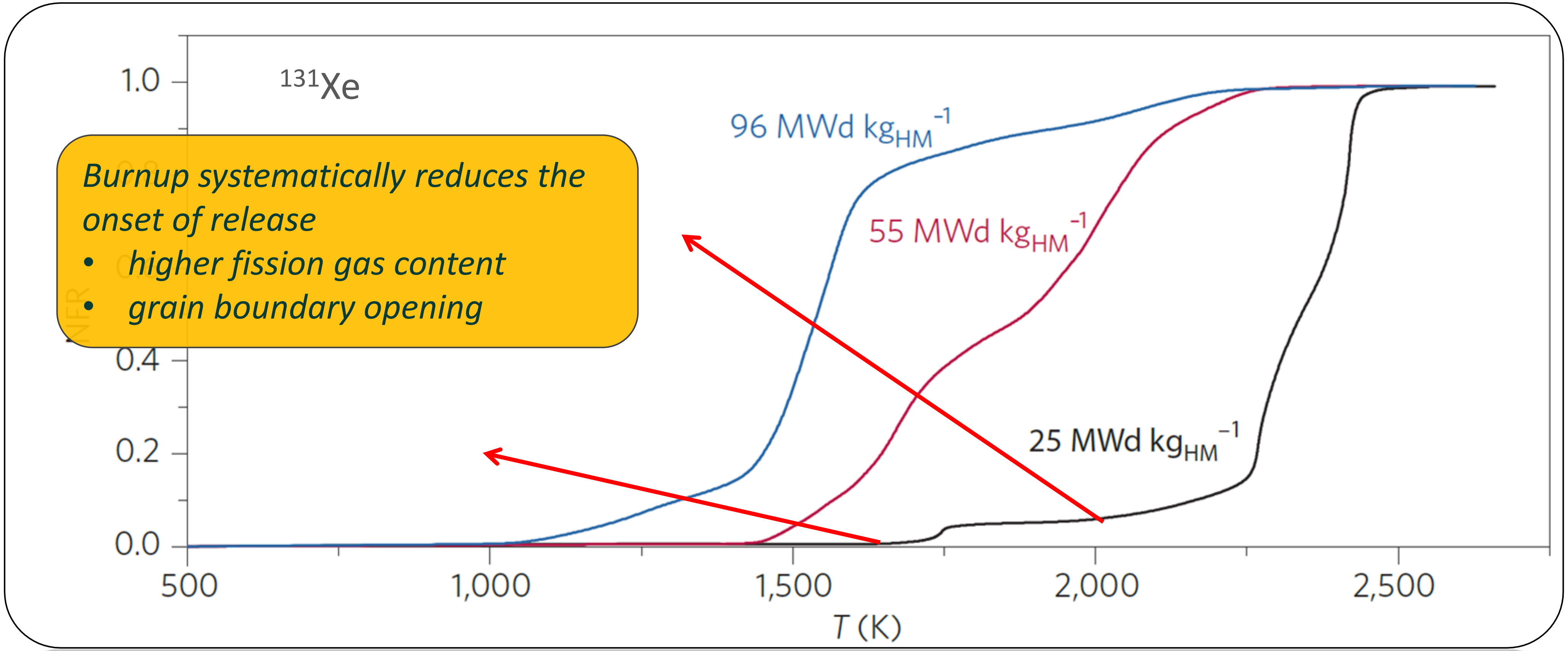
SEM after heating at T = 1800 K in KEMS



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

Irradiation behaviour: Fission Product Release

