Radiation damage studies: fuels and actinides compounds

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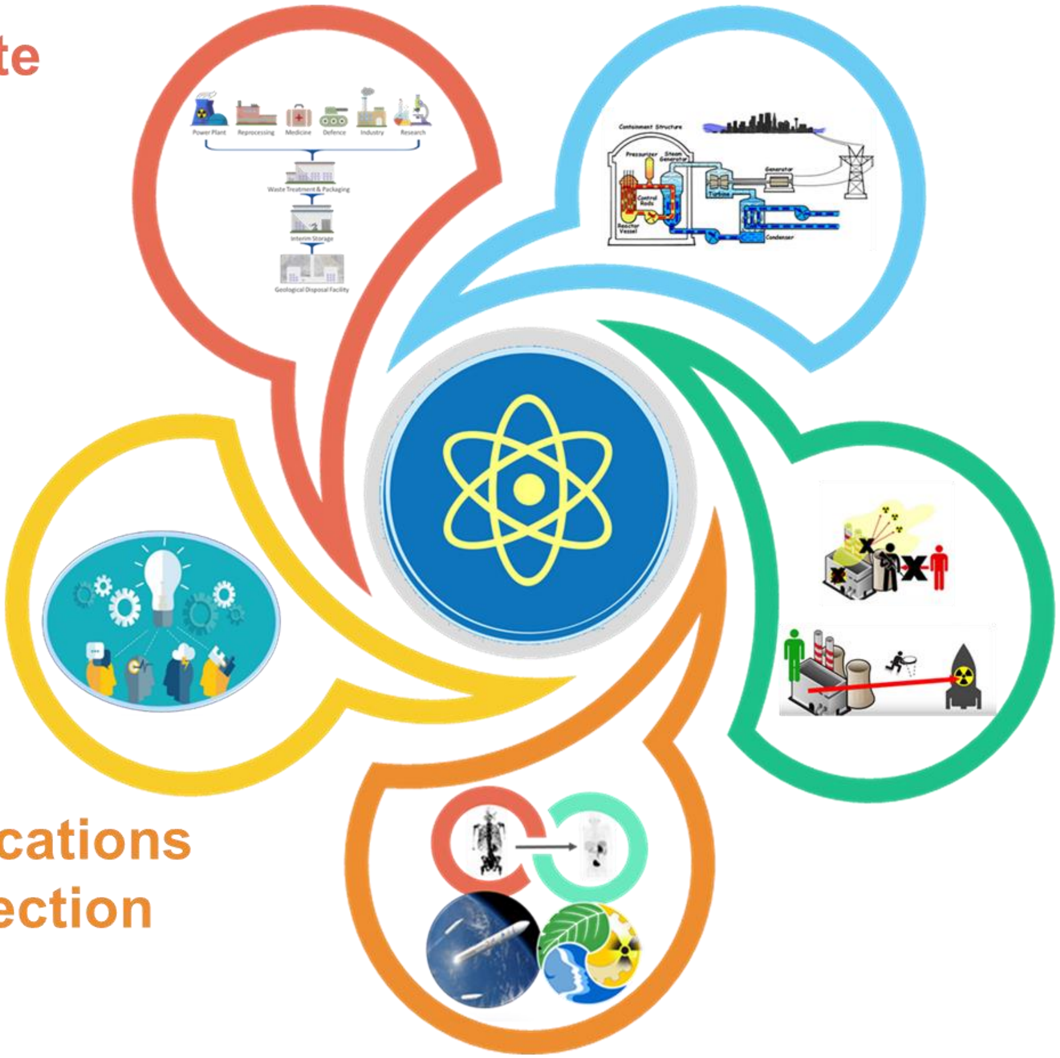
European Summer School, 1 - 5 July 2024 (Strasbourg- Karlsruhe) -Radiation Measurements and Radiochemistry in Environment and Decommissioning

Joint Research Centre

JRC EURATOM Horizon Europe
Research, Training & Education Thematic Areas



Radioactive Waste Management

Deep Geological Disposal
Extended Interim Storage
New Waste Forms (ATF, SMR)
Regulatory framework
E&T, KM, Open Access

Nuclear Knowledge & Competence

Maintain Competence (E&T)
Human Resources Observatory
Support JRC Open Access
Reference Data & Standardization
Innovation & Technology
from Research to Industry

Non-power Applications & Radiation Protection

Medicine, Environment, Space
EU beating Cancer
Standardization
Accelerators
Open access, E&T

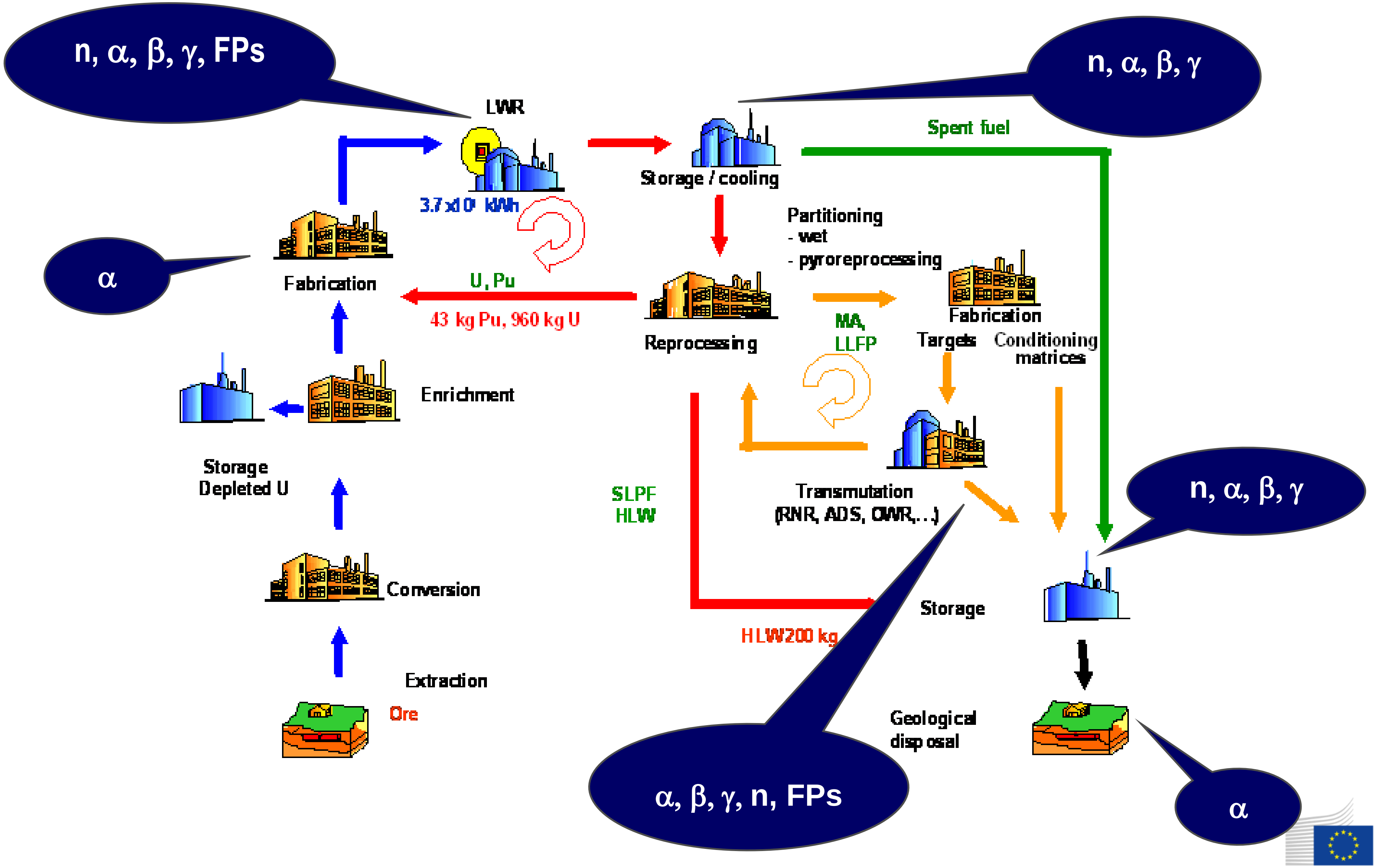
Nuclear Safety of Nuclear Power Plants

Nuclear reactor safety
Update of safety regulations
LTO, SMR, Gen-IV
Innovative materials
Fuel development and testing
Infrastructures: JHR, HFR and Open Access
Emergency Preparedness

Nuclear Safeguards and Security

EU Safeguards obligations
EU nuclear non-proliferation
Synergies with Security Union & Defense
International Partnership
E&T, KM

Radiation Damage in the nuclear fuel cycle



n, α, β, γ, FPs

n, α, β, γ

α

n, α, β, γ

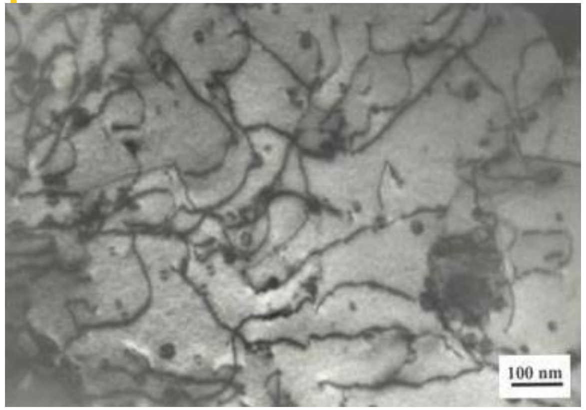
α, β, γ, n, FPs

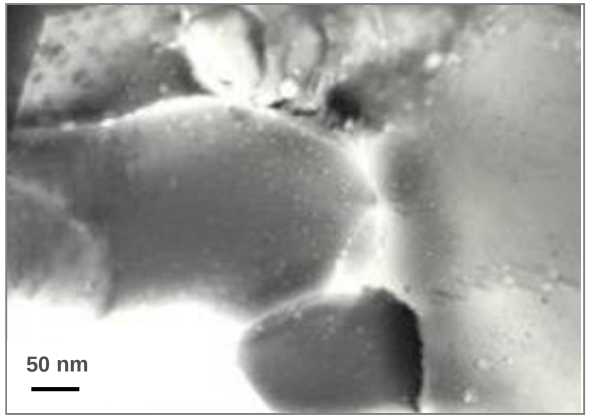
α

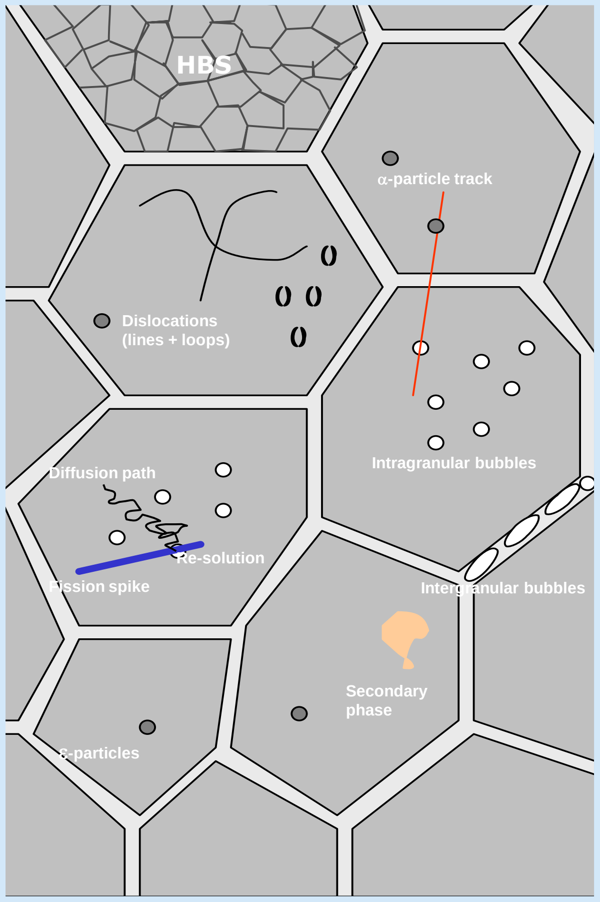
Context

- Nuclear materials lifetime is (partly) reduced due to radiation damage build-up
- The properties of actinide containing materials are impacted by thermal gradient, chemical composition change, mechanical stress, and radiation damage.
- Understanding and predicting the evolution of materials could help in the design of more stable materials but also allow to design migration barriers, waste confinement matrix, accident tolerant fuels (ATF), transmutation fuels...

Rational







During storage
or
During operation

Fundamental data
thermodynamic solubility, diffusion coefficients, HBS

Influence of: grain boudaries, bubble formation, radioactive decay, fission, temperature, gas re-solution, impurities, microstructure,oxygen potential,..

On: gas mobility, release, material property changes (mechanical, integrity, thermo-physical)

Layout

- General on radiation damage processes / defects

- Radioisotopic Thermal Generator (RTG)
  - Am
  - Pu

- Ageing of irradiated fuel (spent fuel)
  - Acceleration of damage by alpha-doping
  - Archive materials
  - Irradiated transmutation fuel

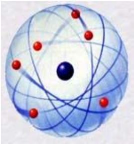
“Les rayons alpha sont des projectiles matériels susceptibles de perdre de leur vitesse en traversant la matière” (1900)

H. Becquerel

P. Curie

M. Curie-Slodowska

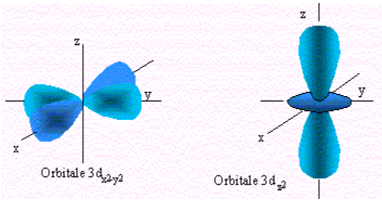
E. Rutherford

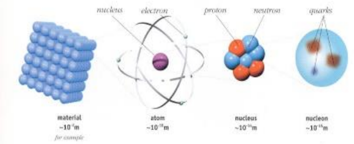


N. Bohr

E. Fermi

E. Schrödinger





1896

1911

today….

Interaction of a charged particle with matter

Inelastic collisions with an electron

main process of energy loss producing excitation and ionization

Inelastic collisions with a nucleus

Bremsstrahlung and coulombic excitation

Elastic collisions with a nucleus

Rutherford diffusion

Elastic collisions with an electron

Particle/ion-matter interactions

Slowing down of a particle/ion in a target

- history of the particle

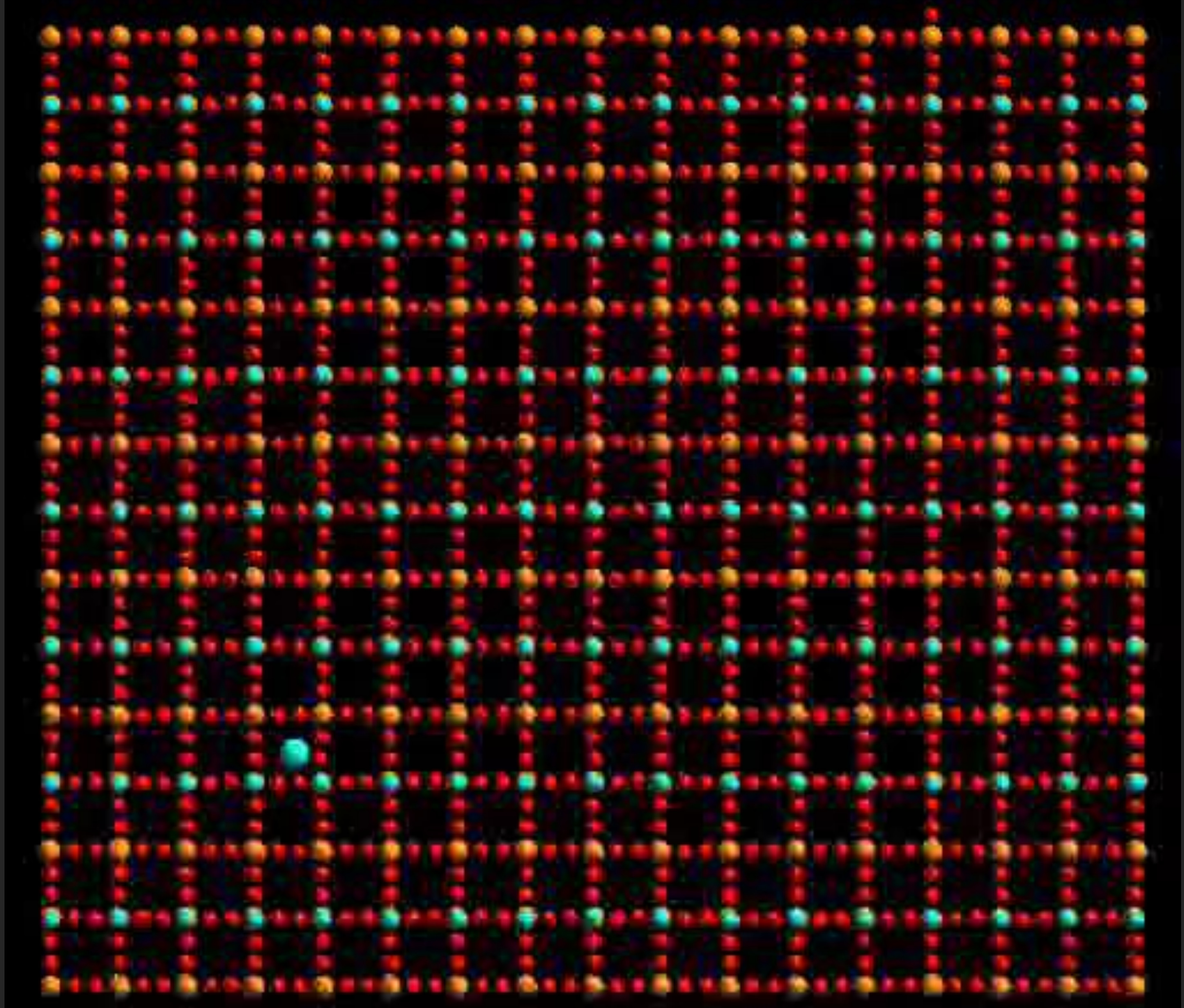
  energy loss of a particle, range, interactions

- history of the target atoms

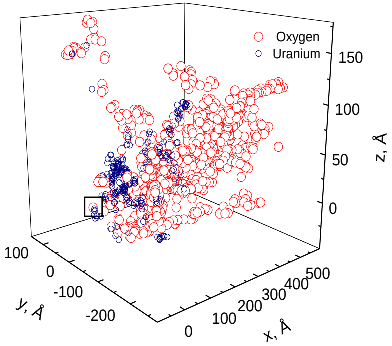
  displacements, recombinations, ionization, excitation, radiation damage build-up

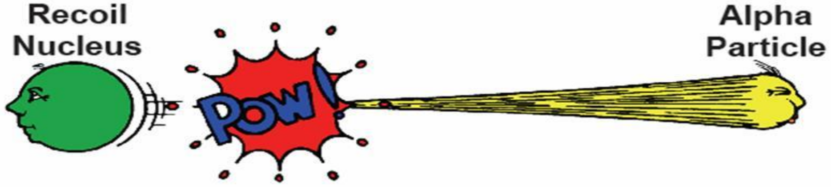
Areas of interest

Nuclear industry, nuclear medicine, space applications, semi-conductor, geology...

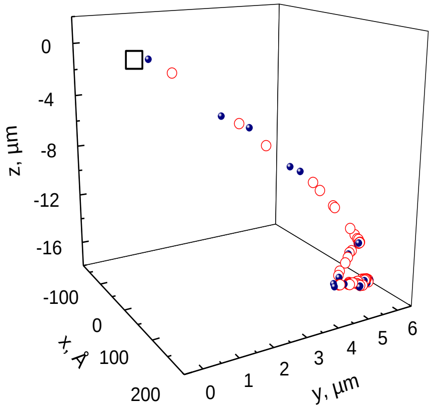


Displacement energy - cascades





Courtesy Bill Weber



Communication of kinetic energy to a lattice atom sufficient to break bonds - lattice elastic forces cannot bring it back

☞ energy to transfer, E_d, threshold displacement energy.

