The Abdus Salam International Centre for Theoretical Physics

2359-12

Joint ICTP-IAEA Workshop on Physics of Radiation Effect and its Simulation for Non-Metallic Condensed Matter

13 - 24 August 2012

JRC testing facilities and experimental tools for study of radiation damage

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Commission européenne

JRC testing facilities and experimental tools for study of radiation damage

Thierry Wiss

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JRC testing facilities and experimental tools for study of radiation damage

TMS + MMSNF

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Studies on properties and behaviour of nuclear fuels

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Content

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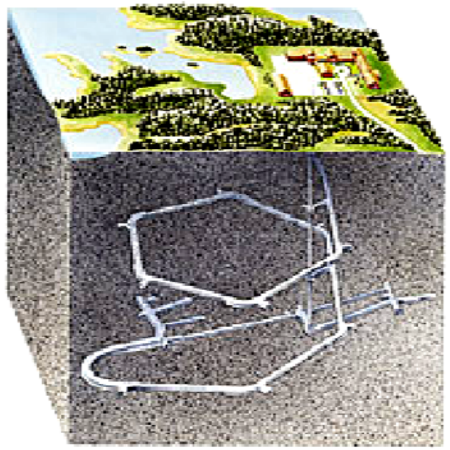
- Forewords
- PIE
- Studies on spent fuel
- Cladding
- Conclusion / perspectives

Repository



Scope

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waste composition/irradiation history

Aging: solid state

radiation damage mechanisms, effects

helium accumulation

property evolution
- bulk
- interface

microstructure evolution

oxidation

recovery (annealing)

High-level wasteforms:

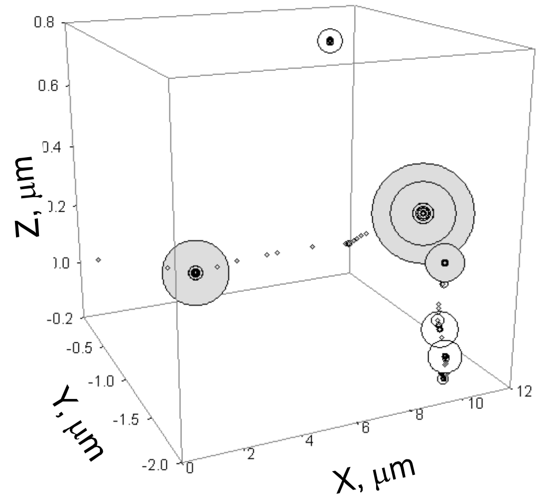
high bu UO_2
MOX
new compounds
(crystalline) ***conditioning matrices***

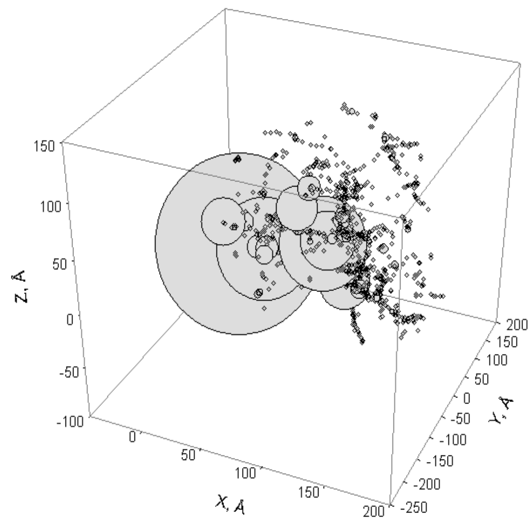
Timeframes:

short-term
interim (<500 y)
geologic periods

long term
mechanical stability

5 MeV alpha-particle





100 keV recoil atom

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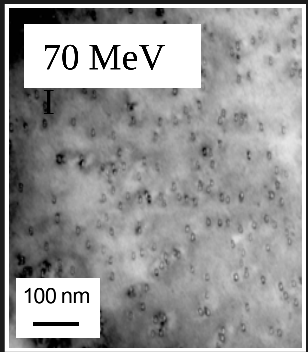
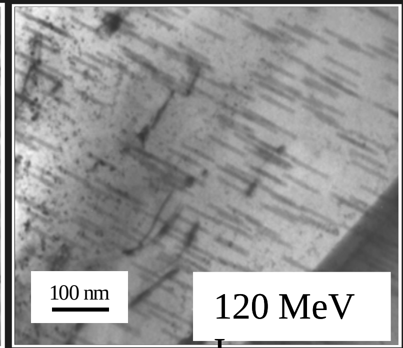
Methodology