Konings et al., Nature Materials 14 (2015) 247



NFR = Normalised release fraction

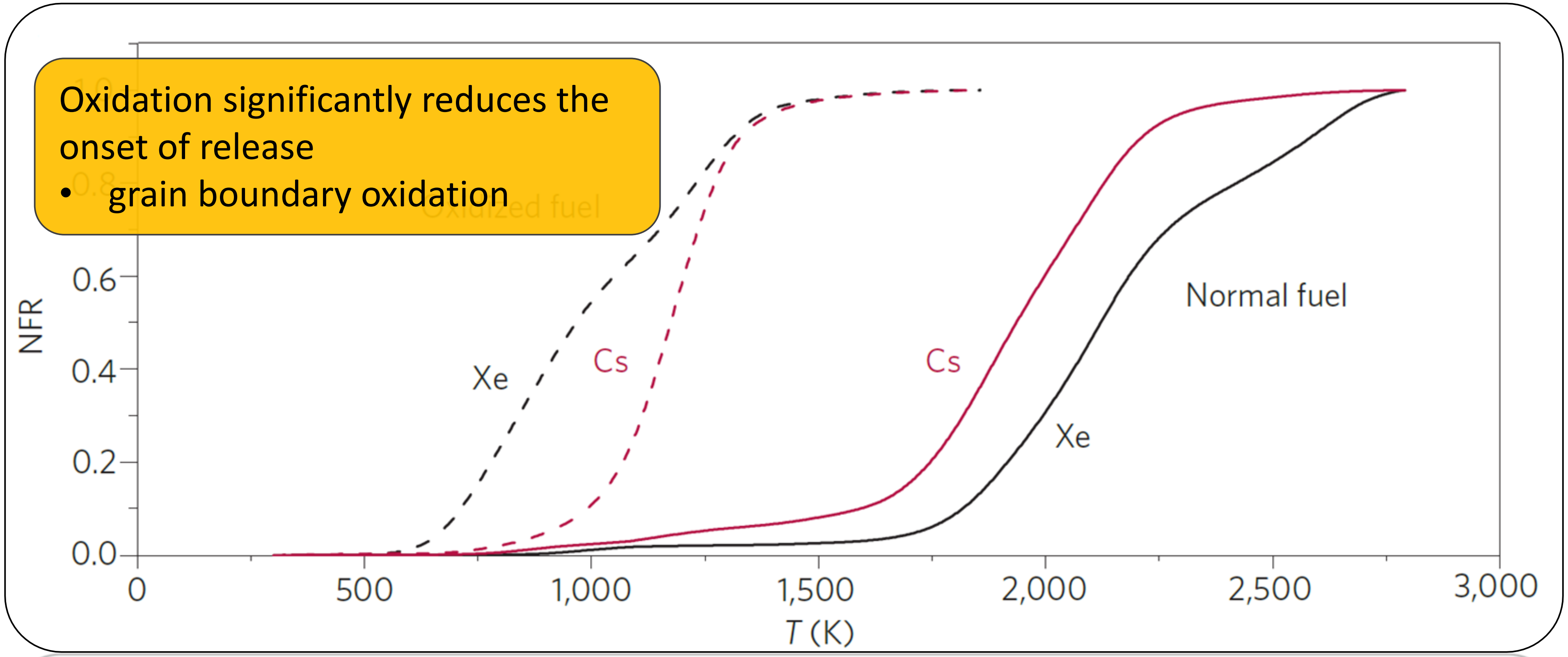
Source: Konings et al. Nature Materials 14 (2015) 247