☞ formation of a Frenkel pair

Dependence on lattice vibrations (temperature), crystallographic directions.

If sufficient energy is transferred to the primary knock-on atom (pka) further collisions/displacements can occur.

☞ Displacement cascade

Number of displaced atom, n(T) per pka (Kinchin and Pease, 1955)

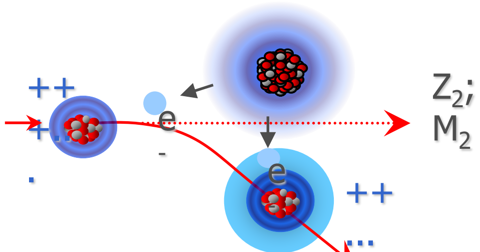
$n(T) = 0$ if $T < E_d$

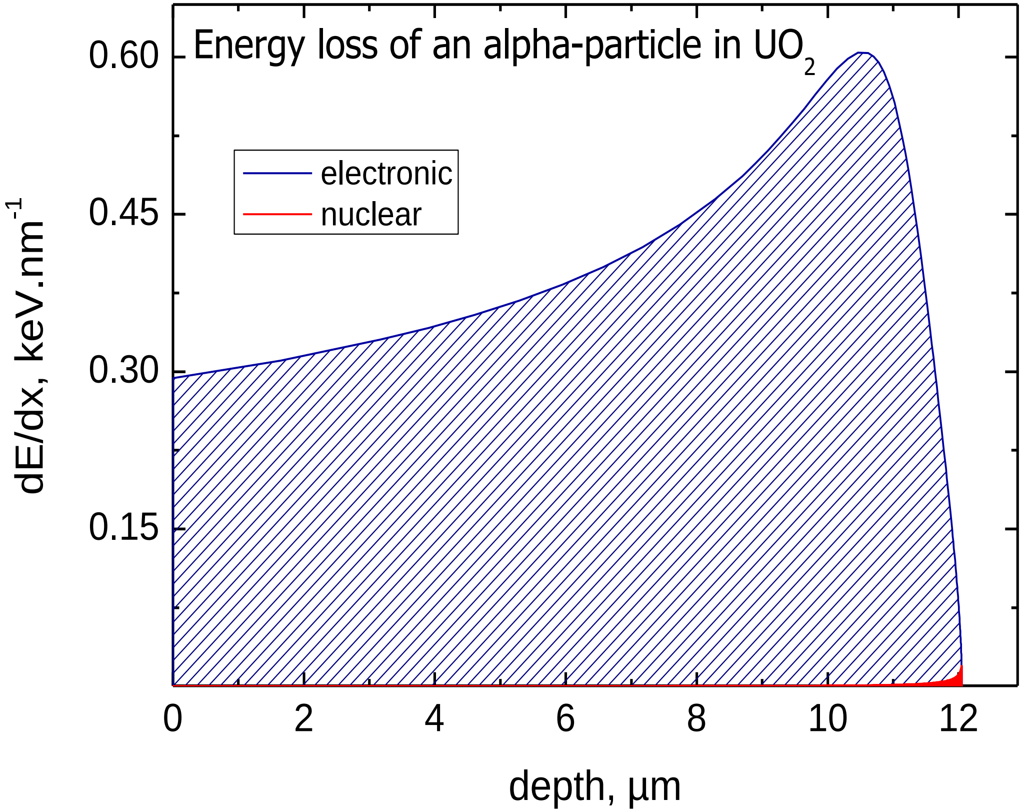
$n(T) = 1$ if $E_d < T < 2\ E_d$

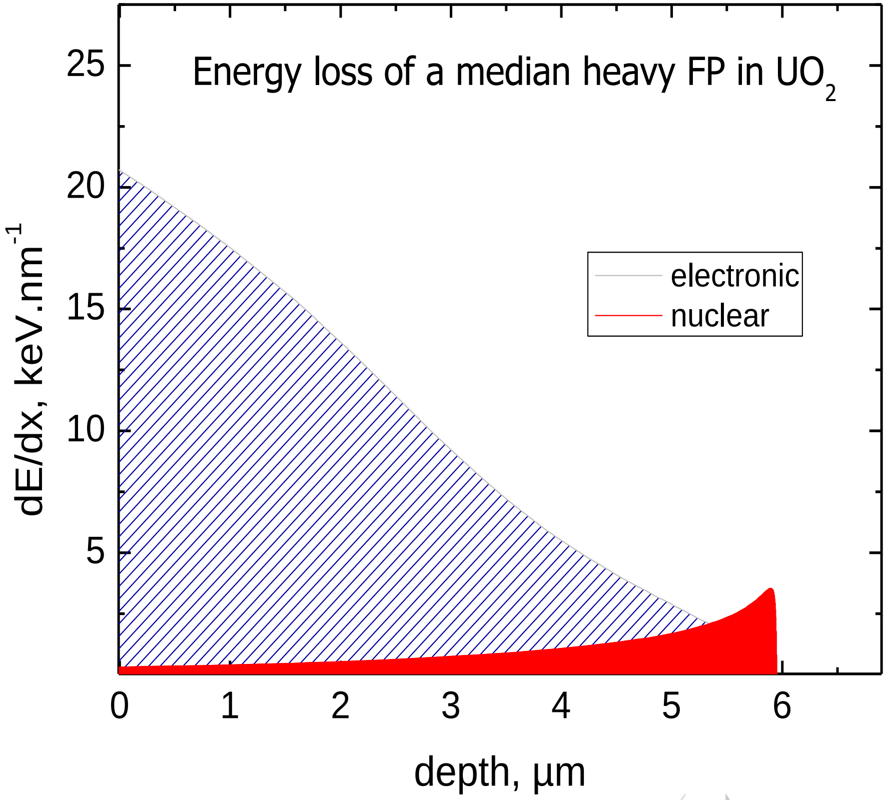
$n(T) = T/2\ E_d$ if $T \geq 2\ E_d$

Better approximation: $n(T) = 0.8\ (T-E_{ioniz})/2\ E_d$ for $T \geq 2\ E_d$

Energy loss in UO_2





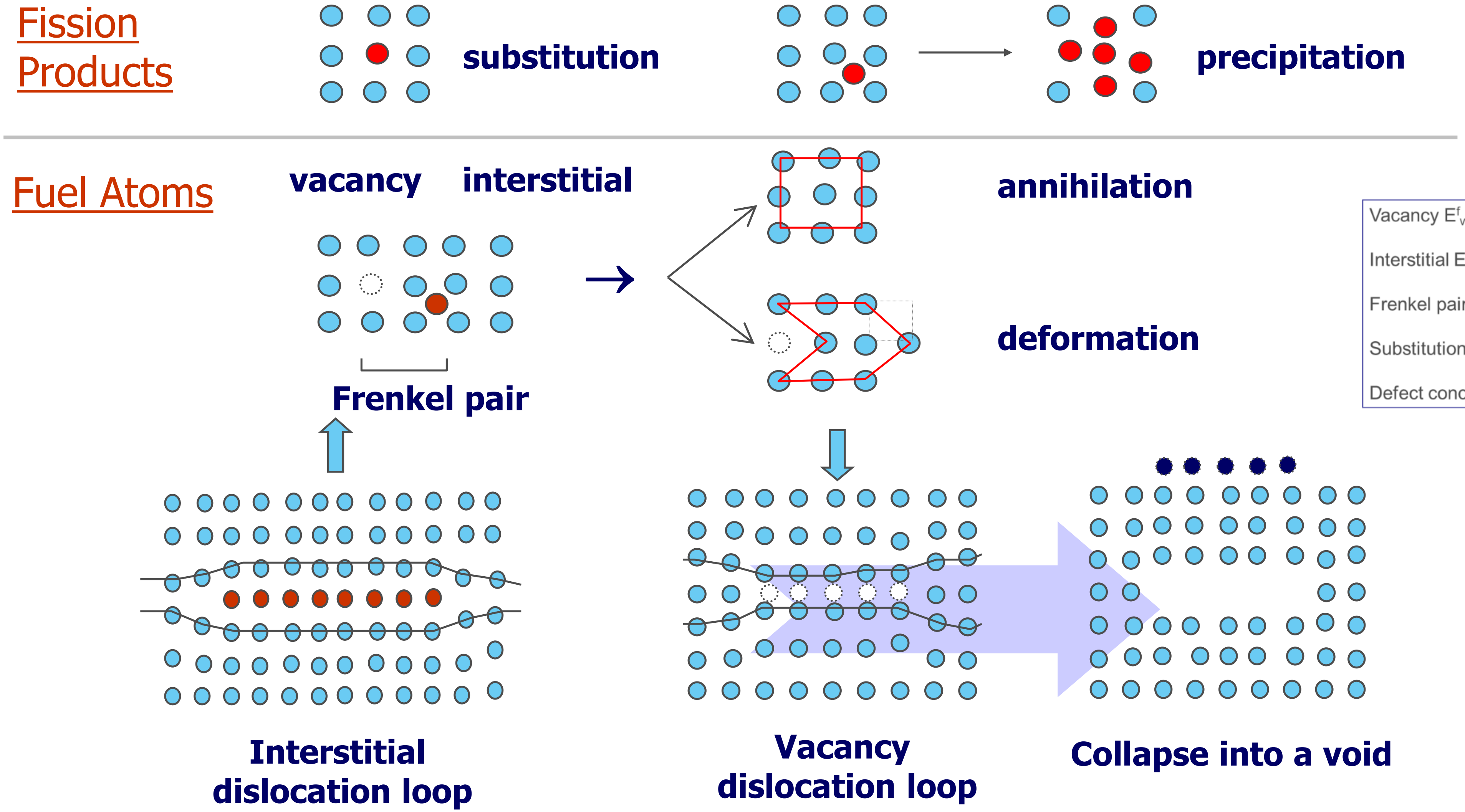


SRIM: Ziegler, J. F., Biersack, J.P., Littmark, U., The Stopping and Range of Ions in Solids, Pergamon Press, Oxford, 1985

Range of different particles/ions

	Energy, keV	Range, μm	dE/dx, Nucl./Elec.	Defects formed
Light FPs	95000	9	0.03/0.97	40000
Heavy FPs	67000	7	0.06/0.94	60000
α-particles	5000	12	0.01/0.99	200
Recoil nucleus	95	0.02	0.90/0.10	1500
Cosmic rays (p^+)	10^{17} 10^6 (typical)	Light years !		

Defect types



Vacancy E^f_v ~ 1 eV

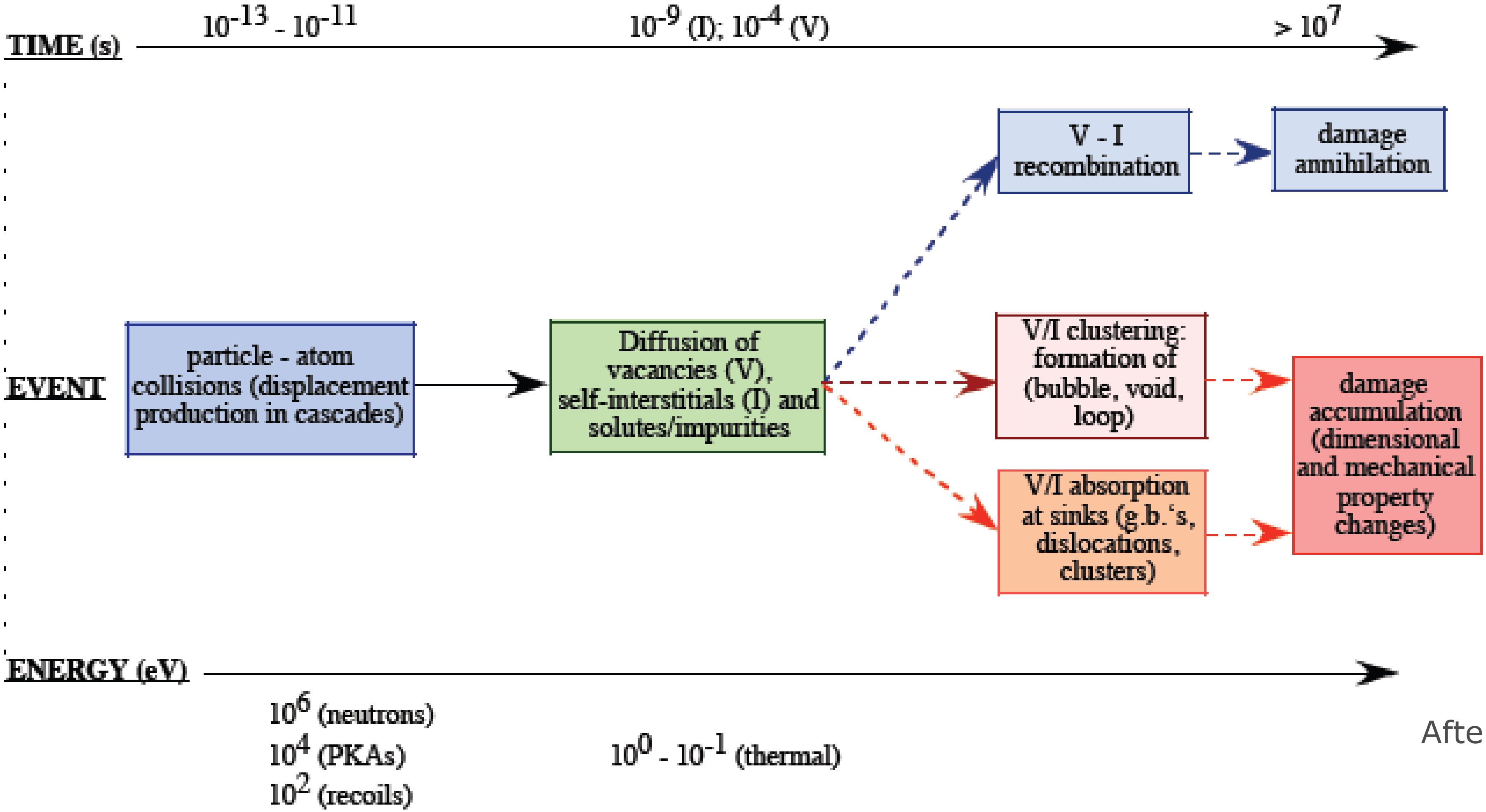
Interstitial E^f_i ~ 3-5 eV

Frenkel pair: 4-6 eV

Substitutional

Defect concentration is T dependent

Damage evolution



After B. Wirth

Stability of defects

Low temperature:

athermal recombination by newly produced defects
(overlap of displacement cascades)

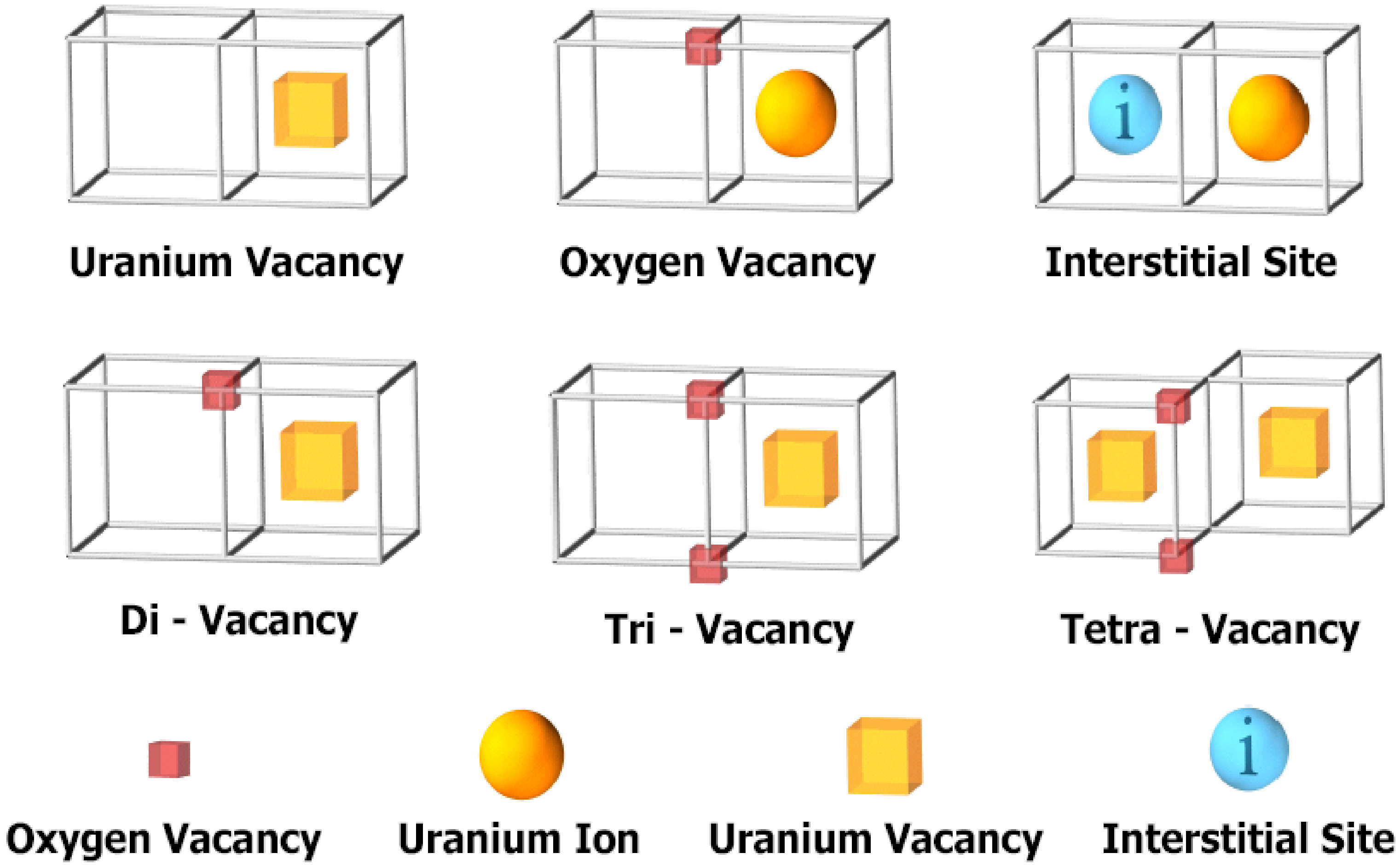
Higher temperature:

migration of interstitials
- correlated recombination (interstitial with "its" vacancy)
- non-correlated recombination
- trapping on impurity
- trapping on sinks (dislocations, clustering)

The number of remaining defects is lower than the total number of displaced atoms.

e.g. in operating fuel values as high as 1500 dpa are reached during the fuel lifetime and the material remains crystalline !!