CeO

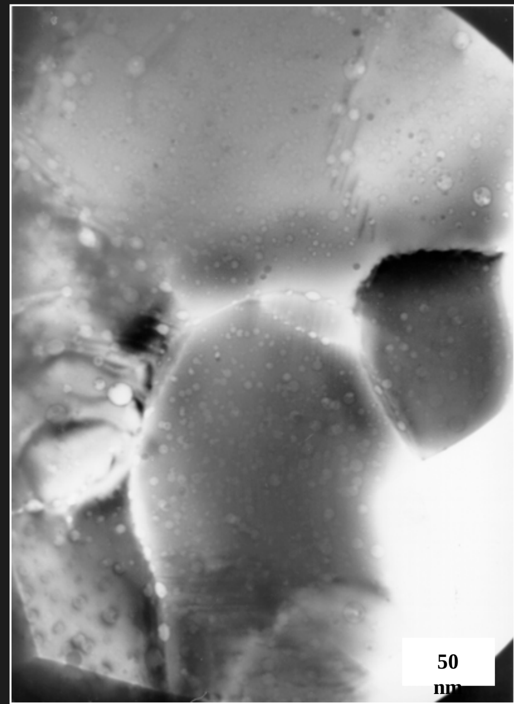


Nd_2Zr_2O

Single effect studies: irradiation with selected ions at given energies

$(U,^{238}Pu)O_2$

Doping with alpha-emitters for homogeneous damage and helium distribution



UO_2 - 75 GWd/t_U

Concomittant effect of different damage sources

Damage in UO_2

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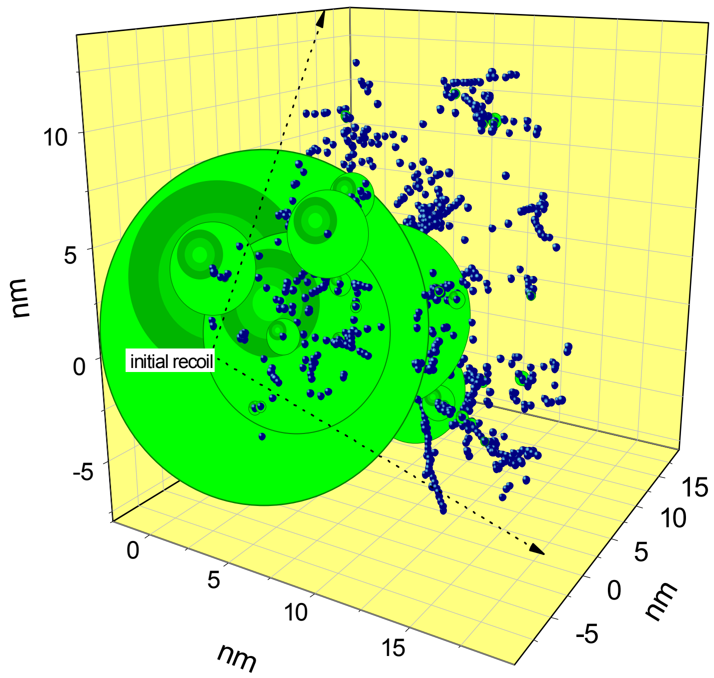
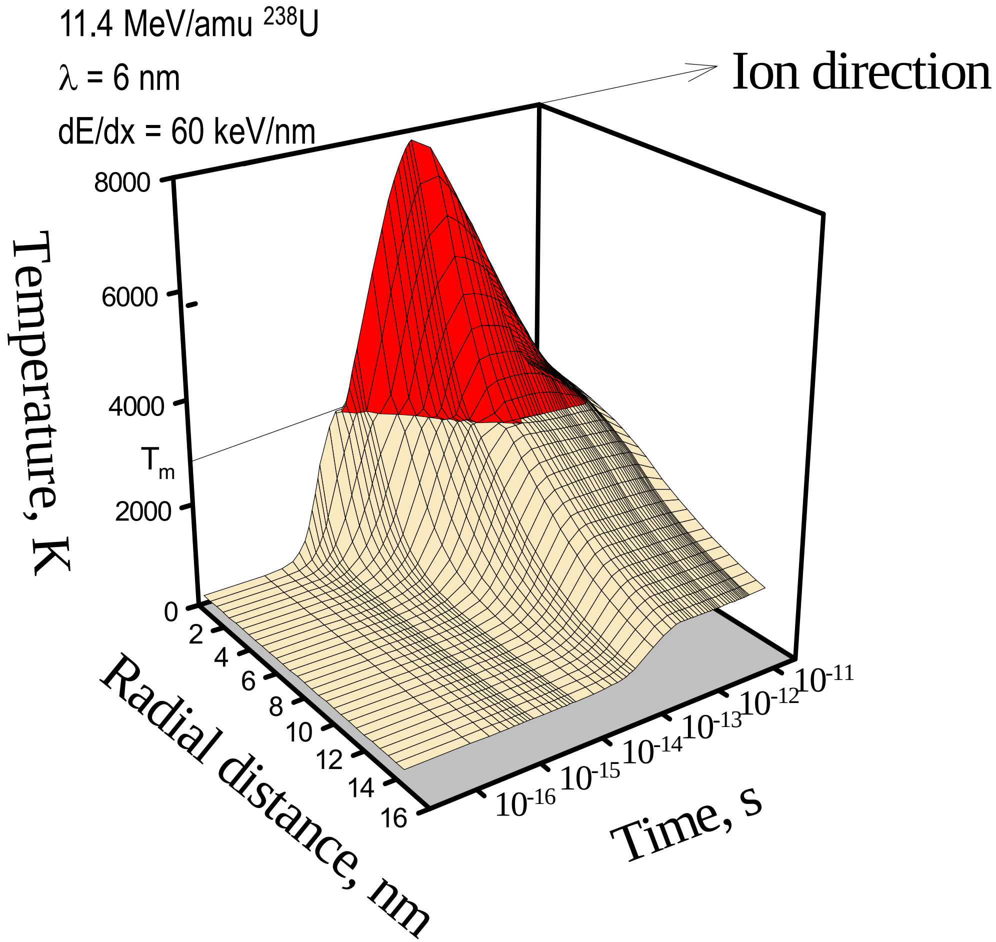
Cascade from a recoil (SRIM2000)



Thermal spike



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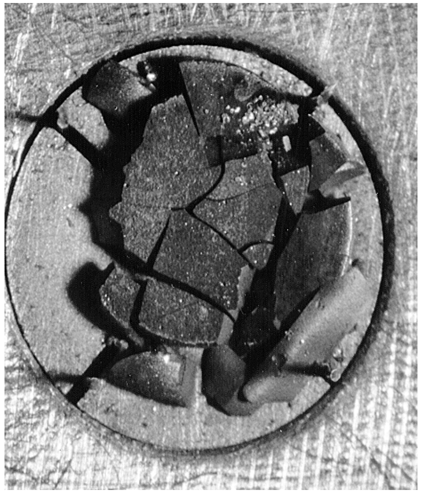
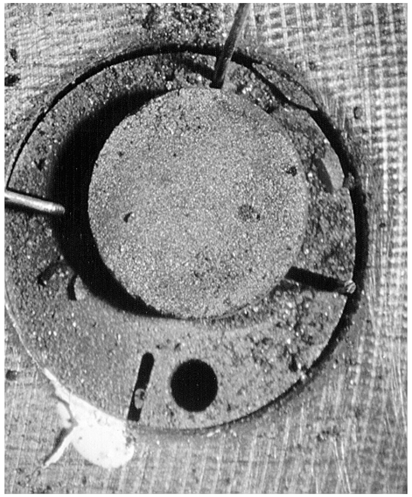
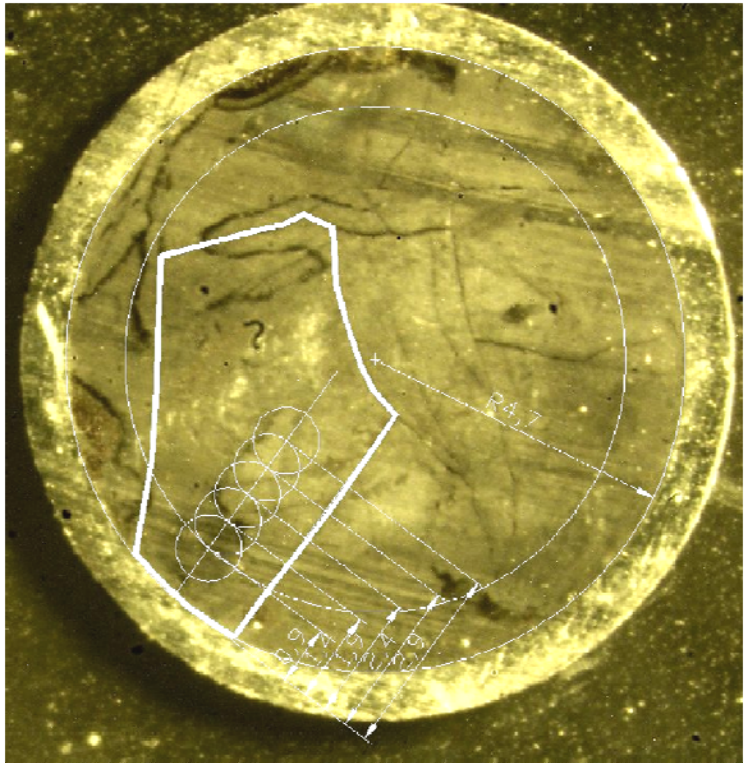
How to measure properties as a function of burnup, T_{irr}?

The combination of commercial fuel investigations and tailor-made irradiations provides optimum results.



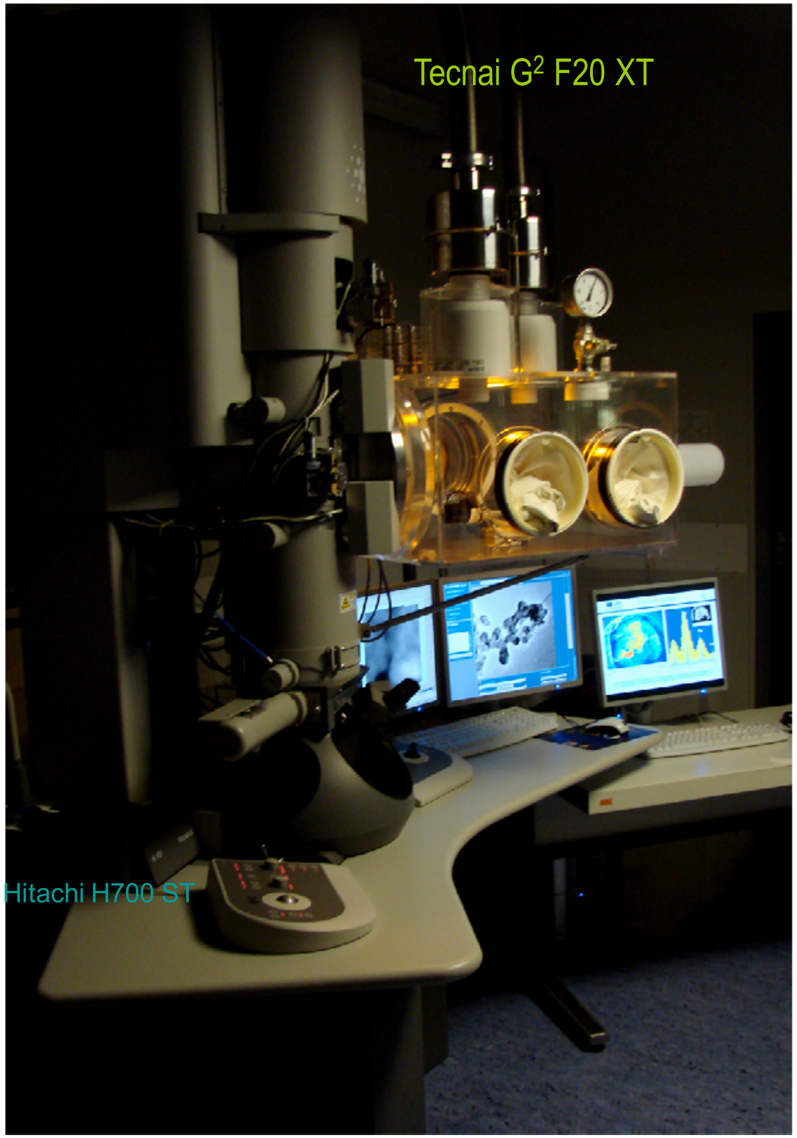


UO_2 discs irradiated to a flat burnup and T_{irr} radial profile. (HBRP project)