Irradiation behaviour: Fission Product Release

Konings et al., Nature Materials 14 (2015) 247

Oxidation significantly reduces the onset of release

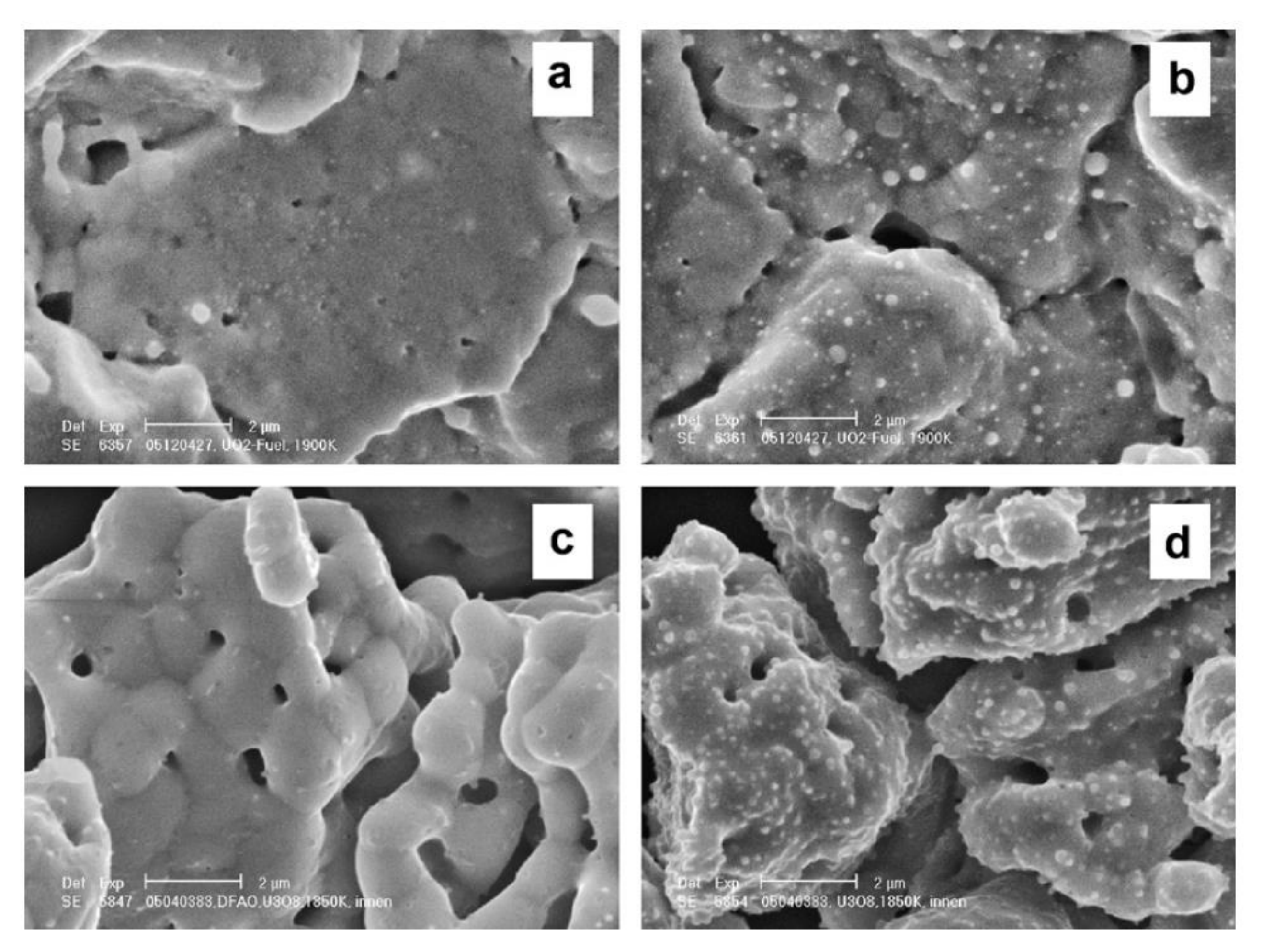
- grain boundary oxidation



Source: Konings et al. Nature Materials 14 (2015) 247

Irradiation behaviour: Fission Product Release

SEM after annealing
at T = 1900 K
Un-oxidised



SEM after annealing
at T = 1900 K
Pre-oxidised

Source: Hiernaut et al., J. Nucl. Mater. 372 (2008) 215

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Irradiation behaviour: CsI formation?