Possible solution sites for FP’s in $UO_{2\pm x}$

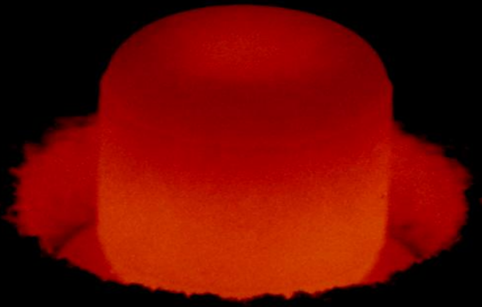


Source: R. Grimes (2005), ITU Summer School

Radioisotopic Thermal Generators (RTG) Electrical Power Sources (EPS)

$T_{1/2}$ $^{238}\mathrm{Pu} \sim 87.74$ y
Peak power 390 $\mathrm{mW.g^{-1}}$

$T_{1/2}$ $^{241}\mathrm{Am} \sim 432$ y
Peak power 114 $\mathrm{mW.g^{-1}}$

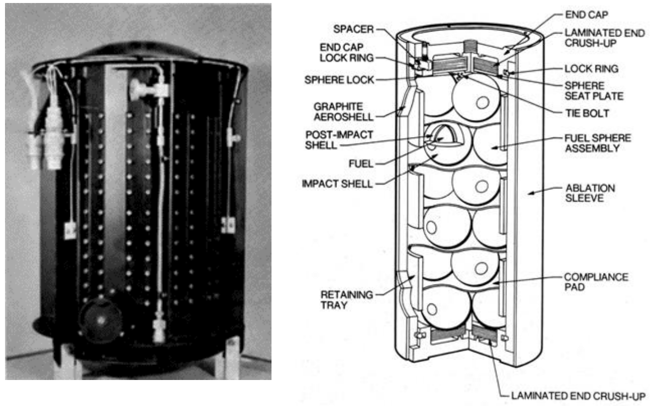


RTG

In addition to spacecraft, the Soviet Union constructed many uncrewed lighthouses and navigation beacons powered by RTGs.
The United States Air Force uses RTGs to power remote sensing stations for Top-ROCC and SEEK IGLOO radar systems predominantly located in Alaska.
In the past, small "plutonium cells" (very small ^{238}Pu-powered RTGs) were used in implanted heart pacemakers to ensure a very long "battery life".

SNAP 3B in 1961 powered by 96 grams of plutonium-238 metal, aboard the Navy Transit 4A spacecraft

Aniva lighthouse was built by the Japanese in 1939, on a chunk of rock off the southern coast of Sakhalin



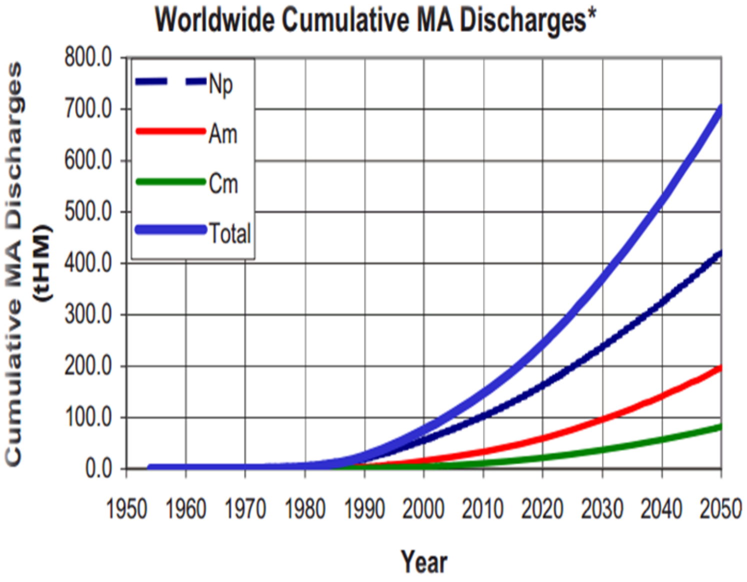
A common RTG application is spacecraft power supply. Systems for Nuclear Auxiliary Power (SNAP) units were used for probes that traveled far from the Sun rendering solar panels impractical. As such, they were used with Pioneer 10, Pioneer 11, Voyager 1, Voyager 2, Galileo, Ulysses, **Cassini**, New Horizons, and the Mars Science Laboratory. RTGs were used to power the two Viking landers and for the scientific experiments left on the Moon by the crews of Apollo 12 through 17

https://en.wikipedia.org/wiki/Radioisotope_thermoelectric_generator

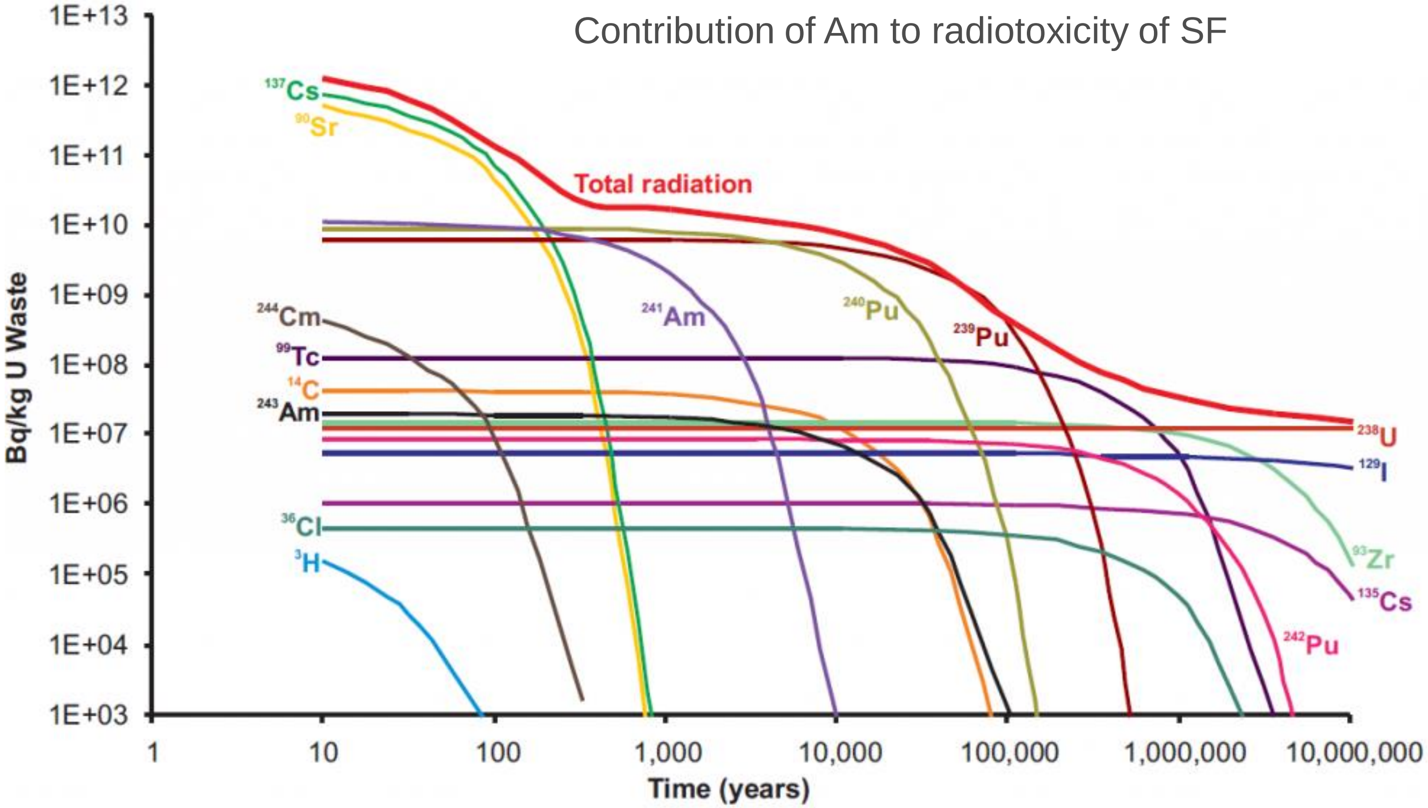
Americium

European Commission

Am inventory

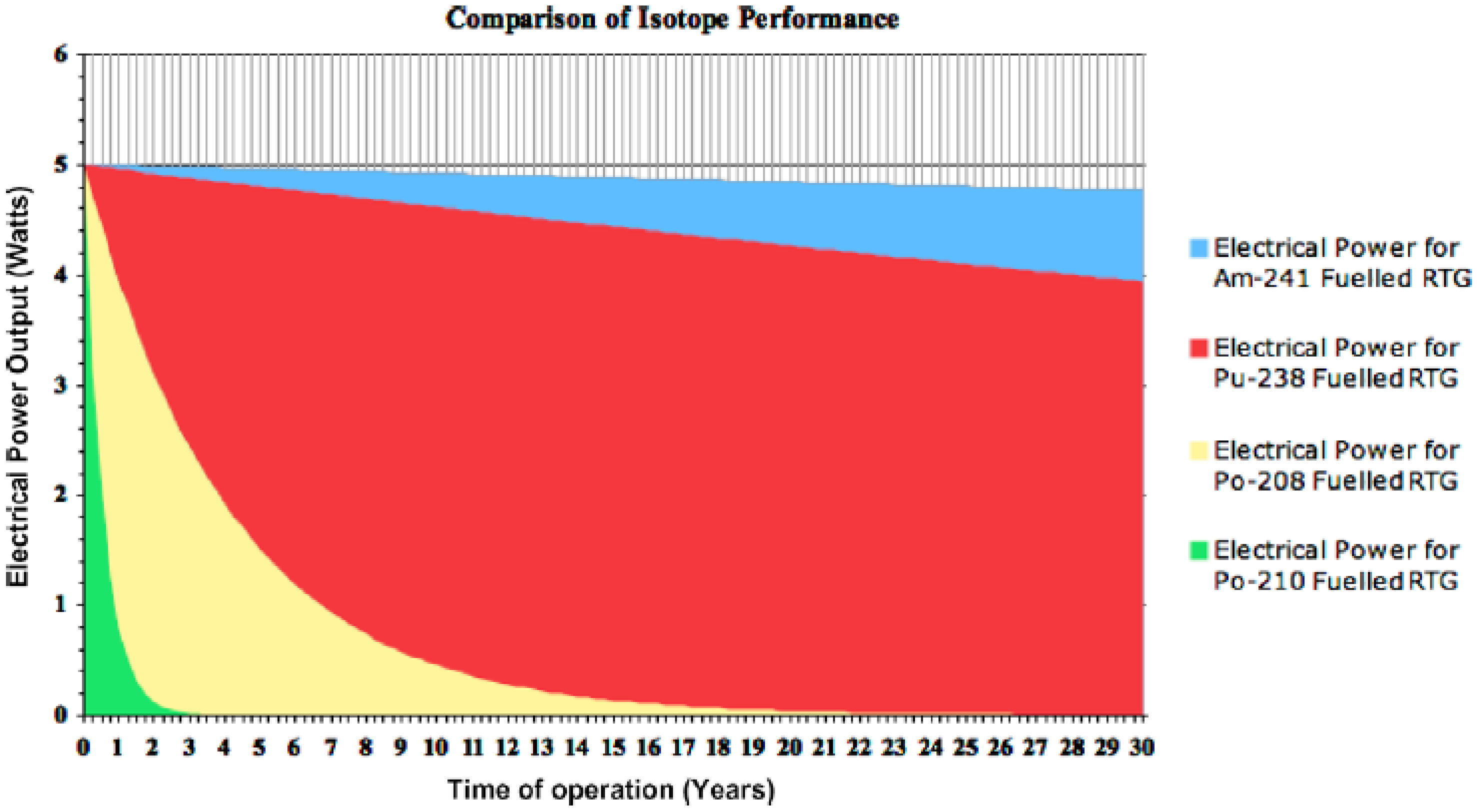


Source : Status of MA fuel development, IAEA, 2006



Contribution of Am to radiotoxicity of SF

EPS



Neutron yield 10x lower for the same thermal power for AmO_2
Mostly from (α, n) reactions on ^{17}O and ^{18}O
(Less shielding for equal O enrichment) i.e. less neutron damage from AmO_2

R.C. O’Brien et al. / Journal of Nuclear Materials 377 (2008) 506–521

Some properties of ^{241}Am

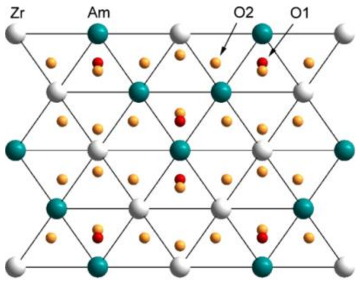
- α **emission**:   E=5486 keV (85%), E=5443 keV (13%)
- γ **emission:** E=60keV (36%)
- **X-emission:** L-shell 17 keV (40%)
- **Specific activity:** 127 GBq.g^{-1}
- **(α, n) reaction (n.s^{-1}.g^{-1}):** 5.321x10^{3} (natural oxide) 1.080x10^{2} (enriched oxide)
- **Shielding (1/10)**: 3mm steel

European
Commission

Different Am-compounds

- $AmAlO_3$ (perovskite, from spinel degradation in EFTTRA T4)

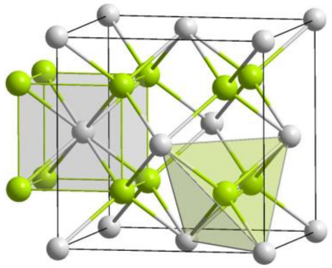
- Am_2O_3 (lattice swelling, Horlait et al. J. Sol. Chem., 2014 vol. 217, pp. 159-168)
- Evolution from type A to C



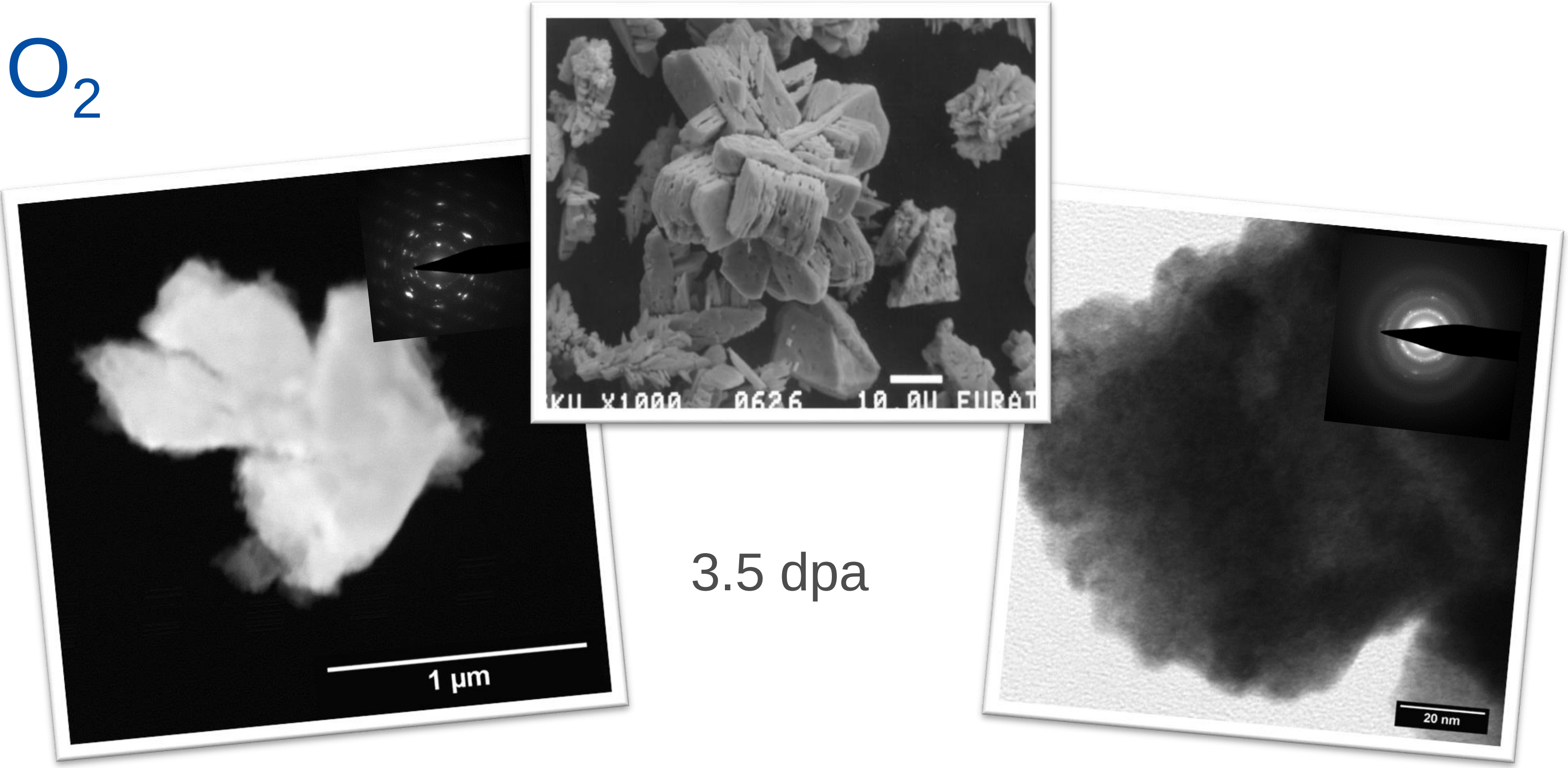
- $AmPO_4$ (monazite) Amorphization after 300 days