High burnup LWR UO_2 radial averaging of measured quantities; (white circles indicate spots for Laserflash measurements)

Electron microscopy



SEM Philips XL40

SE, BSE, EDX

Irradiated fuel can be handled.

TEM characteristics

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TEM point resolution (nm) 0.25

TEM line resolution (nm) 0.102

Information limit (nm) 0.14

TEM magnification range 22 x - 930

STEM HAADF resolution (nm) 0.23 (0.18)

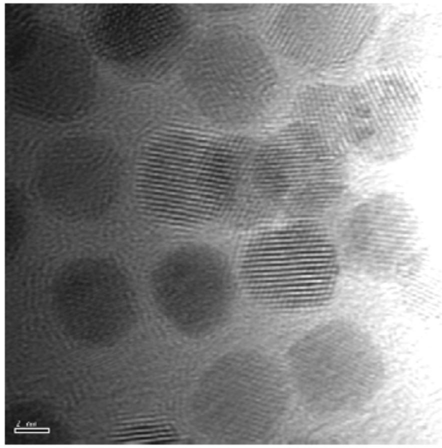
STEM magnification range 150 x - 230 Mx

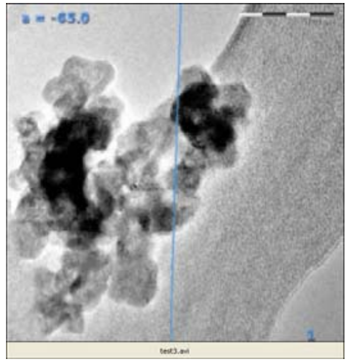
Maximum tilt angle with tomography holder ±70°

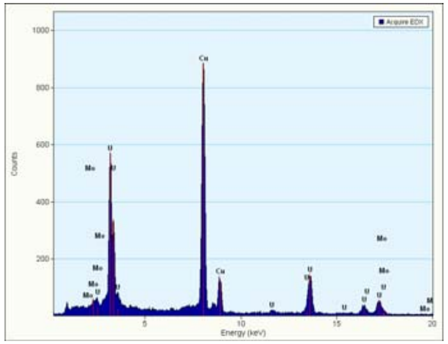
EDS energy resolution 134.6 eV

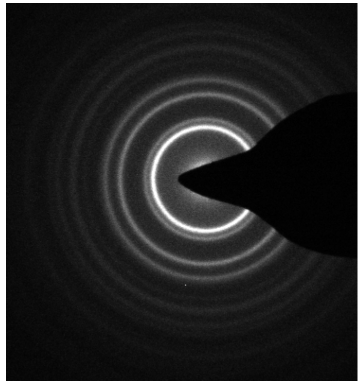
Spot drift 1 nm.min-1

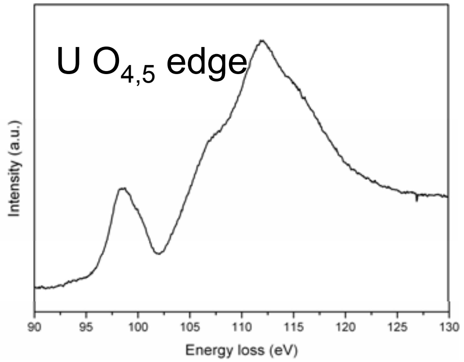
Resolution EELS (ZLP) 0.6 eV

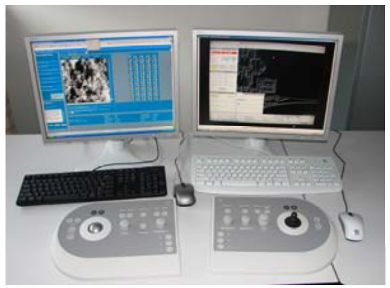


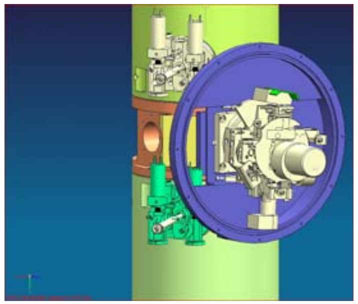


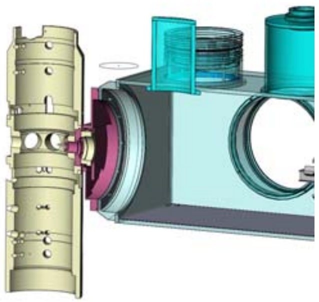












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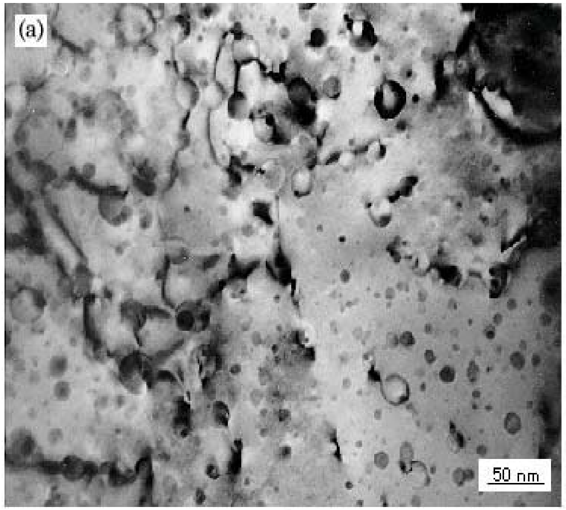
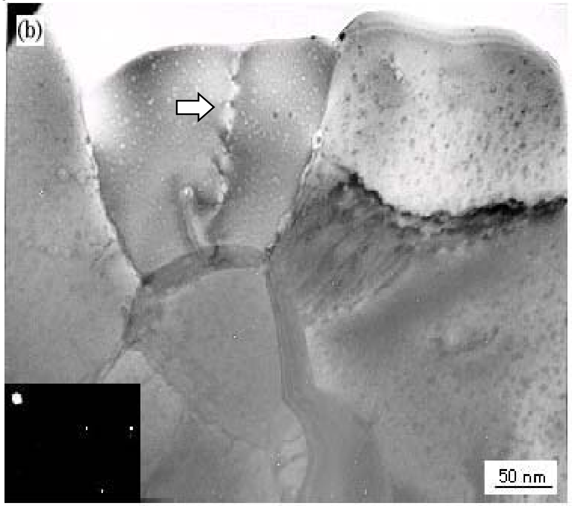
Microstructure of high burnup fuel (TEM)

LWR T_{irr} = 450 C



UO_2 55 GWd/t



UO_2 82 GWd/t

Studies on high burnup fuel properties are continuing along several lines:

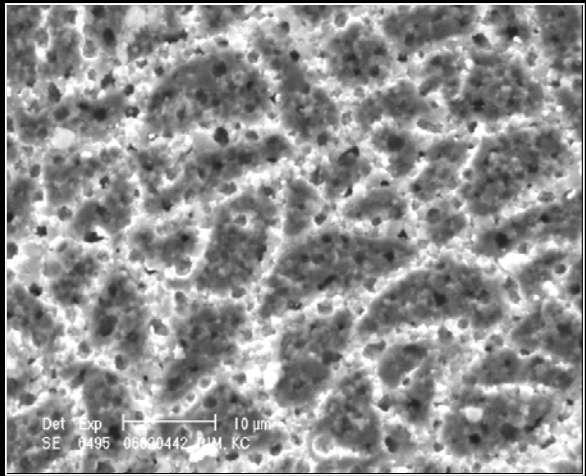
- fundamental studies on (radiation damage) evolution at very high dose/burnup.