$Cs + ½I_2(g) = CsI$

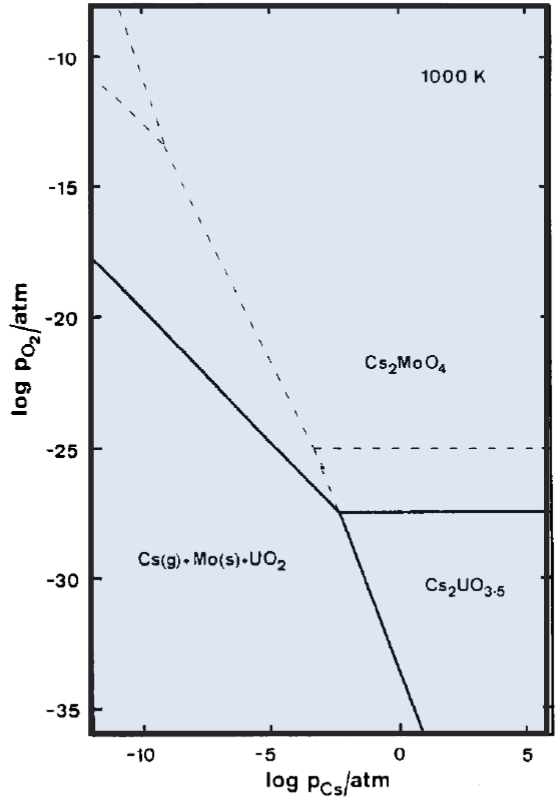
$2Cs + 2Te = Cs_2Te$

$2Cs + MoO_2 + ½O_2(g) = Cs_2MoO_4$

$2Cs + UO_2 + ½O_2(g) = Cs_2UO_4$

Thermodynamically stable

Can form at low oxygen potential

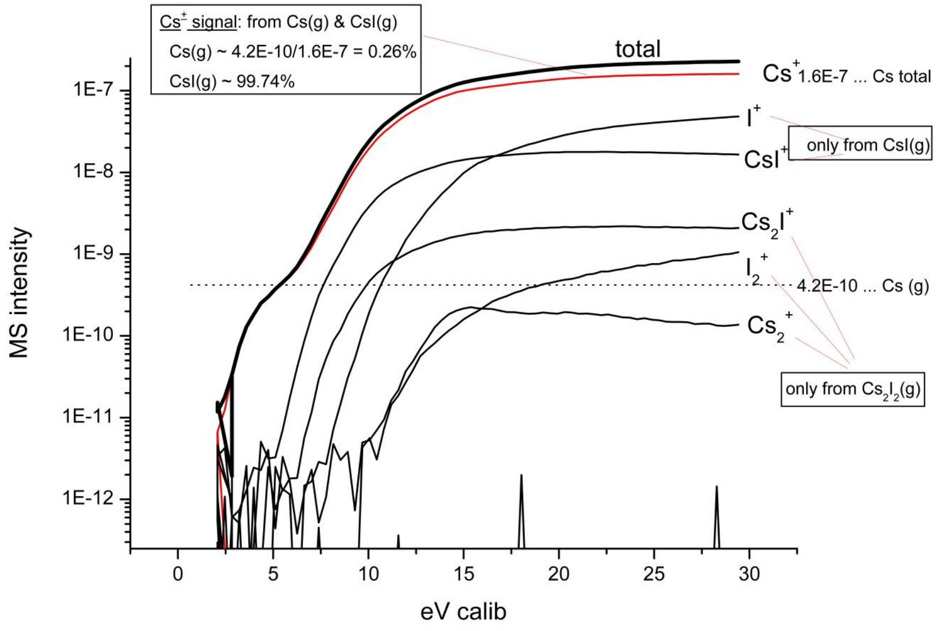


Irradiation behaviour: CsI formation?

Zappey et al., to be published

Cesium iodide

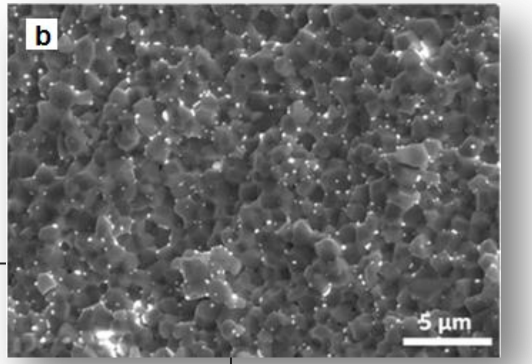
- Fragments: Cs^+, I^+, CsI^+ (monomer), Cs_2I^+, Cs_2^+, I_2^+ (dimer)
- $I^+/CsI^+ \approx 3:1$
- Parallel release profiles

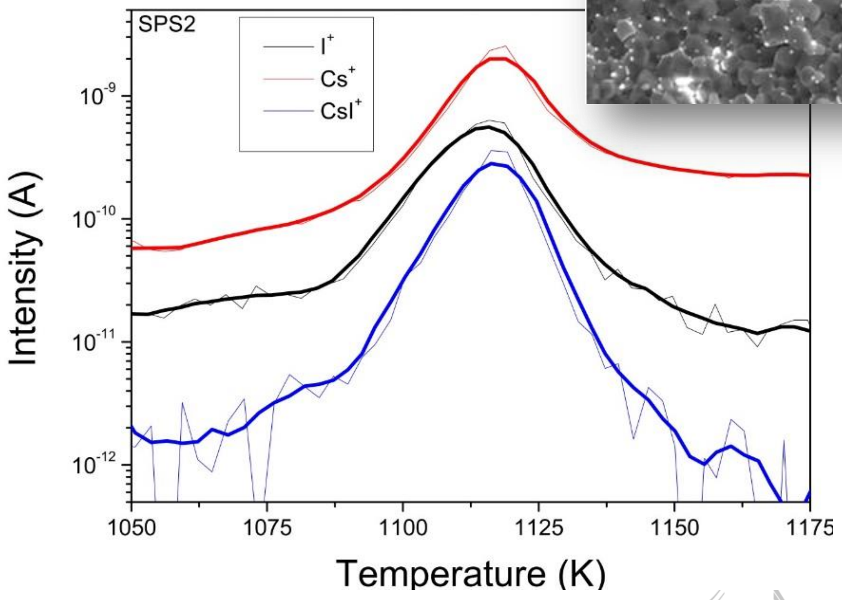


Source: European Communities ©

Simulated Fuel (UO_2+CsI)