- $Am_2Zr_2O_7$ (pyrochlore) Transmutation

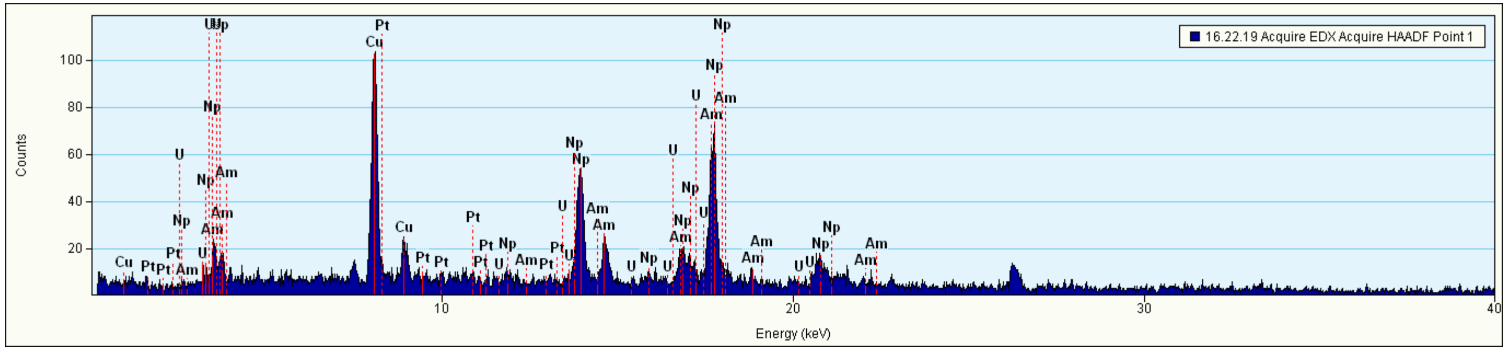
- **AmO_2**



AmO_2

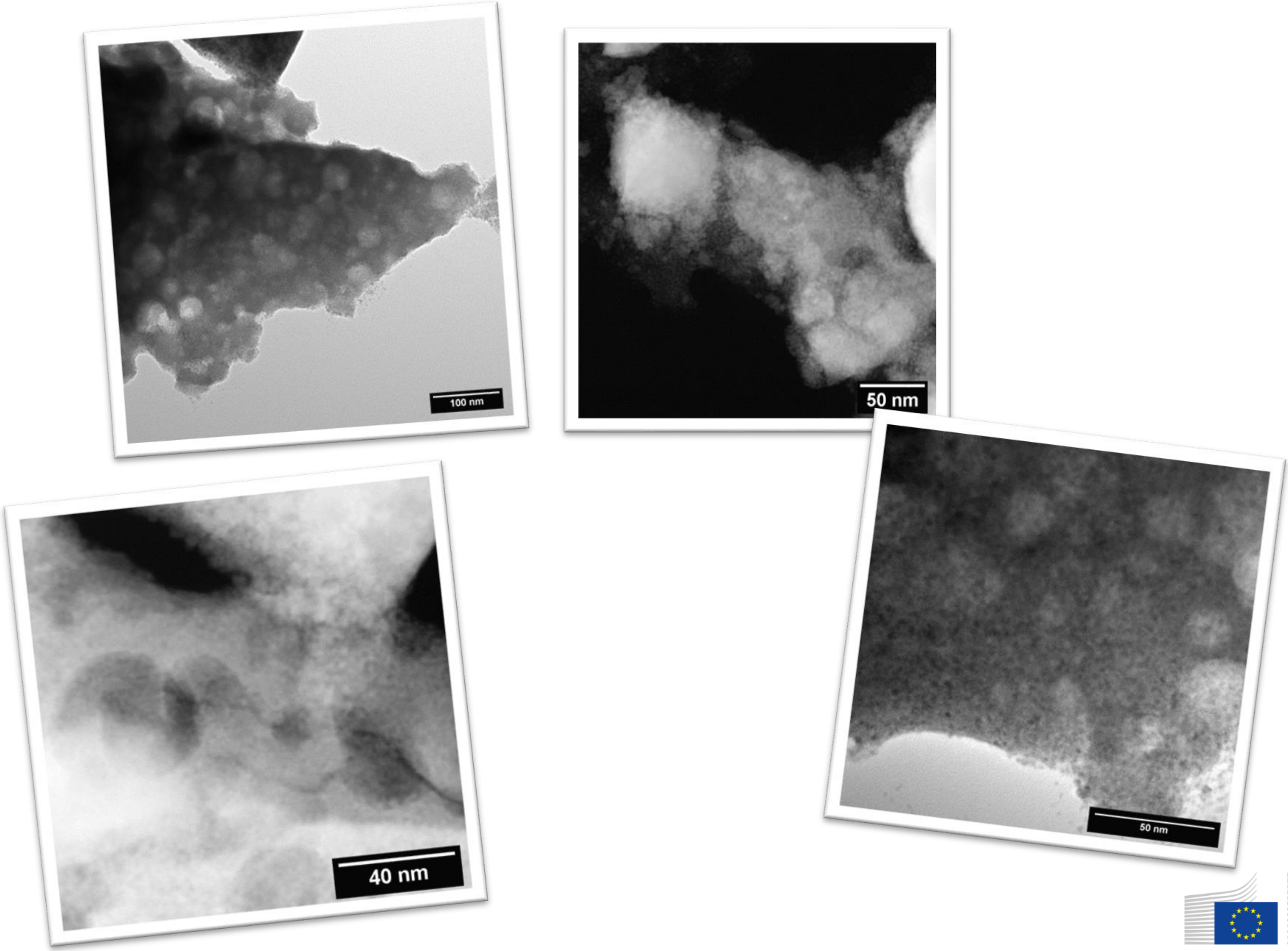

1 µm
KU X1000 0626 10.0U EURAT
20 nm

3.5 dpa


16.22.19 Acquire EDX Acquire HAADF Point 1
Counts
100
80
60
40
20
10
20
30
40
Energy (keV)
Pt
Cu
Np
U
Am
Pt

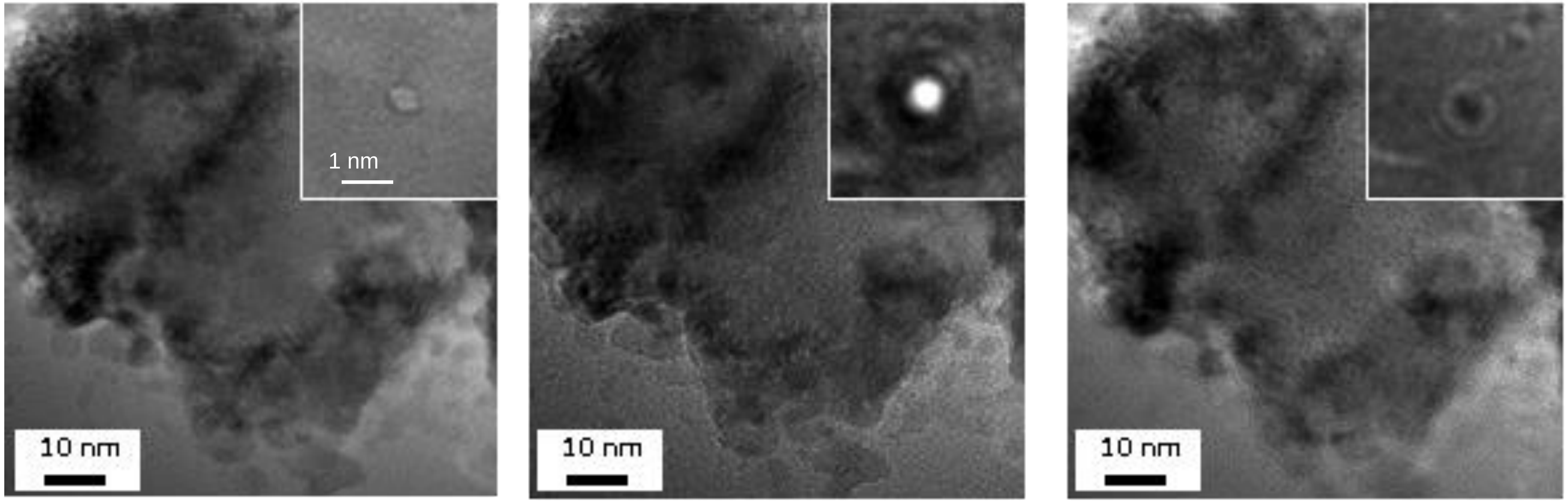
AmO_2

32 dpa $1.4 \times 10^{20}\ \alpha.g^{-1}$



AmO_2

32 dpa $1.4 \times 10^{20}\ \alpha.g^{-1}$

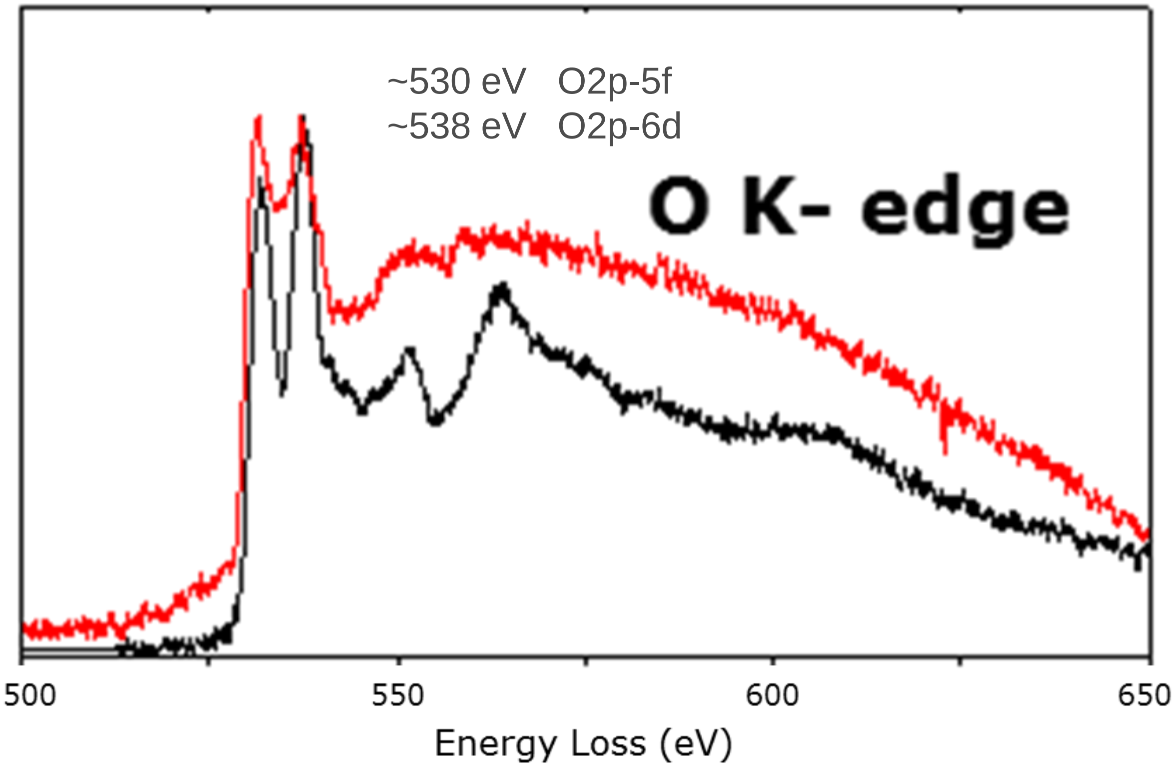


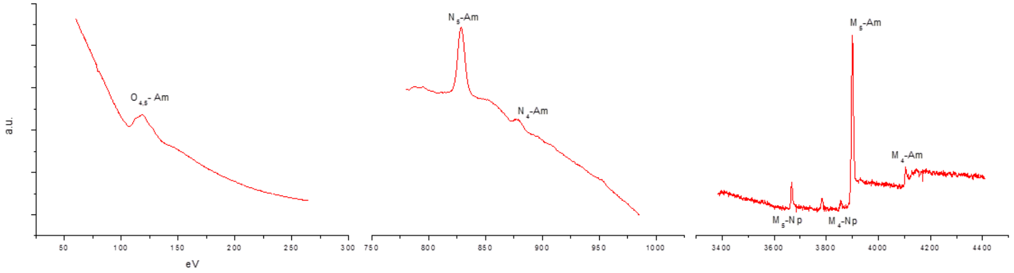
Cavities/bubbles ~ $10^{23}\ m^{-3}$

AmO_2 EELS bef. & aft. annealing

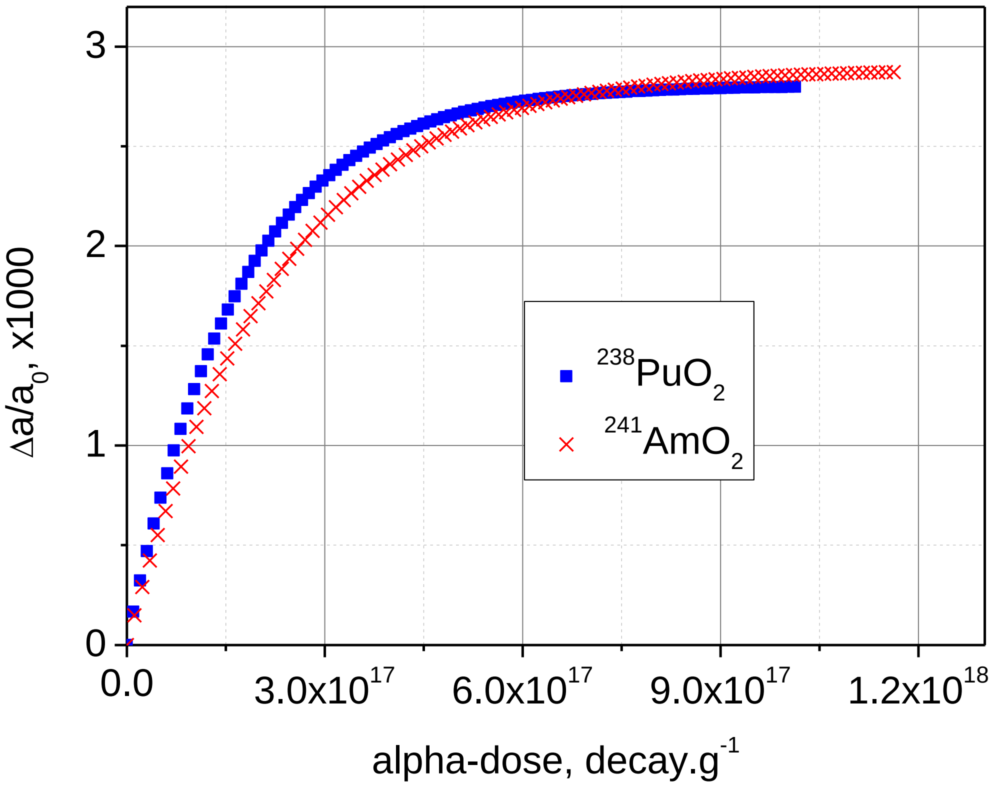
32 dpa 1.4x10^{20} α.g^{-1}

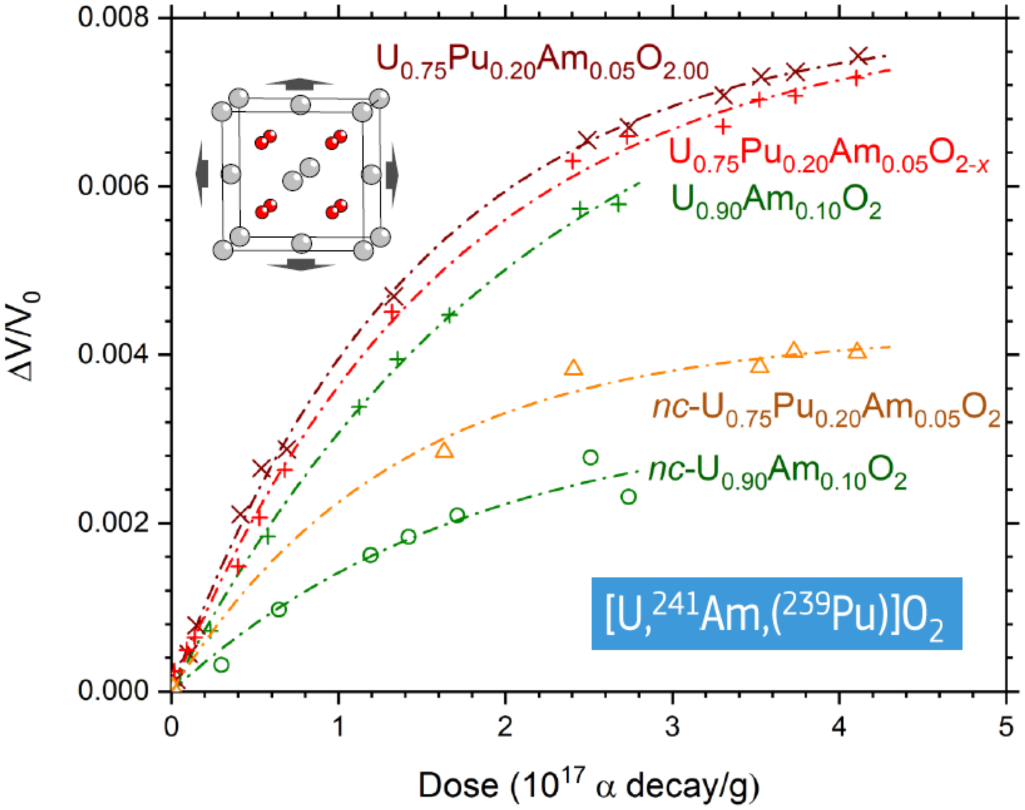
oxygen 2p empty states are probed





Lattice parameter evolution of aged AmO_2 or Am-compounds





- W. Weber, Alpha-irradiation damage in CeO_2, UO_2 and PuO_2, Radiat. Eff., 83 (1984) 145-156.
- Hurtgen, C., Fuger, J. (1977), Self-irradiation Effects in Americium Oxides, Inorg. Nucl. Chem. Lett. 13, 179-188

Summary aged AmO_2

- EELS shows the lack of short range order around oxygen atoms (multiscattering peaks)

- two peaks at ~530 eV and ~538 eV are still clearly observed which is consistent with the FCC structure observed

- Bimodal distribution of bubbles

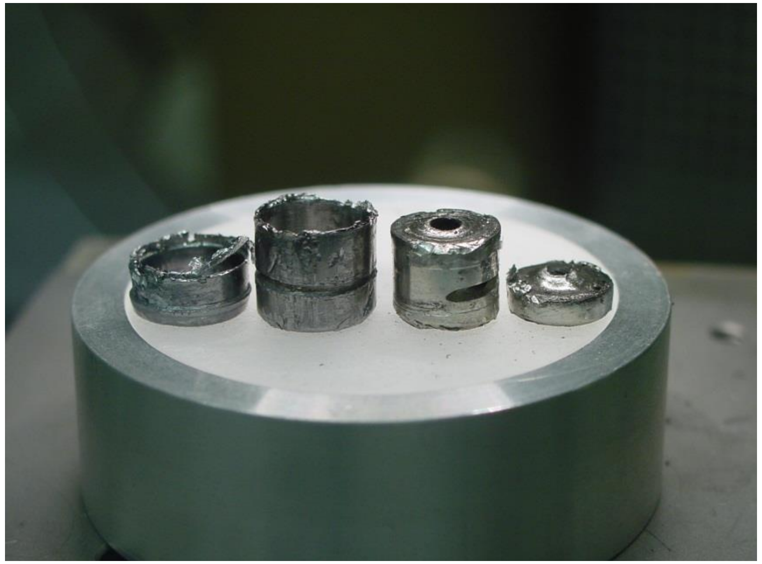
- No (high conc.) extended defects but polygonized areas

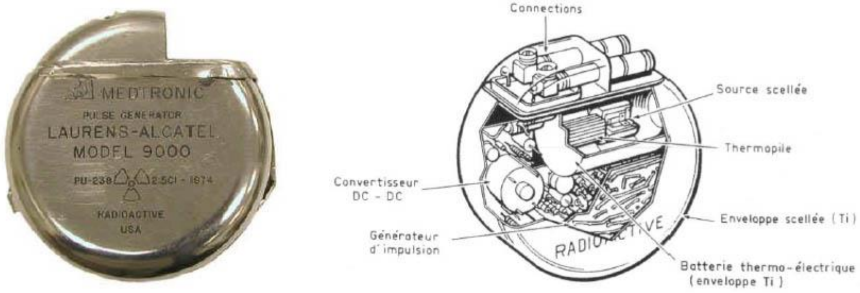
- Saturation of the lattice parameter around 0.3%