- microstructure examination of high burnup fuel to 'map' relevant features and quantify distribution of gas bubbles, extended defects

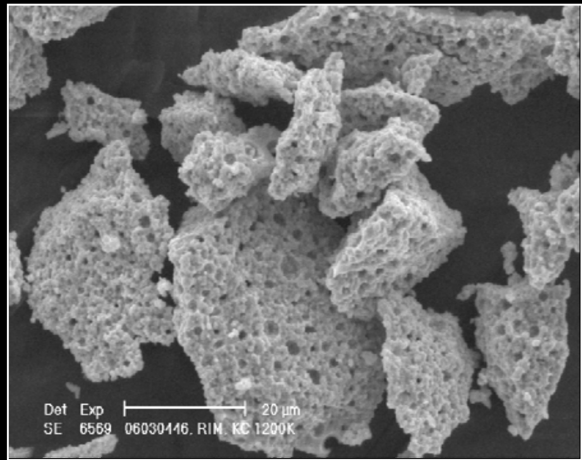
- effects of accumulated strain energy on the fuel restructuring process

models will be implemented in codes.

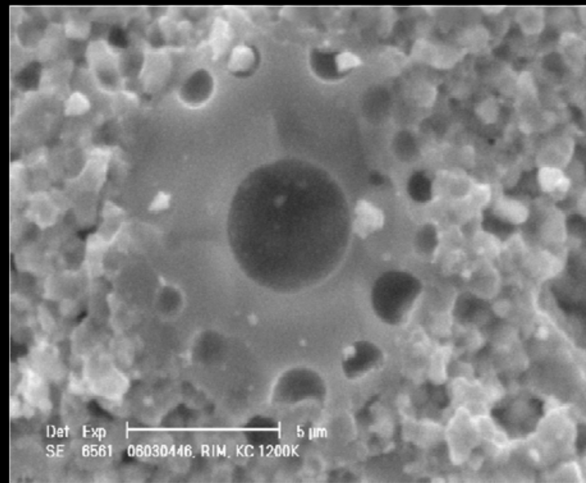
SEM of a 200 GWd / t_U fuel sample



Memory effect after HBS formation



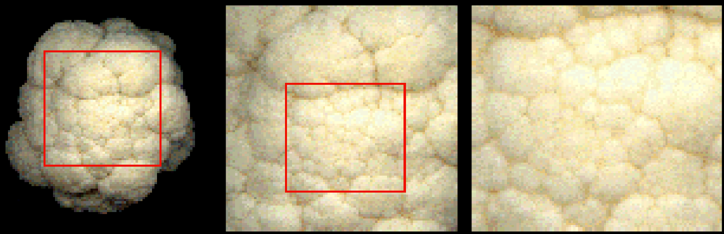
Break of the structure during annealing (1200 K)



Formation of „ultimate“ pores at very high burnup

Fission product release and microstructure changes during laboratory annealing of a very high burn-up fuel specimen,
J.-P. Hiernaut *, T. Wiss, J.-Y. Colle, H. Thiele, C.T. Walker, W. Goll, R.J.M. Konings, J.Nucl. Mater., in press