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





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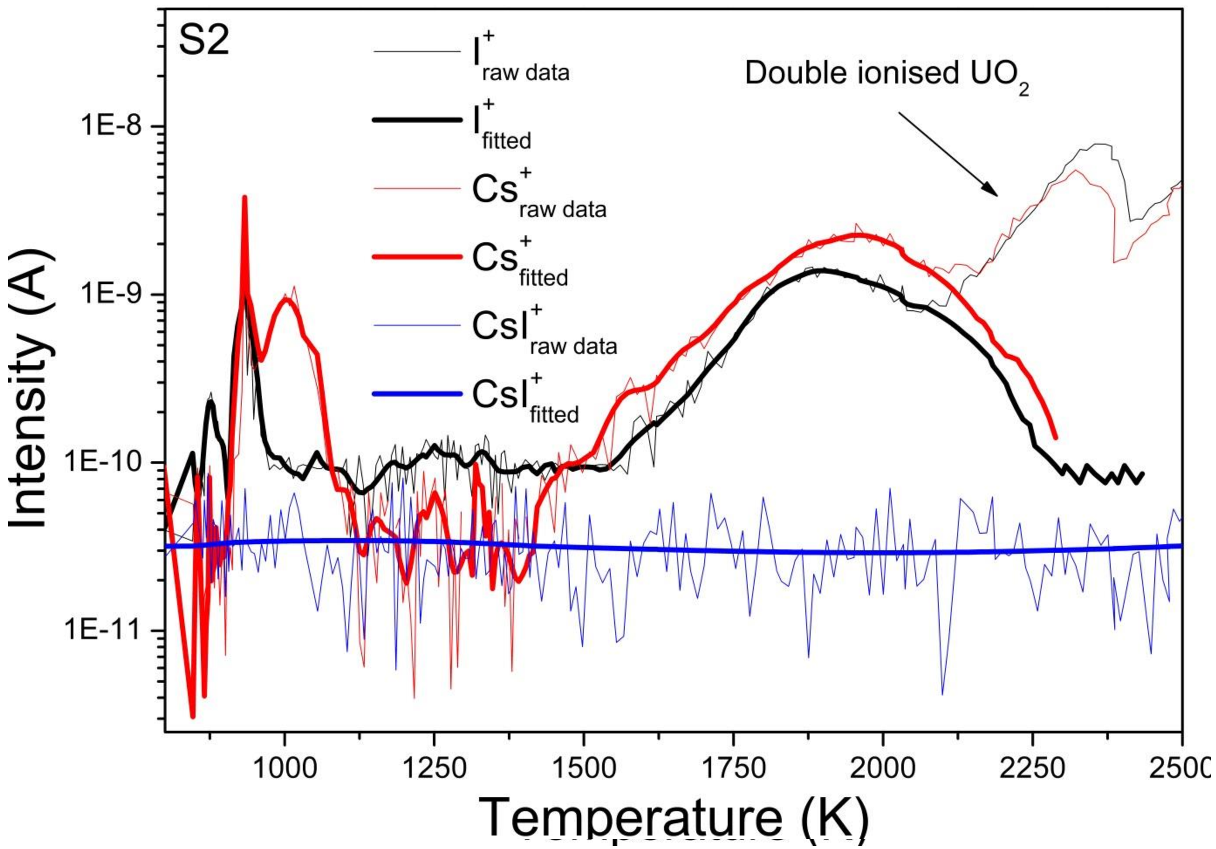
Irradiation behaviour: CsI formation?

Zappey et al., to be published



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?*

Nuclear Fuel: What affects the margins?

Fuel Composition

Microstructure

Radiation effects

Fuel Burn up

Mechanical evolution

Further reading

- R.J.M. Konings, T. Wiss and C. Guéneau, Nuclear Fuels. In: The Chemistry of Actinides and Transactinide Elements, 4th Edition (L.R. Morss, J. Fuger and N.M. Edelstein, Eds.), Volume 6, Chapter 34, p. 3665-3812, Springer Netherlands, 2010.
- R.J.M. Konings (Ed.), *Comprehensive Nuclear Materials*. Elsevier Science & Technology, Oxford, 2012.

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Thank you

Any questions?

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