Plutonium

European Commission

Examination pacemaker RTG

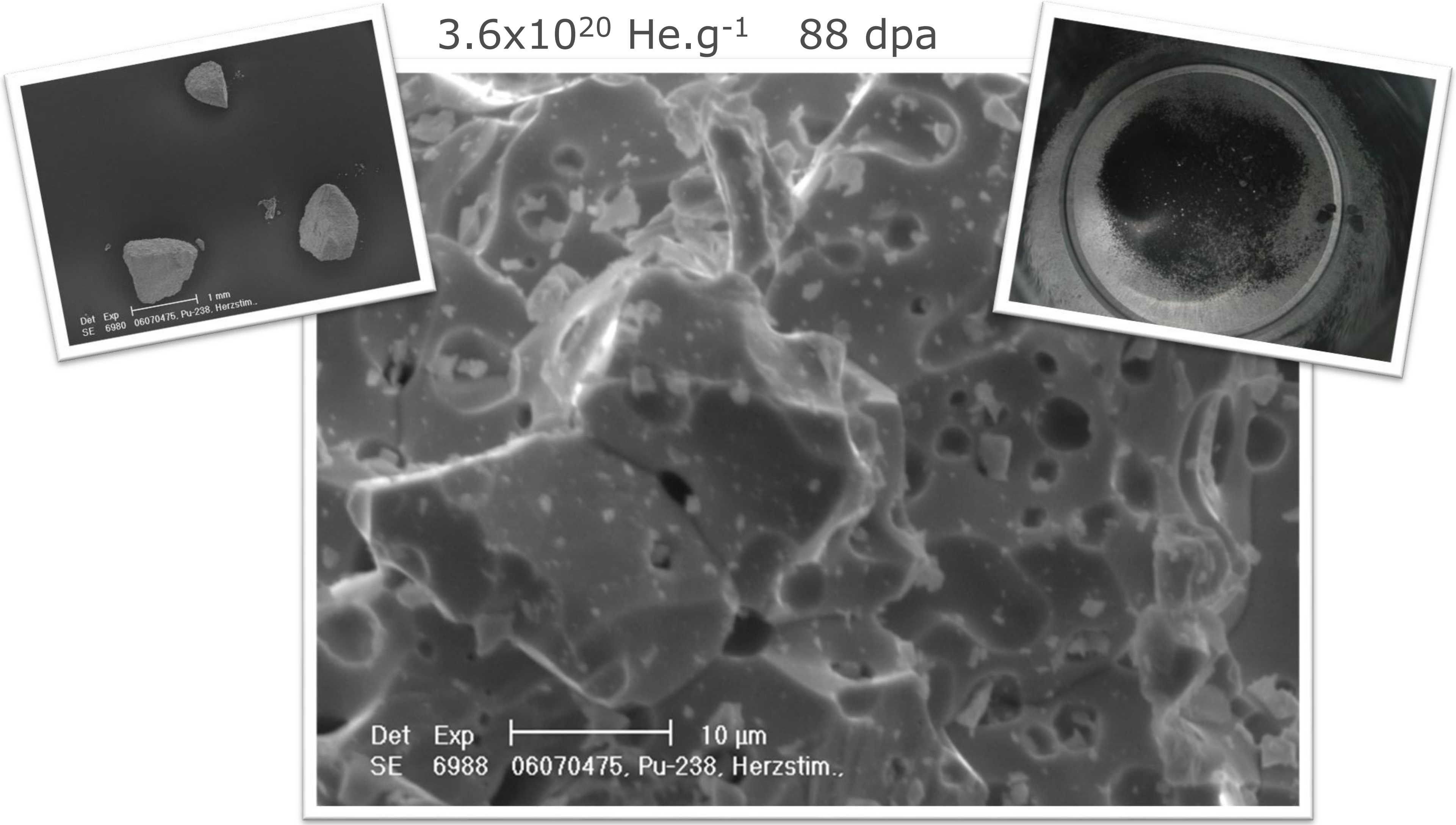




Illustrations d'un stimulateur cardiaque

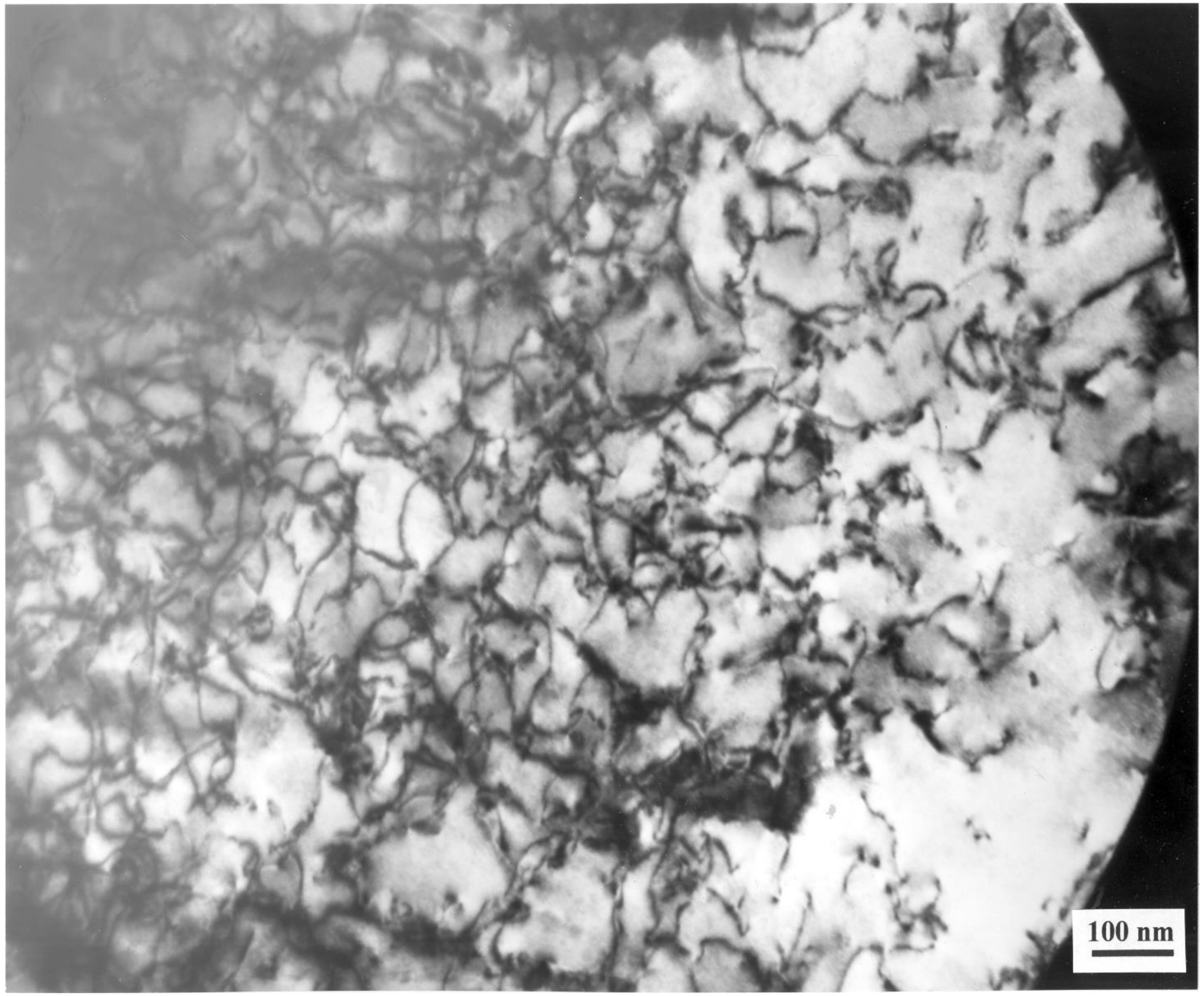
SEM of $^{238}PuO_2$

3.6×10^{20} $He.g^{-1}$ 88 dpa



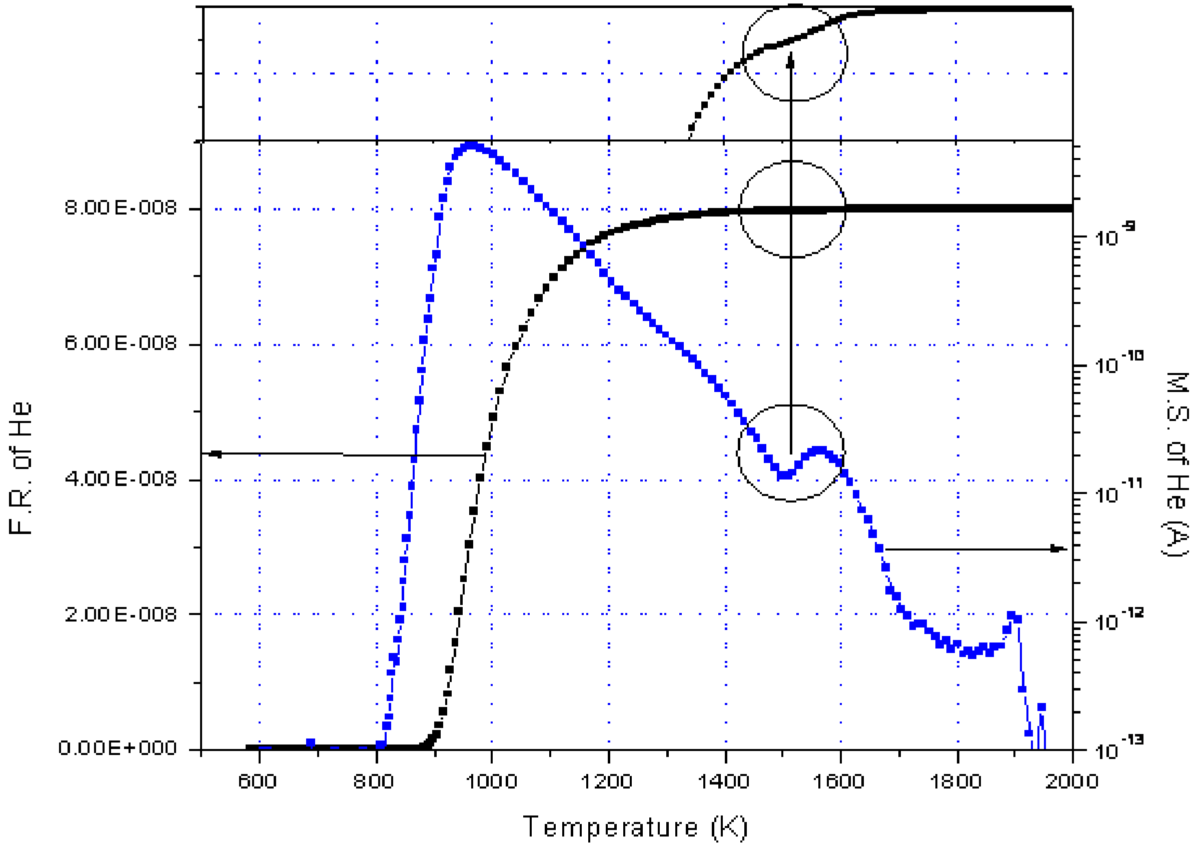
SEM micrograph showing intergranular microcracks.

TEM of $^{238}PuO_2$



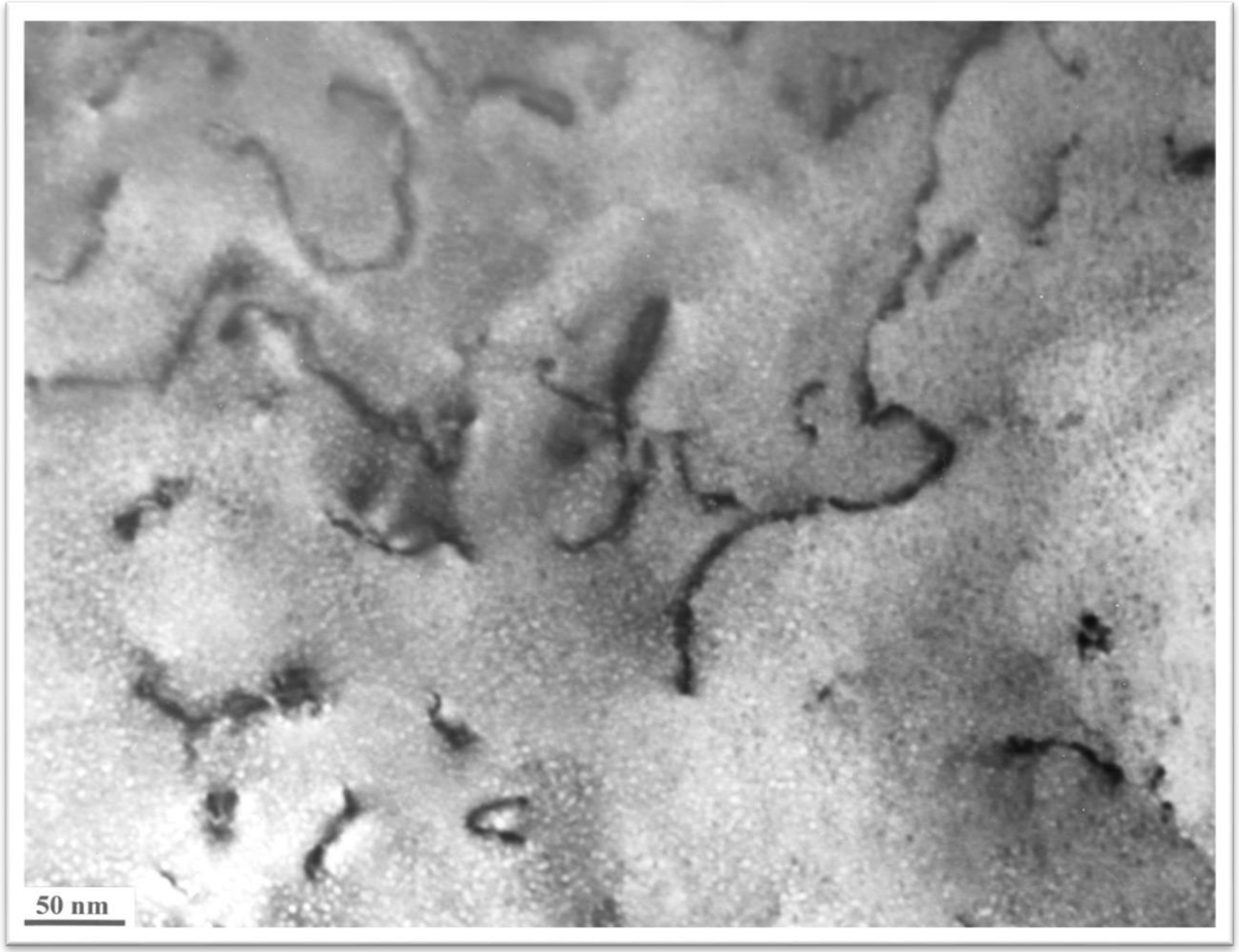
TEM micrograph showing a dislocation network.

He release from $^{238}PuO_2$



- 2/3 of the inventory present
- He-resolution / bubble growth ?

TEM of $^{238}PuO_2$



Bubble swelling 9%
Lattice swelling ~2 %

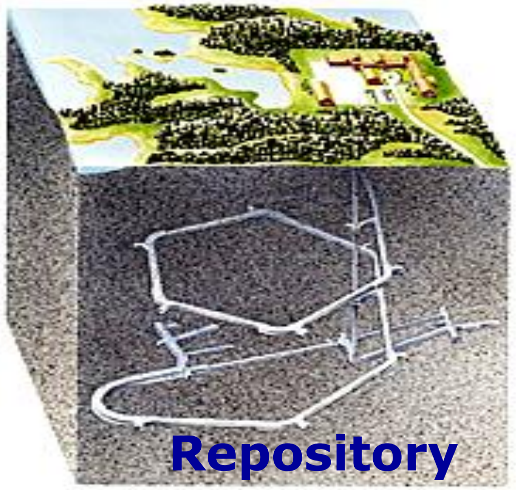
5×10^{23} m^{-3}

Study on alpha-damage - SF

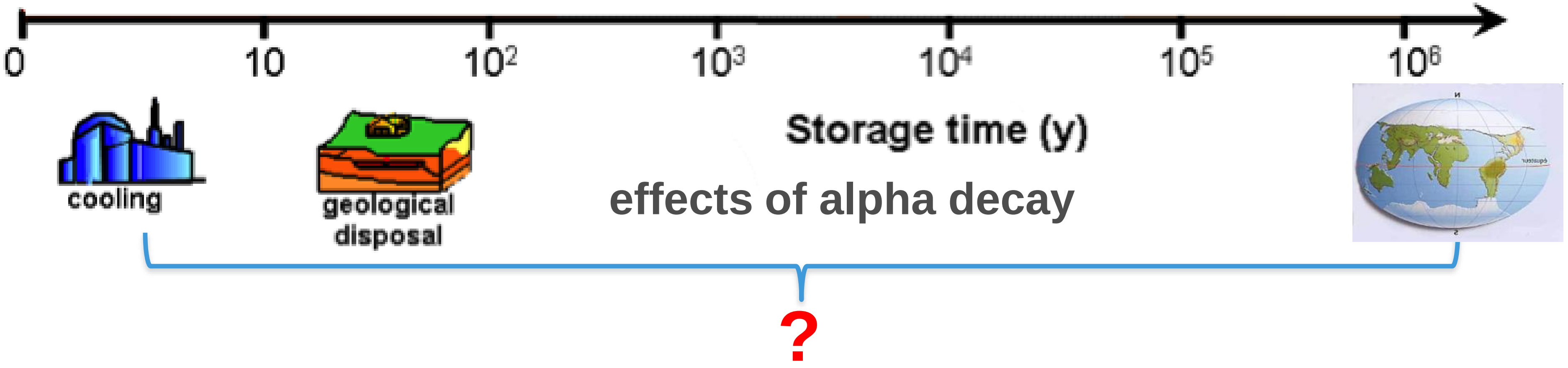
Fresh FR samples – α-doped UO_2

Context

- effect of He/radiation damage accumulation on spent fuel evolution during storage
- Assessment of the contribution of damage prior to PIE's