High Burnup Structure



Fractal dimension

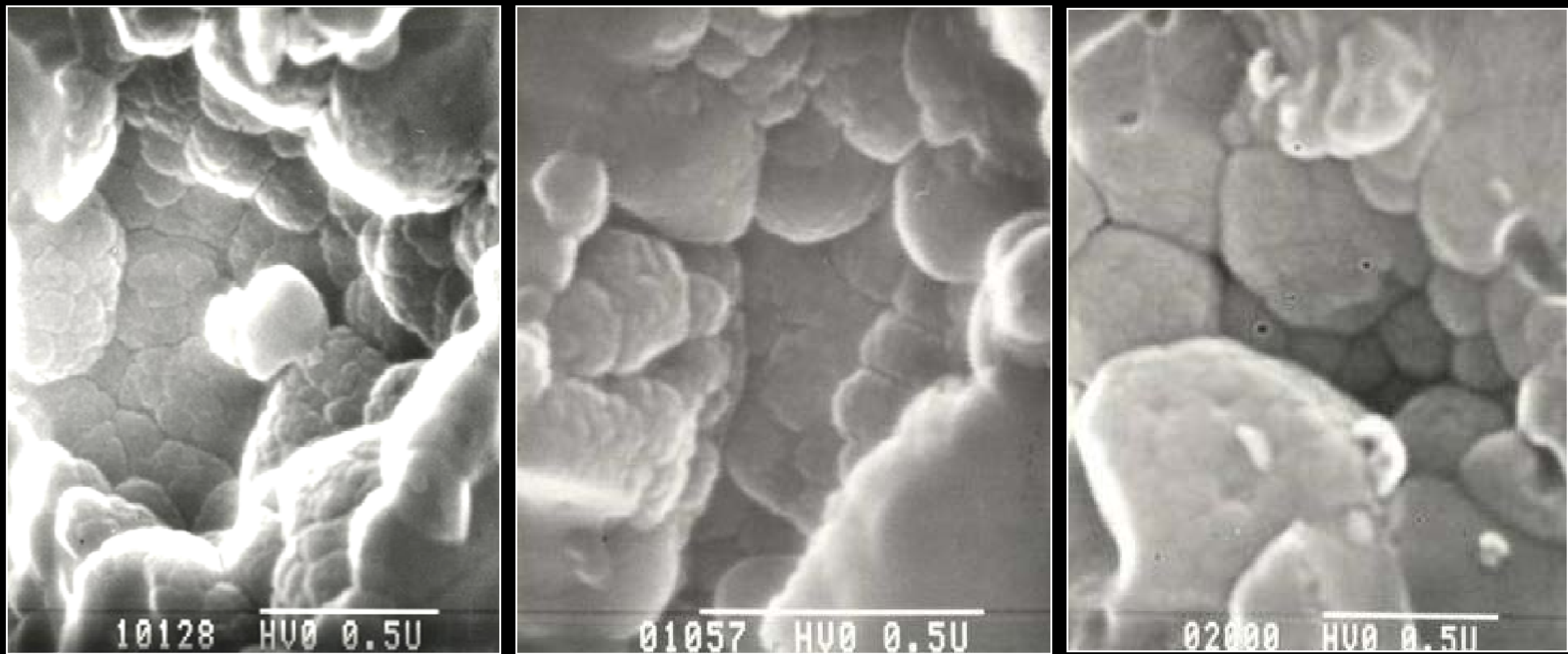
Log p / log q

p: nb of fractals

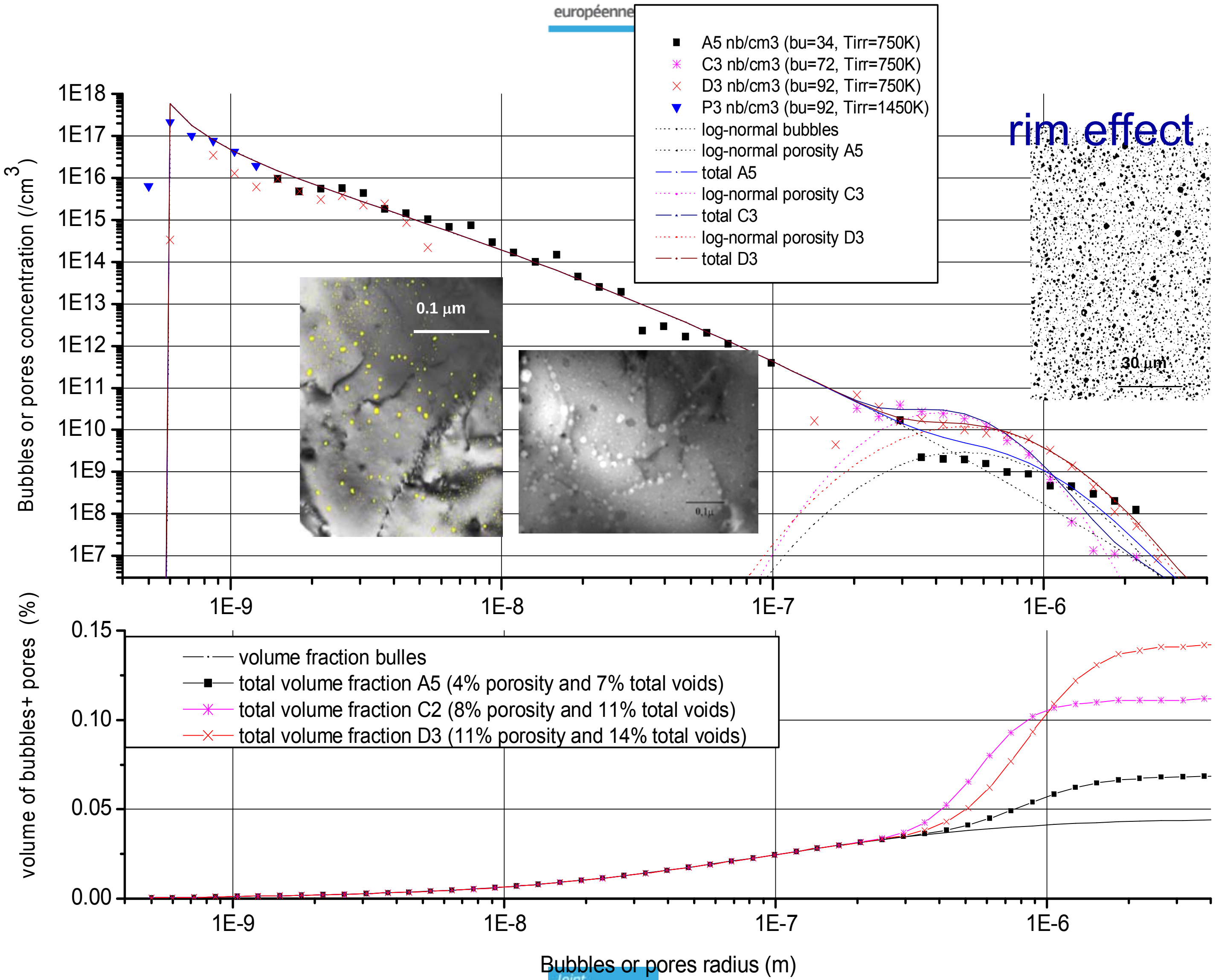
q: magnification

HBS d= 2.2

(Cauliflower d = 2.33)



fg bubbles and pores in irradiated UO_2



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Knudsen cell effusion setup with mass spectrometer

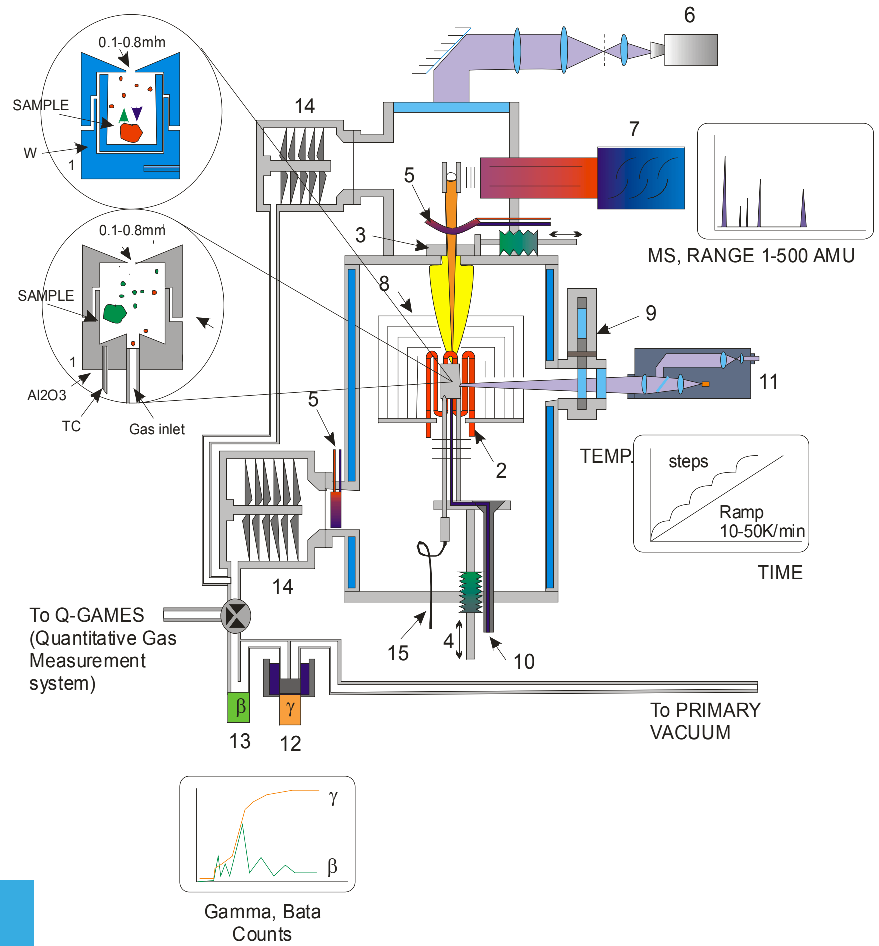
Various types of cells used

in high vacuum or under controlled, chemically reacting atmosphere up to 3100 K.

Effusion and release processes from **irradiated fuel** with temperature programs up to complete vaporization of the sample

<u>*Type of measurements:*</u>

- Equilibrium vapour pressure over the fuel
- Composition changes during annealing
- Release behaviour and analysis of He and *fg*



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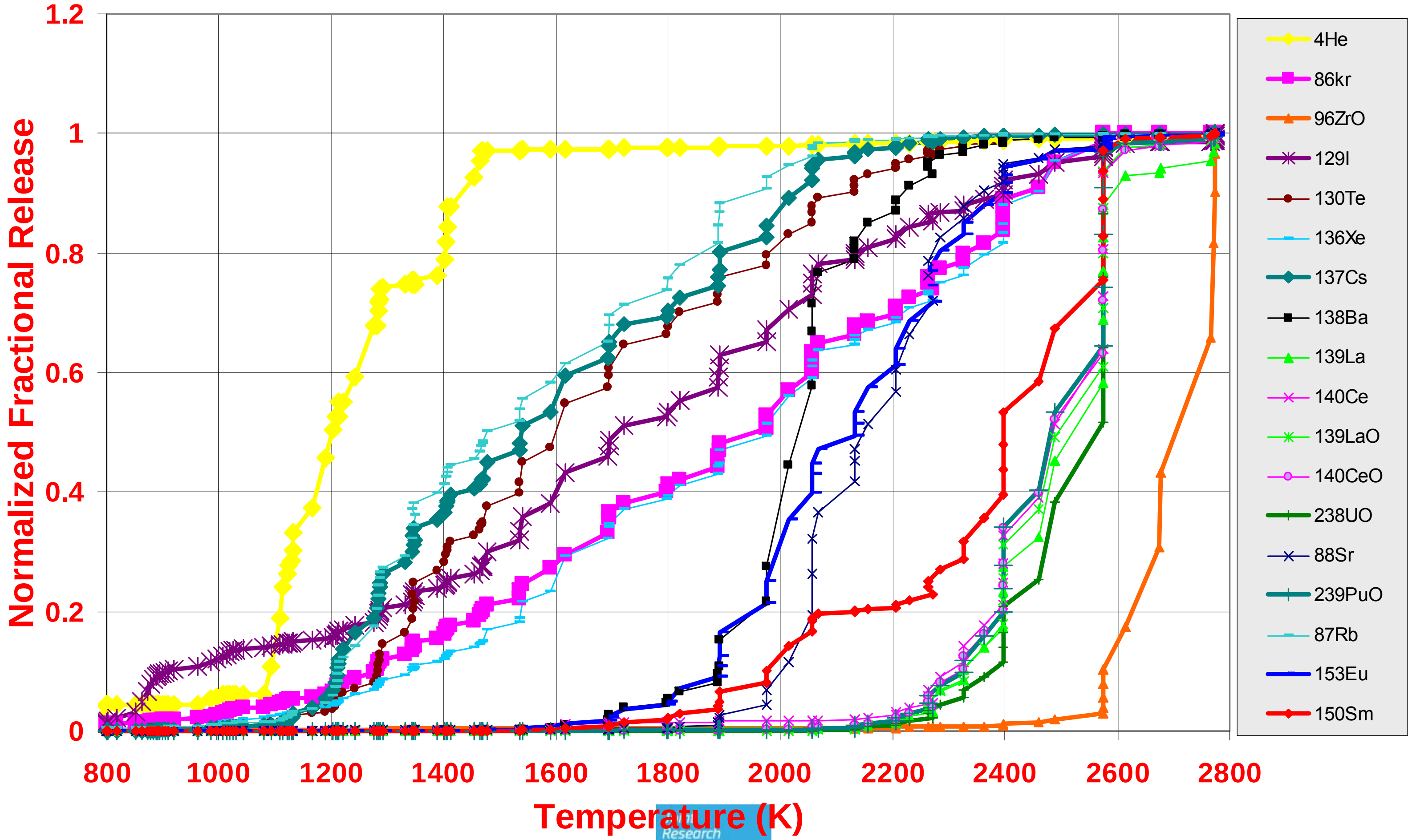
Fractional release of irradiated UO_2