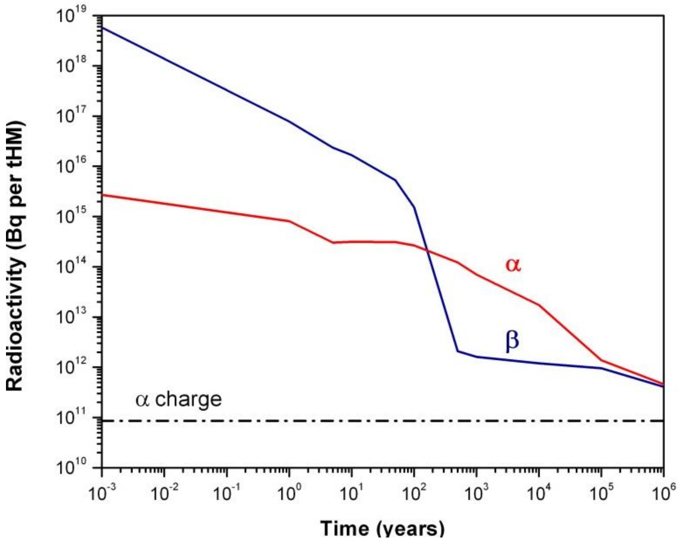


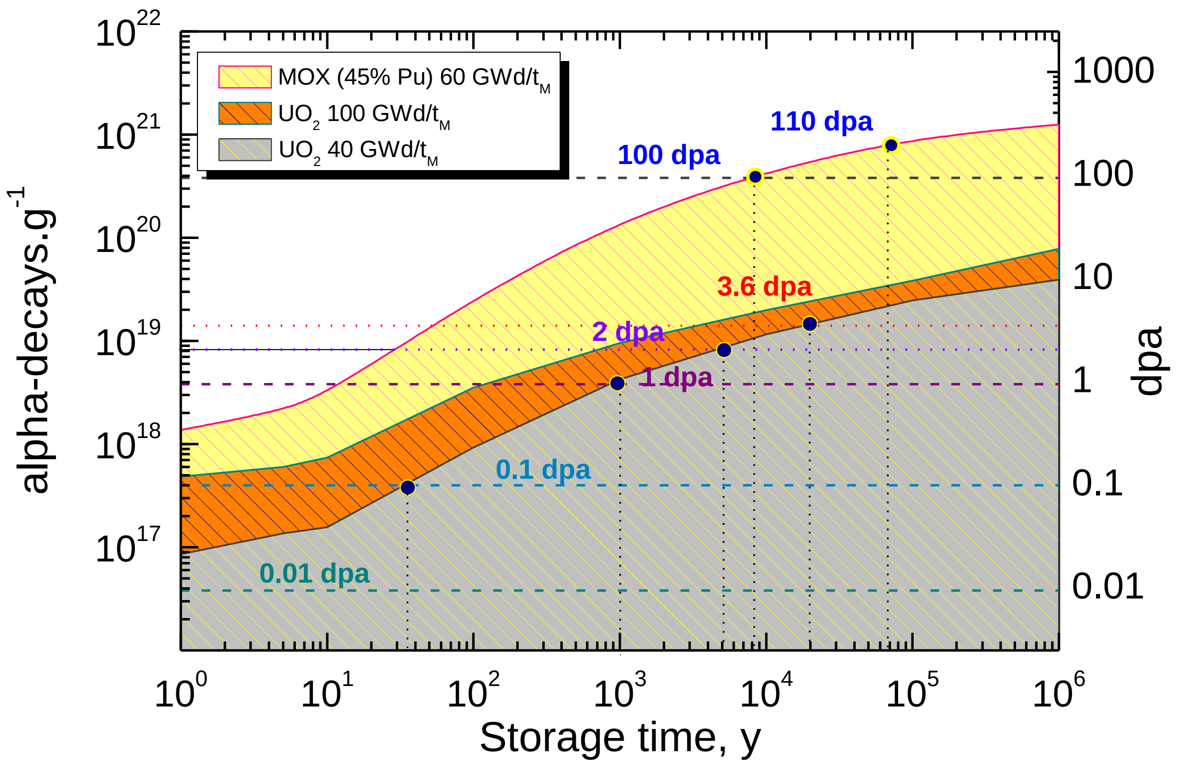
Context



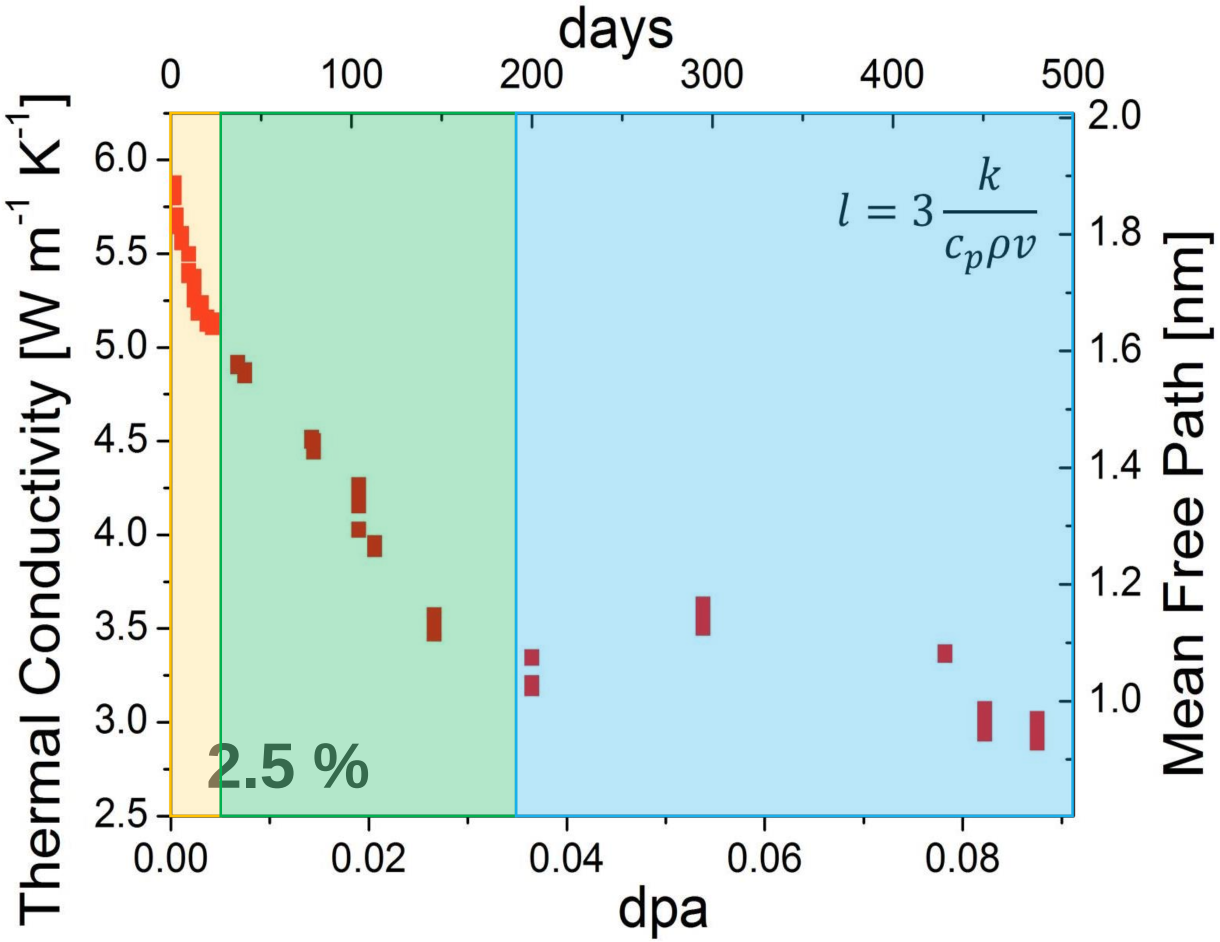
effects of alpha decay

?





Periodic characterization – LAF



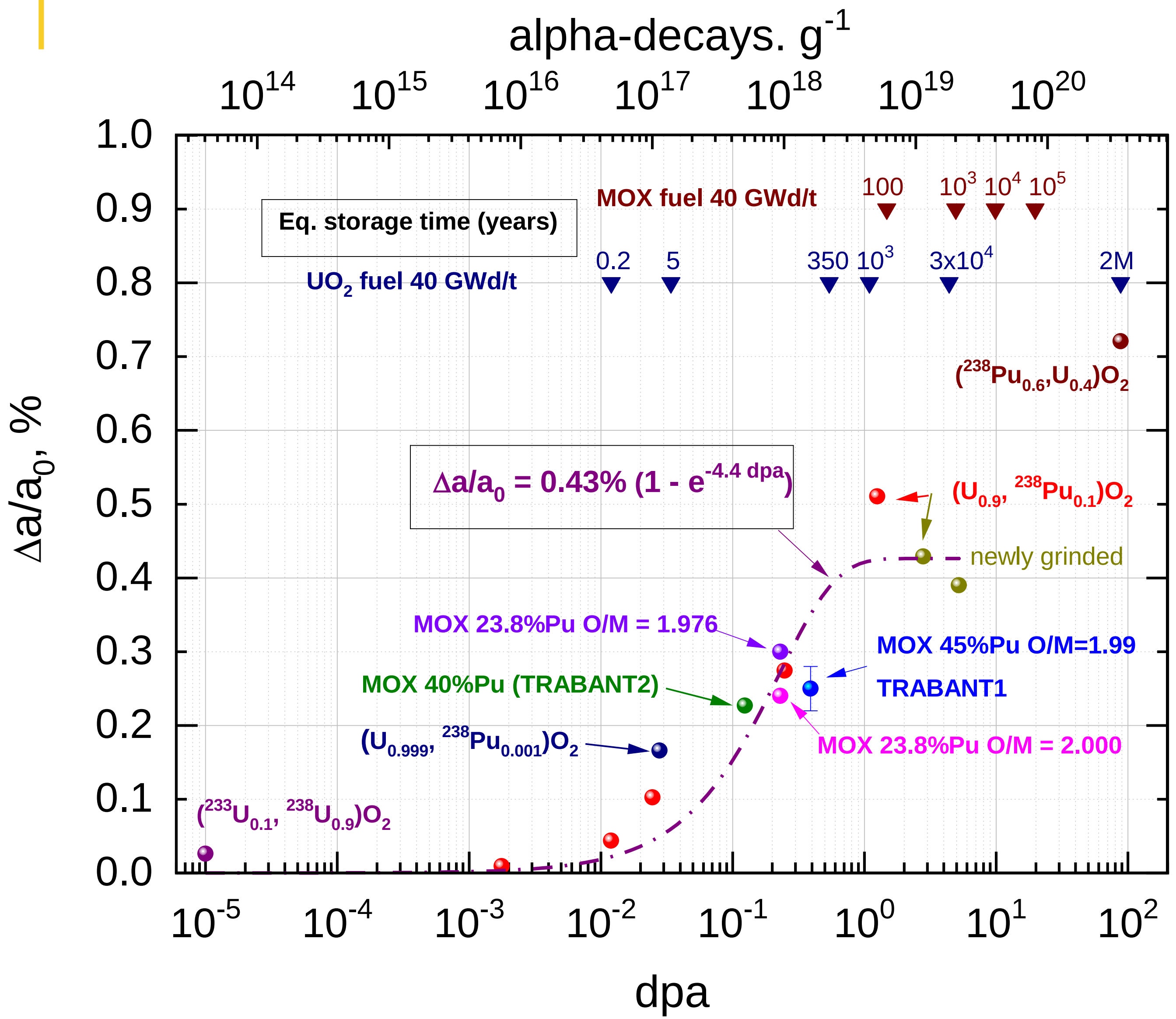
Thermal conductivity decrease:

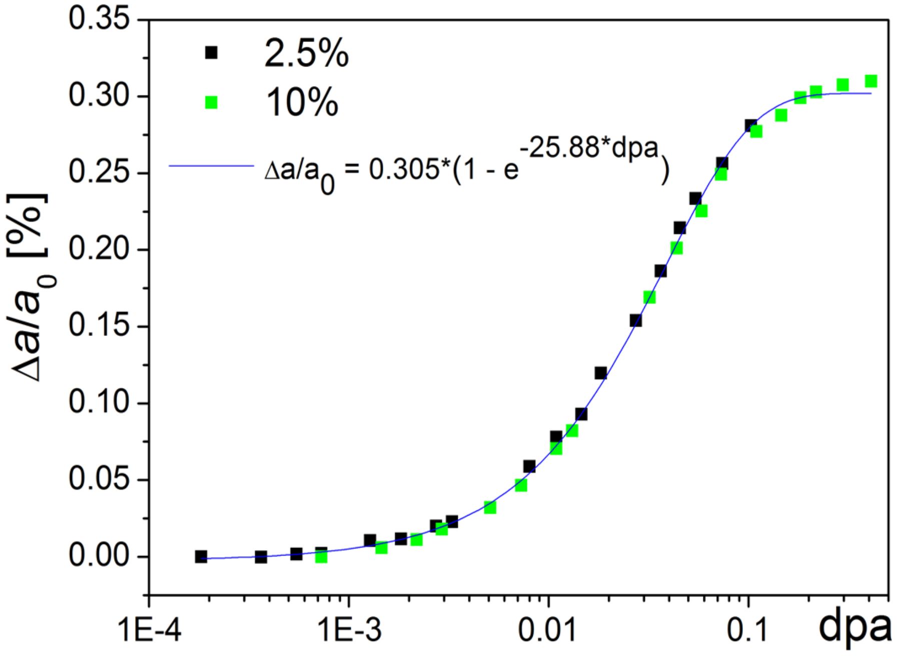
1. Abrupt up to **0.005 dpa** (15 %)
2. Steep up to **0.035 dpa** (up to **40 %**)
3. Plateau after 0.03 dpa

Heat conduction by **phonon transport**

Heavily affected by **point defects**

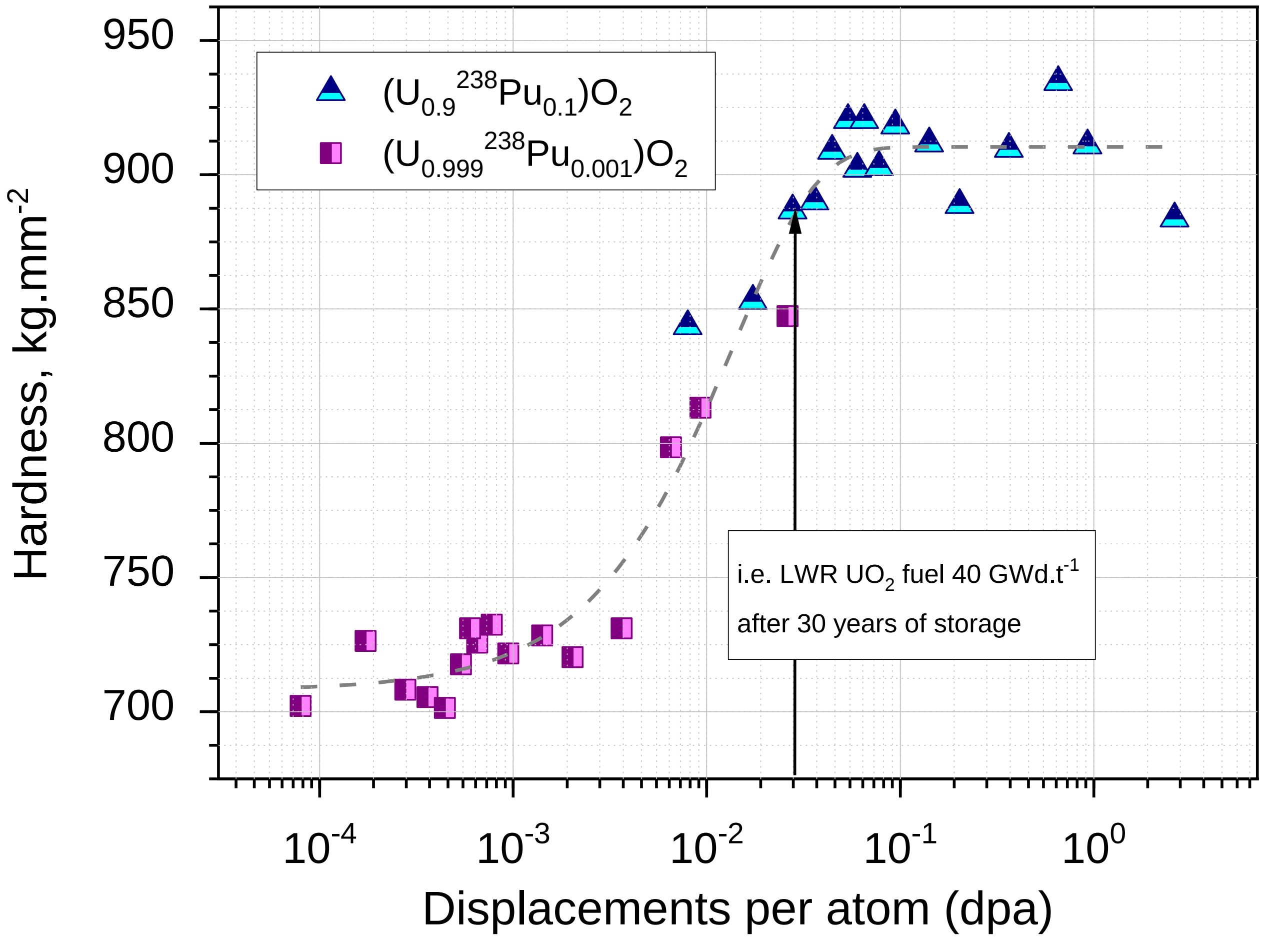
Periodic characterization – XRD





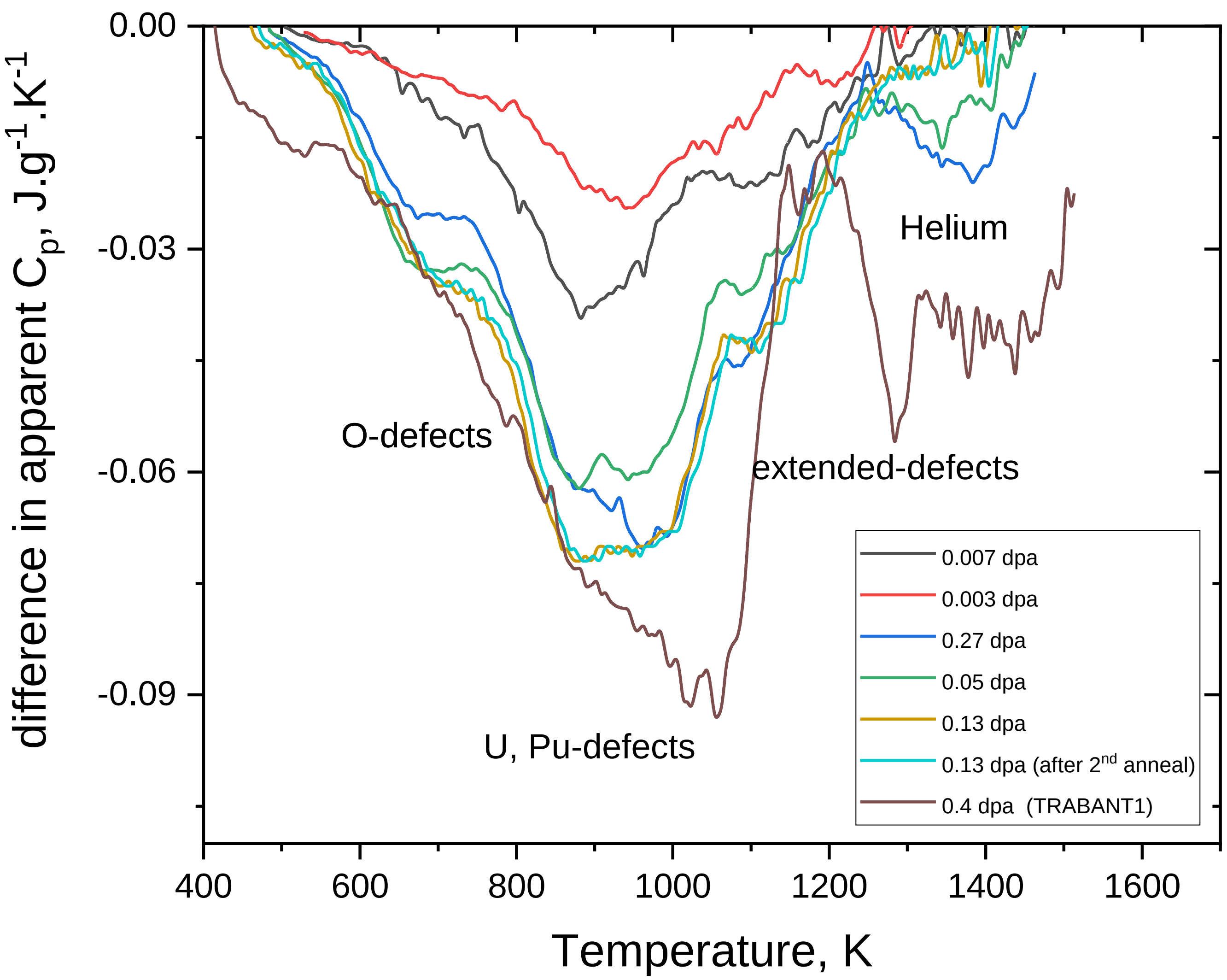
European Commission

Periodic characterization – Vickers Hardness



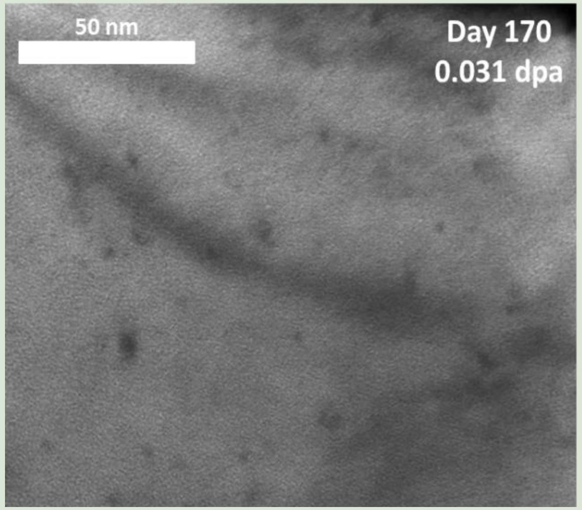
i.e. LWR UO_2 fuel 40 GWd.t^{-1} after 30 years of storage

C_p from alpha-damaged samples



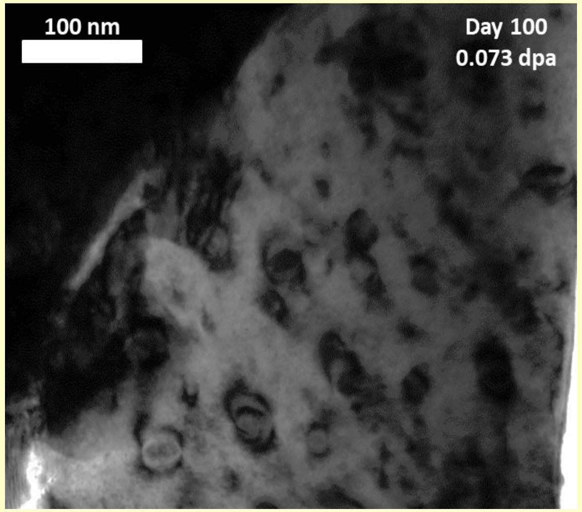
Periodic characterization – TEM

2.5 % – 0.031 dpa



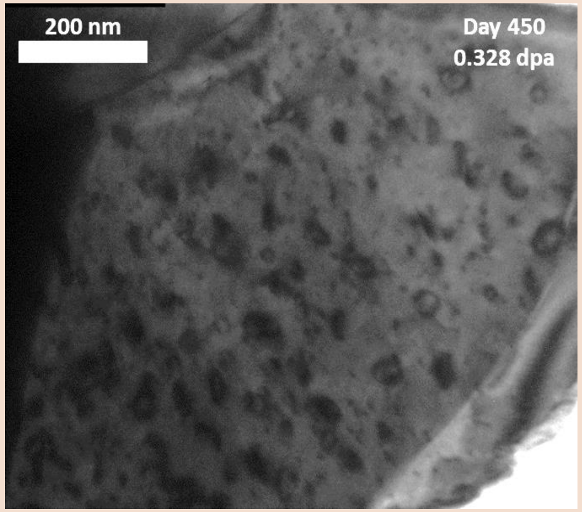
Onset of extended defects population

10 % – 0.073 dpa

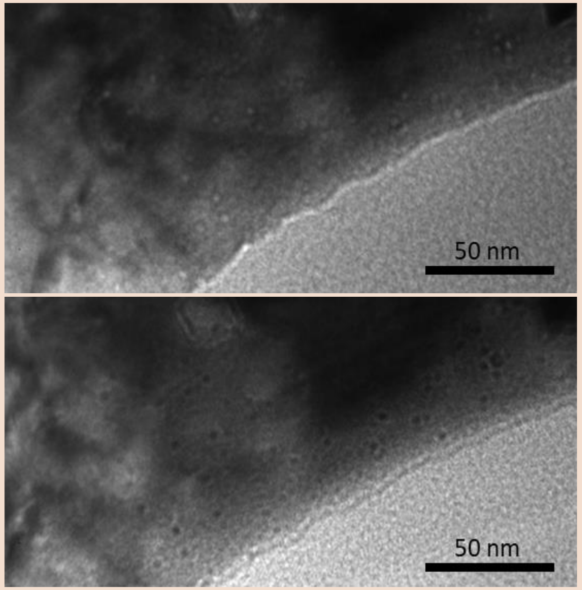


Dislocation loops

10 % – 0.328 dpa

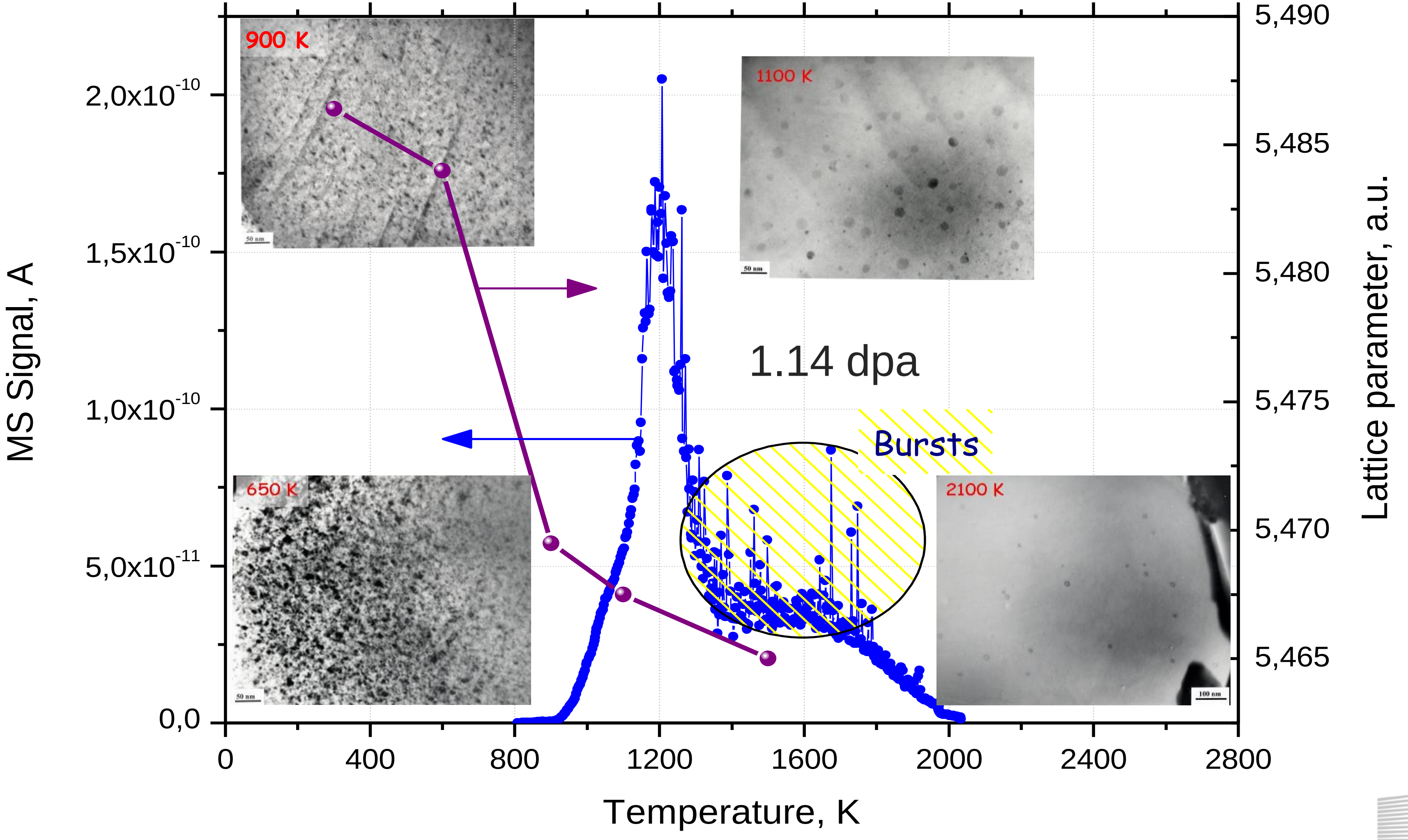


More and larger dislocation loops

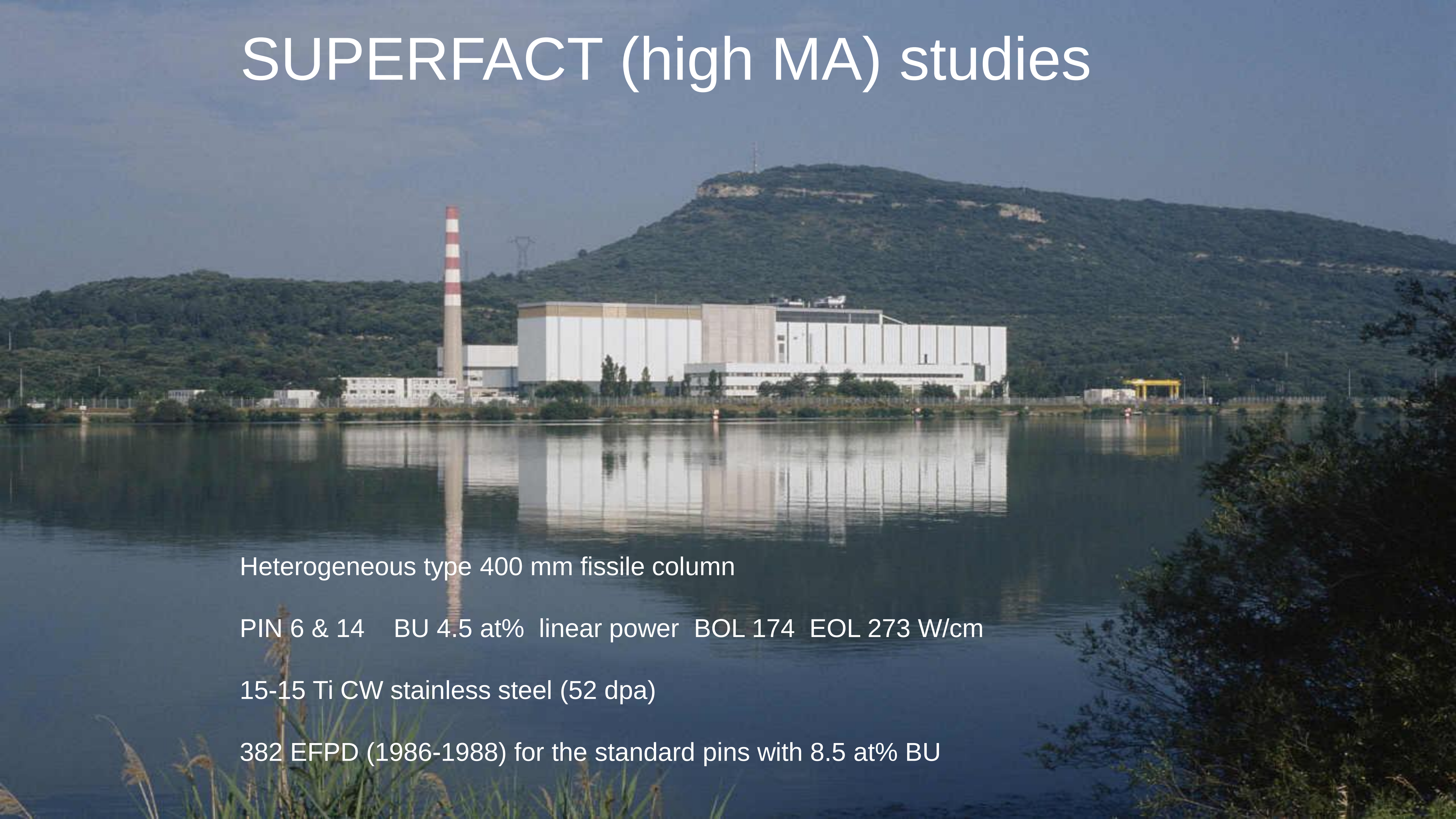


He bubbles

Annealing of alpha-damage + He release



SUPERFACT (high MA) studies



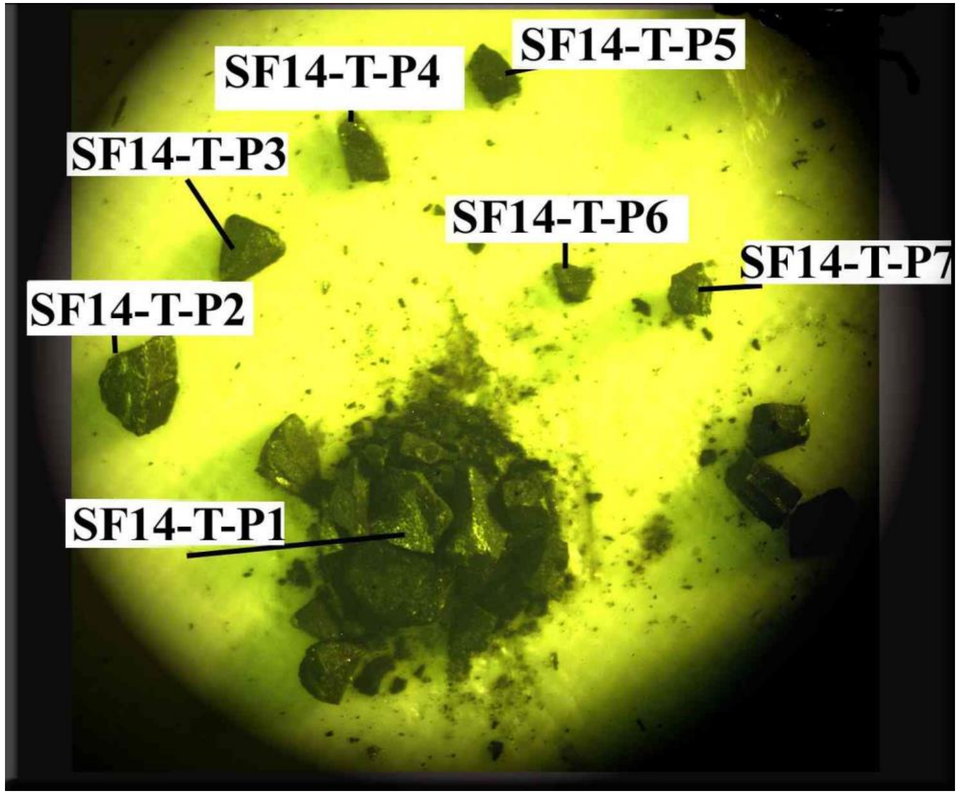
Heterogeneous type 400 mm fissile column

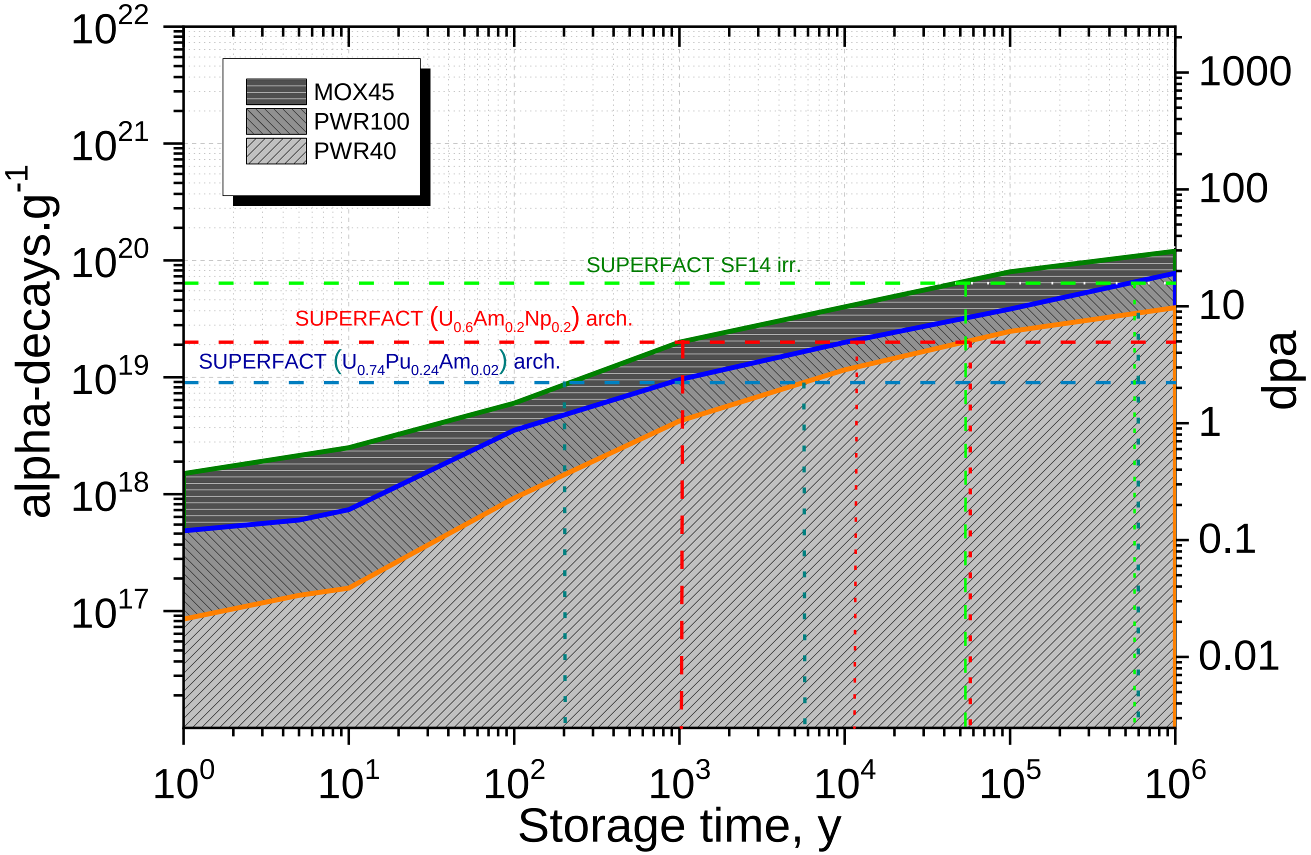
PIN 6 & 14   BU 4.5 at% linear power BOL 174 EOL 273 W/cm

15-15 Ti CW stainless steel (52 dpa)

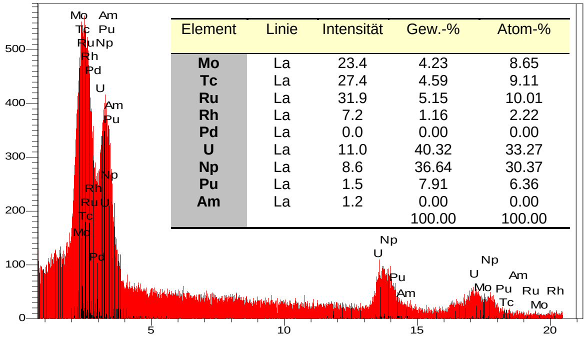
382 EFPD (1986-1988) for the standard pins with 8.5 at% BU