sample from the central region of the pellet



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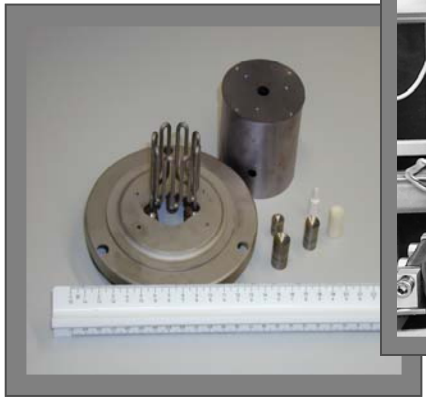
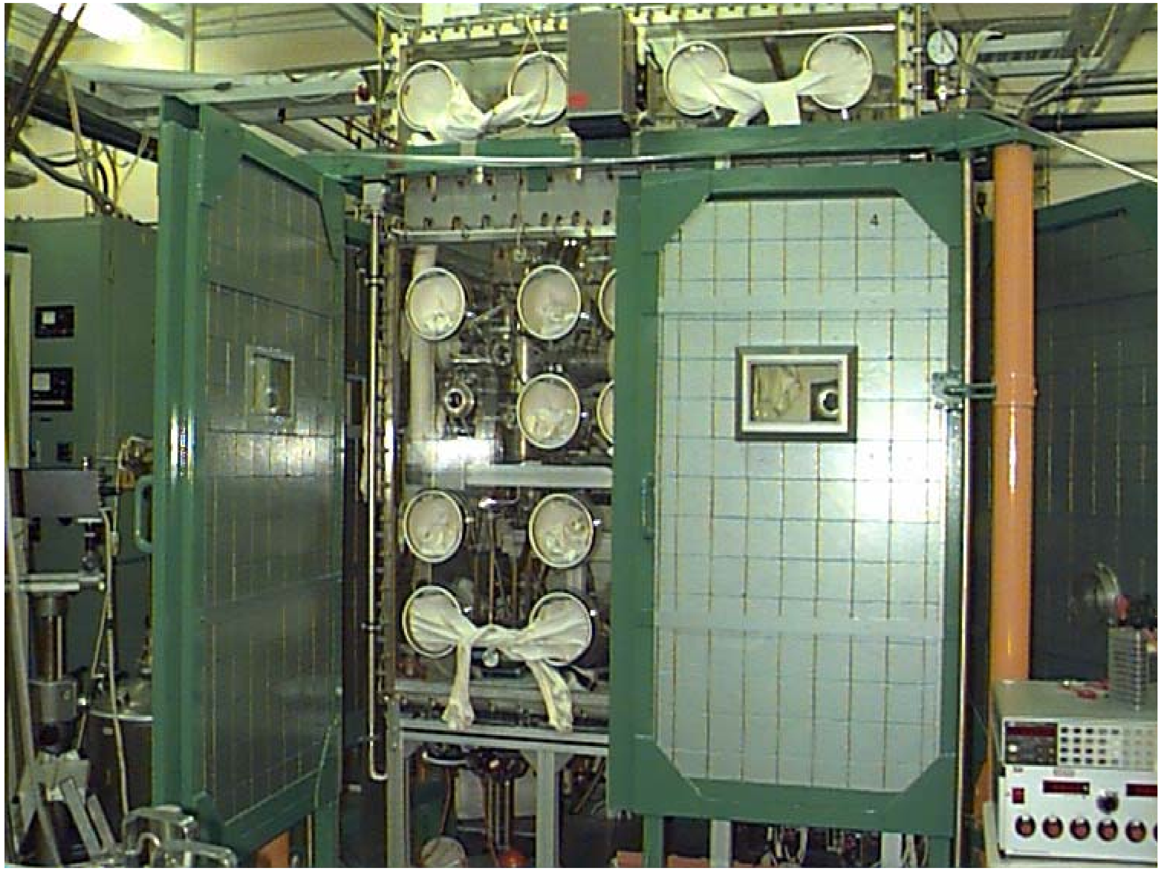
Knudsen cell effusion setup

Various types of cells can be used to work in a ultra-high vacuo or in a low-pressure controlled atmosphere up to 3000 K.

The mass-spectrometer noise is less than 10^{-13} A.

The usable m.s. signal range extends over *six* orders of magnitude.