SUPERFACT SF14

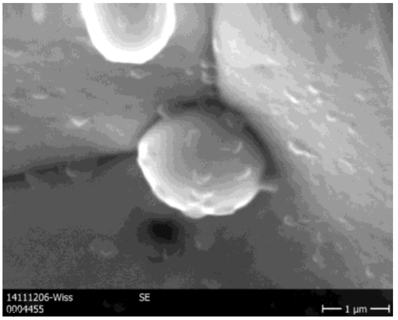




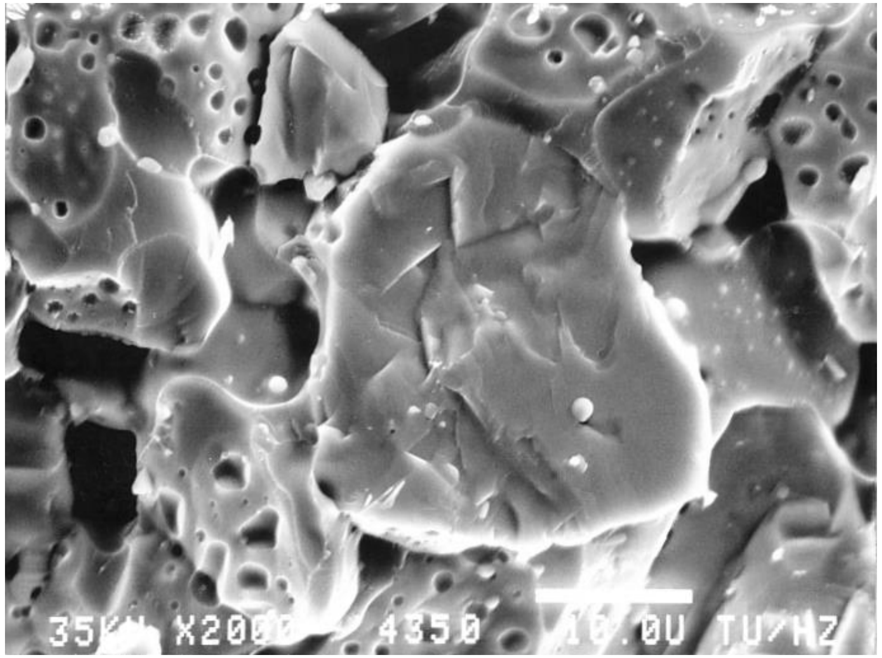
SEM over time – SF14



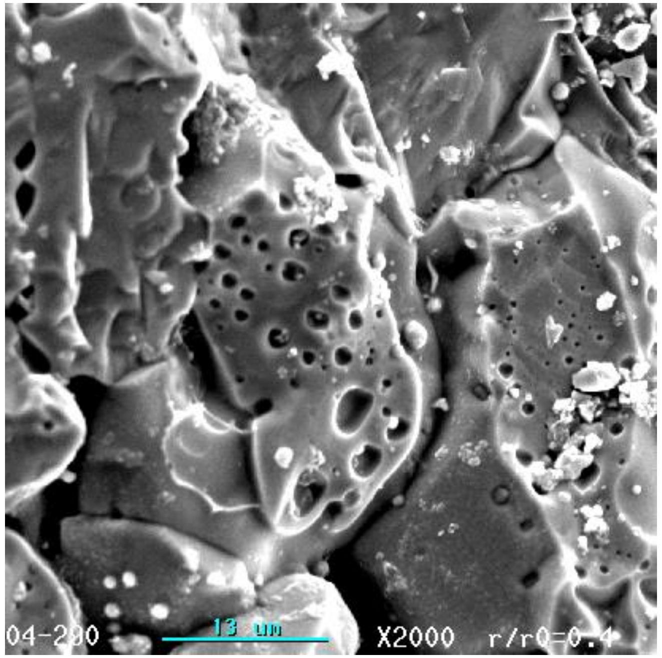
Element	Linie	Intensität	Gew.-%	Atom-%
Mo	La	23.4	4.23	8.65
Tc	La	27.4	4.59	9.11
Ru	La	31.9	5.15	10.01
Rh	La	7.2	1.16	2.22
Pd	La	0.0	0.00	0.00
U	La	11.0	40.32	33.27
Np	La	8.6	36.64	30.37
Pu	La	1.5	7.91	6.36
Am	La	1.2	0.00	0.00
			100.00	100.00



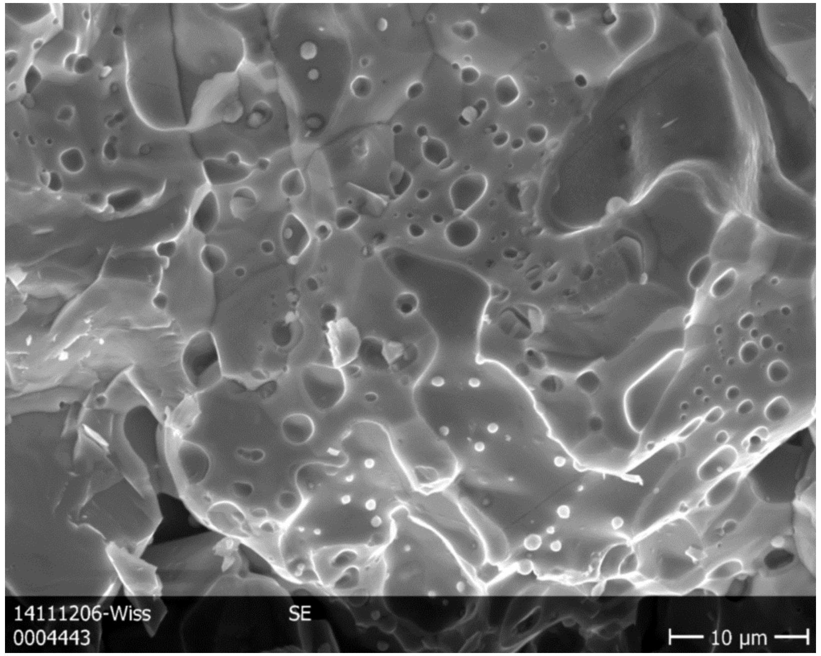
1991



2003

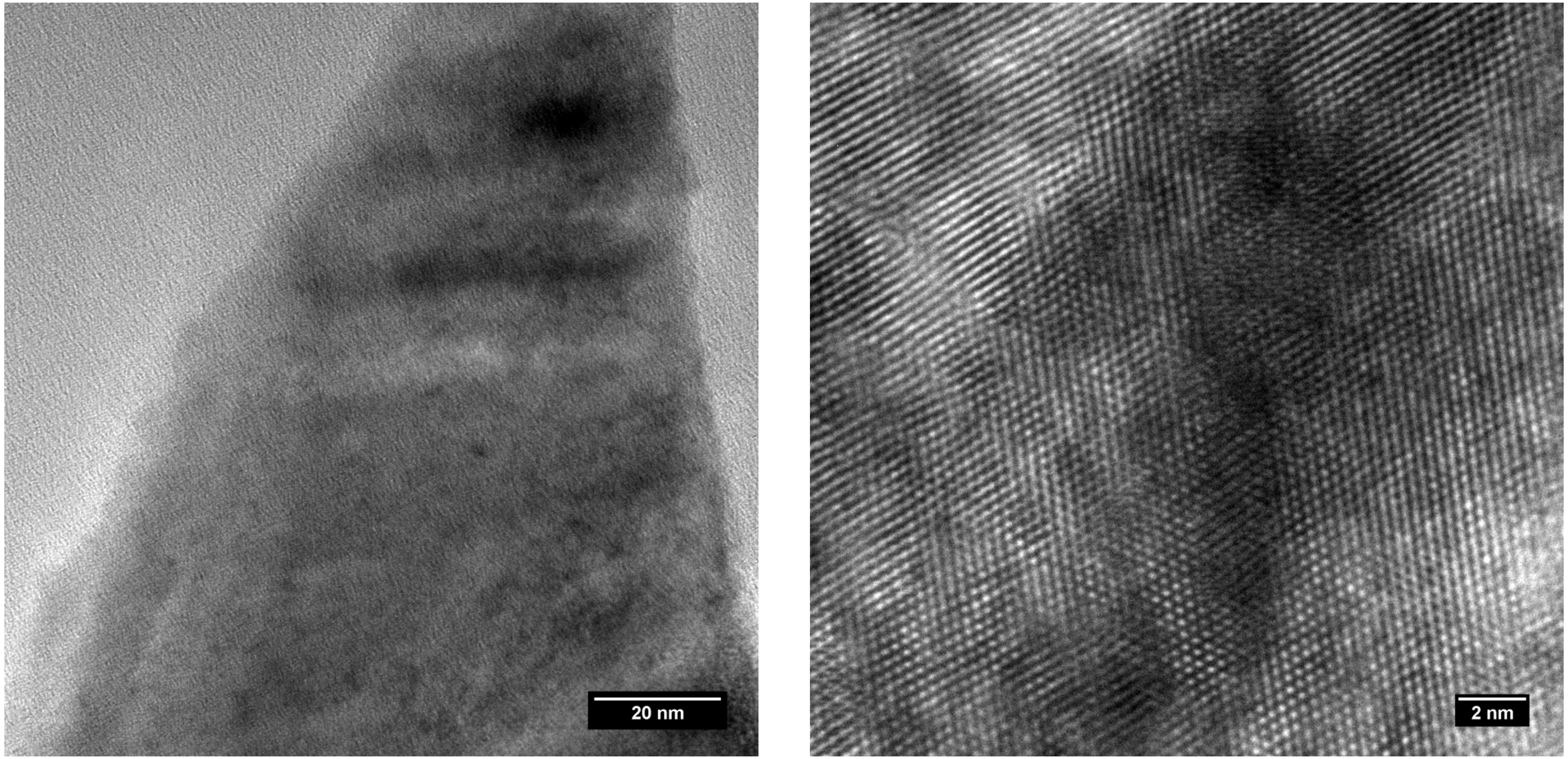


2014



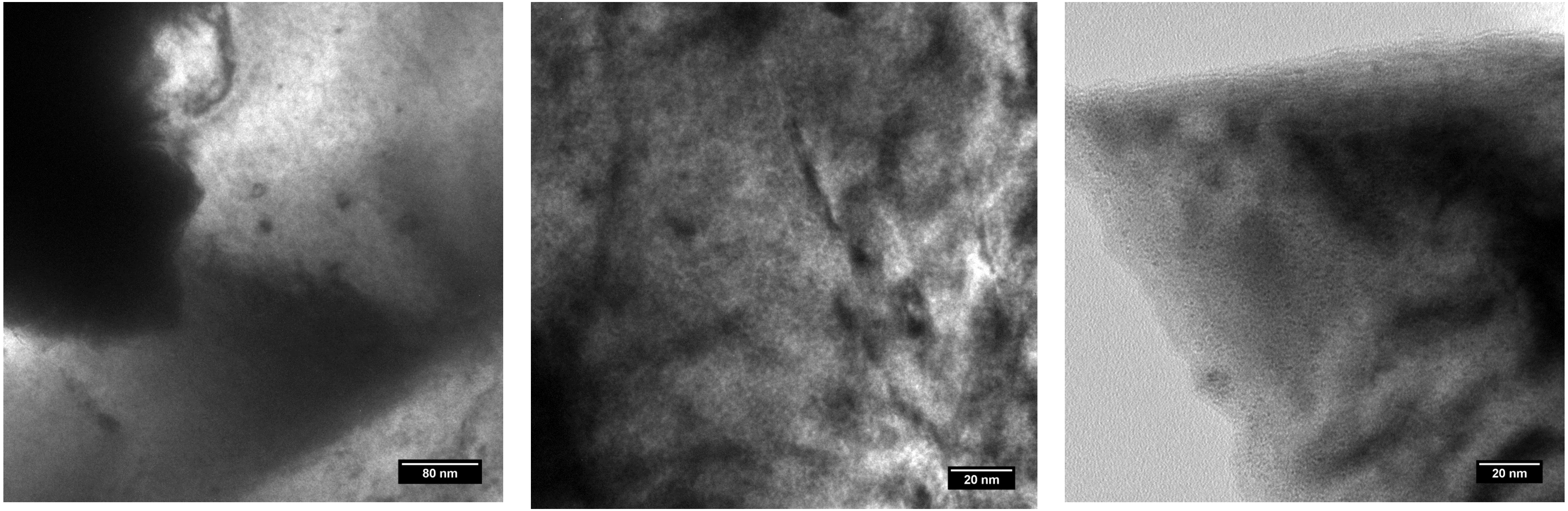
No observable degradation, i.e. GB opening

SUPERFACT Archive



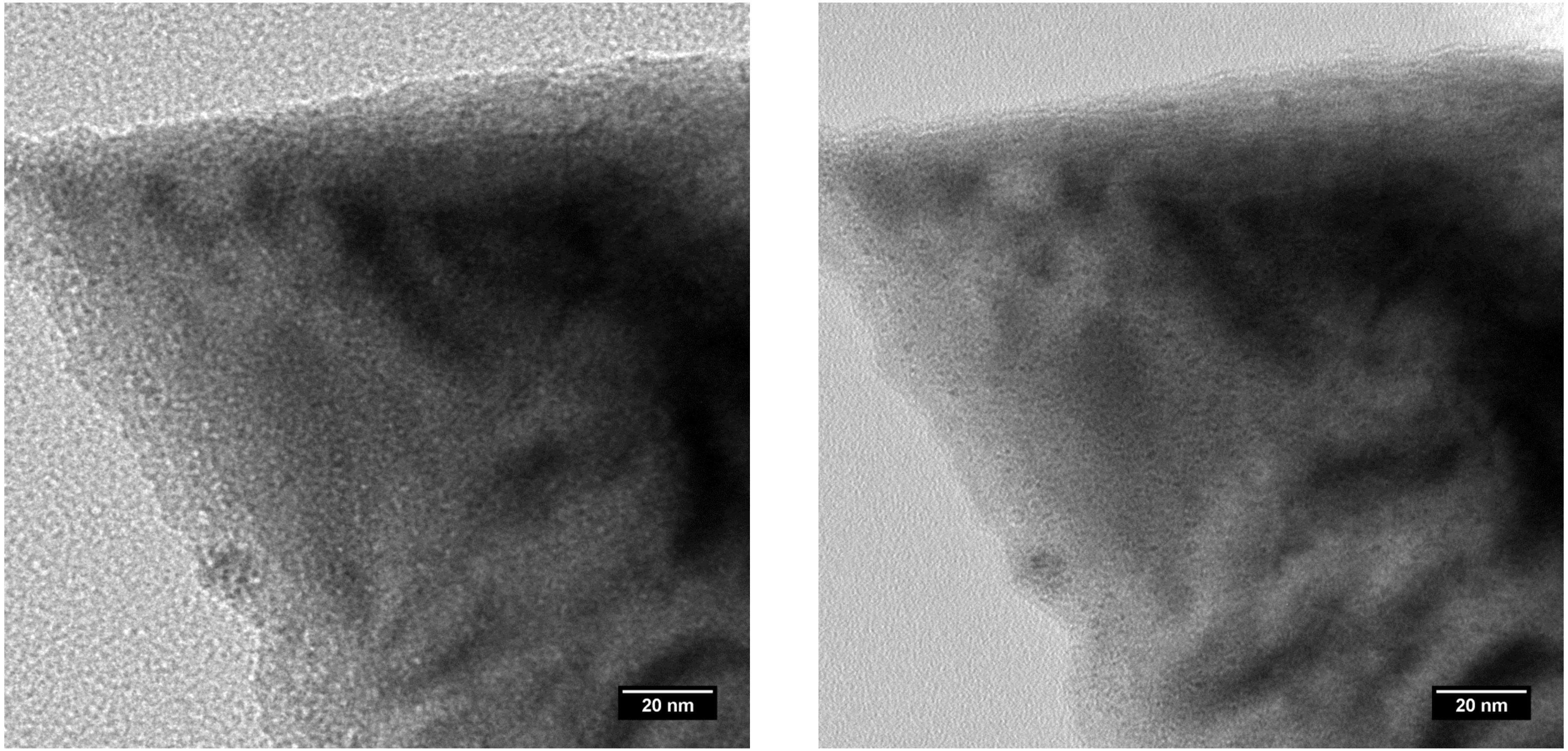
5 dpa $2x10^{19}$ α.g^{-1}

SUPERFACT Irr. SF14-T-P1 TEM



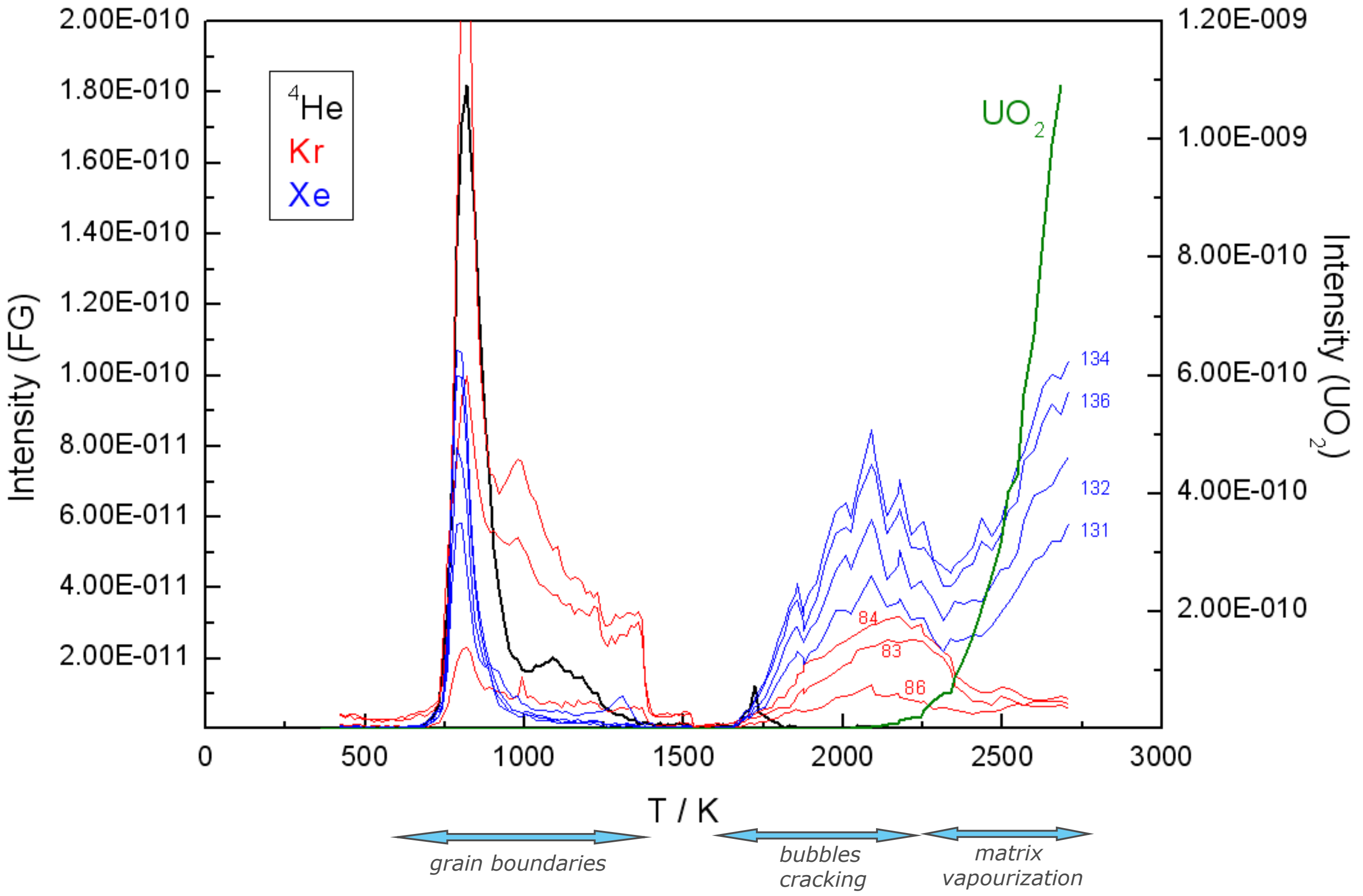
Dislocation loops and lines

SUPERFACT Irr. SF14-T-P1 TEM



Sub-nanometric bubbles

SUPERFACT Irr. SF14-P6 gas-release



PIN6 in-pile release: 61%

Summary SUPERFACT high MA (het.)

- High swelling (high porosity)
- Little restructuring (lower BU)
- FG release 61%
- High helium content (better gap conductance)
- Different fission yield for FGs determined
- Polygonization
- Little He retention

Takeaway message

- Main properties affected by radiation damage (T, BU, linear power, ..)
- Need of data for modelling (FPC, MD, physical models)
- swelling: 0.3% at 0.02 dpa; 1% at 1 dpa (alpha-doped UO_2 i.e. LWR standard BU 1000 years or High BU 200 years or MOX 100 year)
- thermal properties, hardness: saturation at maximum 0.05 dpa
- microstructure: dislocation loops in SNF and α-doped
- Annealing of alpha-damage not below 500 K
- PIE affected by alpha-damage.
- Radiogenic helium and alpha-damage must be considered in storage conditions.

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Many colleagues at CEA

Thank you

Slide 17,18: all pictures, source: https://en.wikipedia.org/wiki/Radioisotope_thermoelectric_generator

; Slide 21: graph, source: R.C. O’Brien et al. / Journal of Nuclear Materials 377 (2008) 506–521

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