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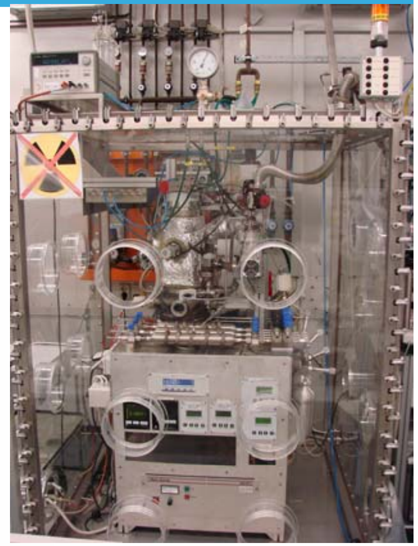
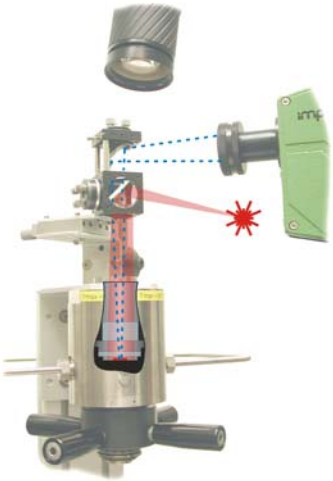
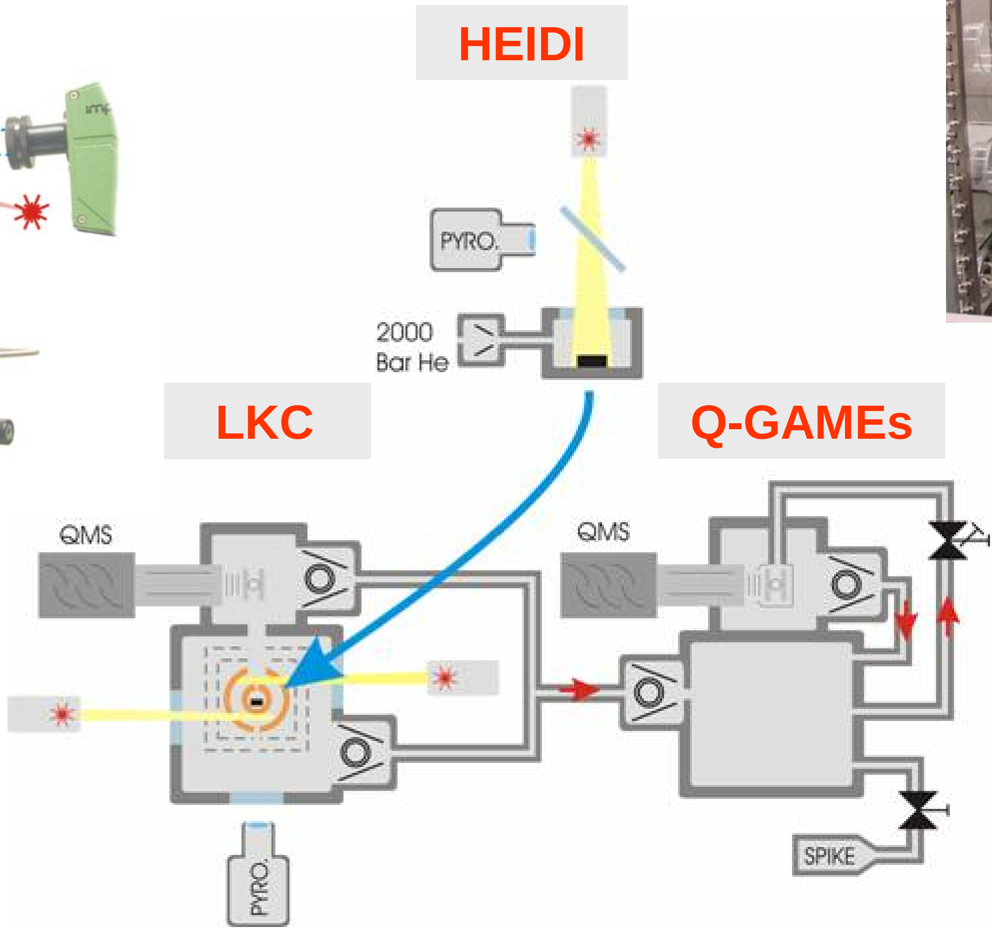
Q-GAMES







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Knudsen cell – SEM analysis

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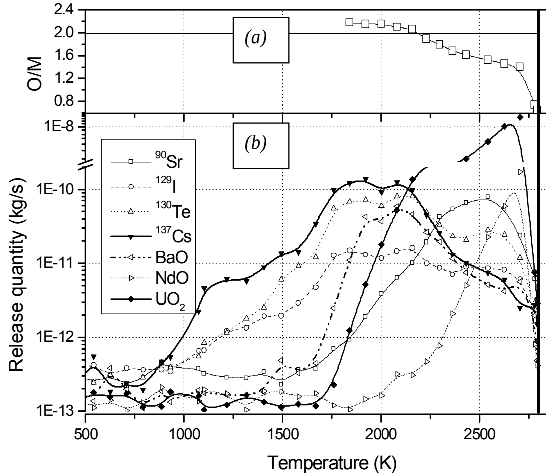
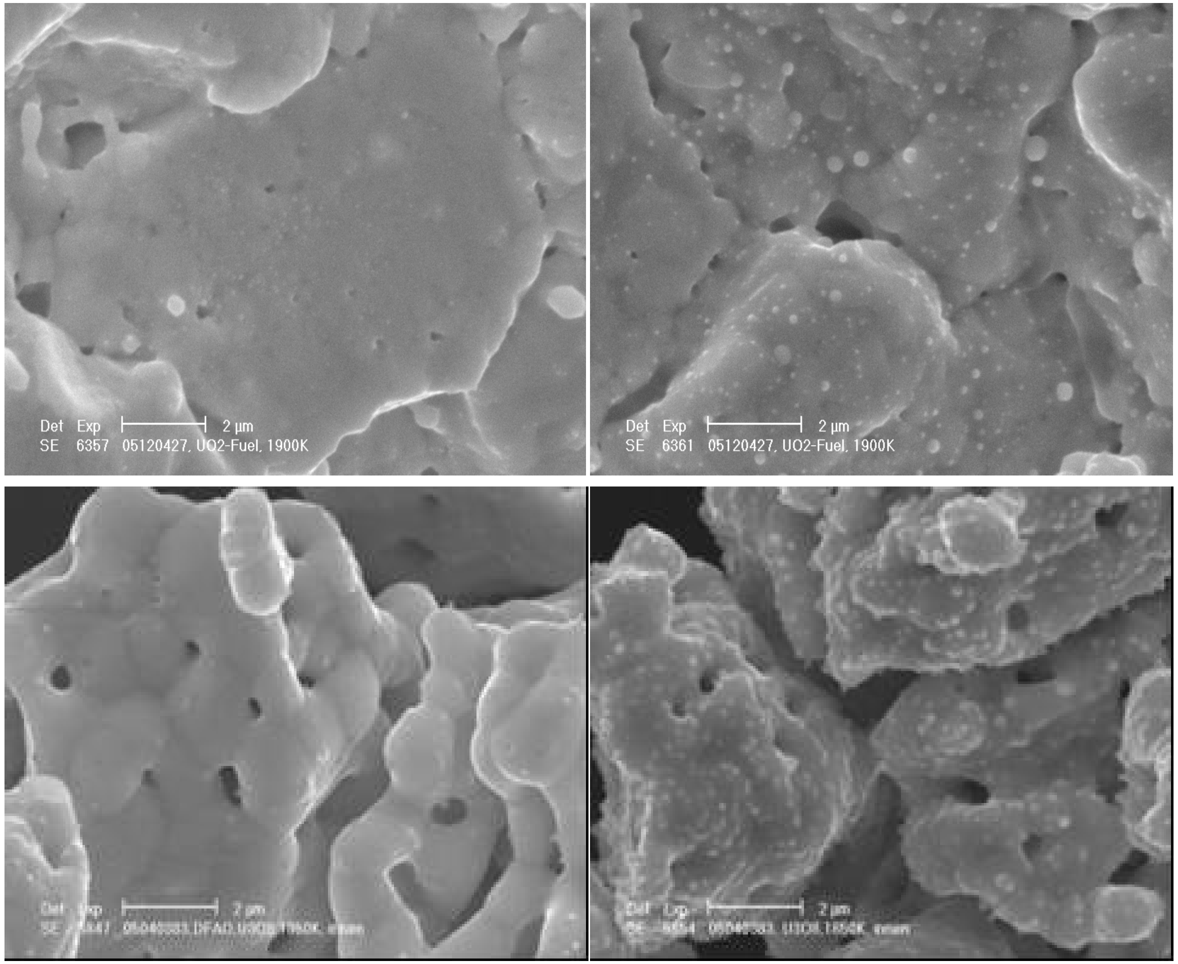
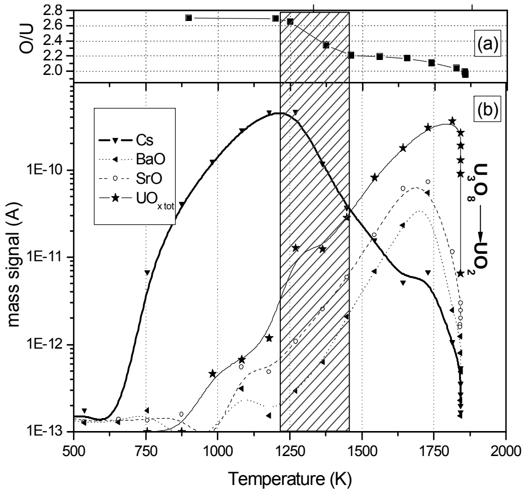
oxidation effects



Annealed 1900 K in vacuum





Annealed 1900 K preoxidized

outer surface **fracture surface**

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Lines of development for Knudsen cell

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• the effects of burnup and of irradiation temperature on the *fg* release behaviour in UO_2 have been characterized and modeled (rim structure effects)

• extension to MOX and irradiated IMF/MX fuels

• microstructural mechanisms responsible for property deterioration: parallel micro- and macro-structural characterizations using several techniques

• *fp*/fission *vs*. He/alpha decay damage

• effects of (e.g.) oxidizing conditions on the effusion behaviour

• basic properties of actinide compounds

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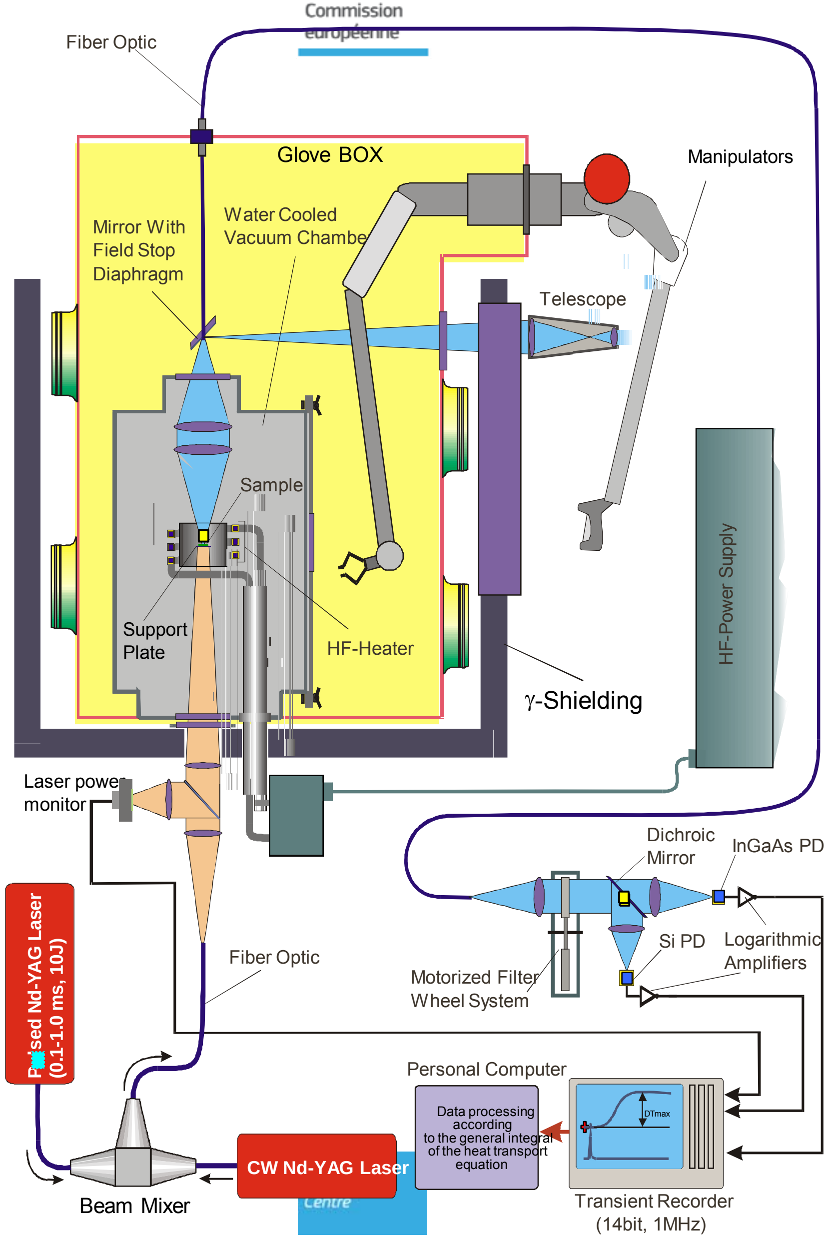
Laser flash apparatus

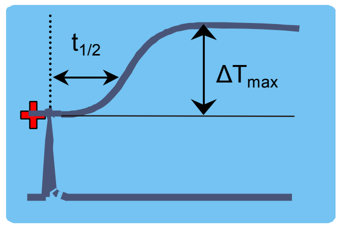
sample subject to uniform T field

vacuum condition

1. HF induction furnace is heating up the sample.
2. when sample is at homogeneous T, laser shot is fired towards sample's front face.
3. the T wave generated by the laser shot moves through the sample towards the rear surface.



4. the heat front reaches the rear sample surface generating a T increase.
5. the increasing T thermogram measured by highly sensitive fast pyrometer.



$a = 0.13885\ L^2/t_{1/2}$

$C_p = Q^*/DTmax$

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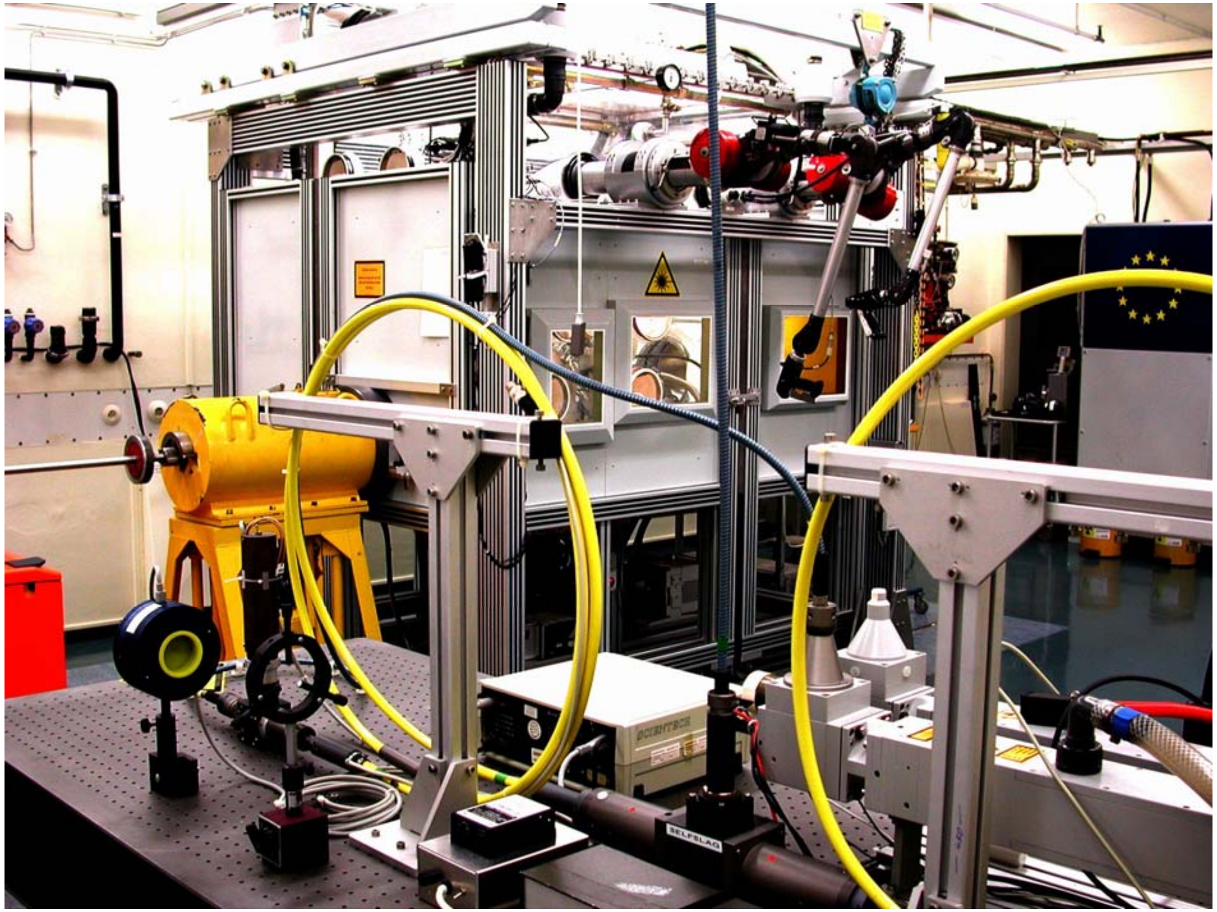
V



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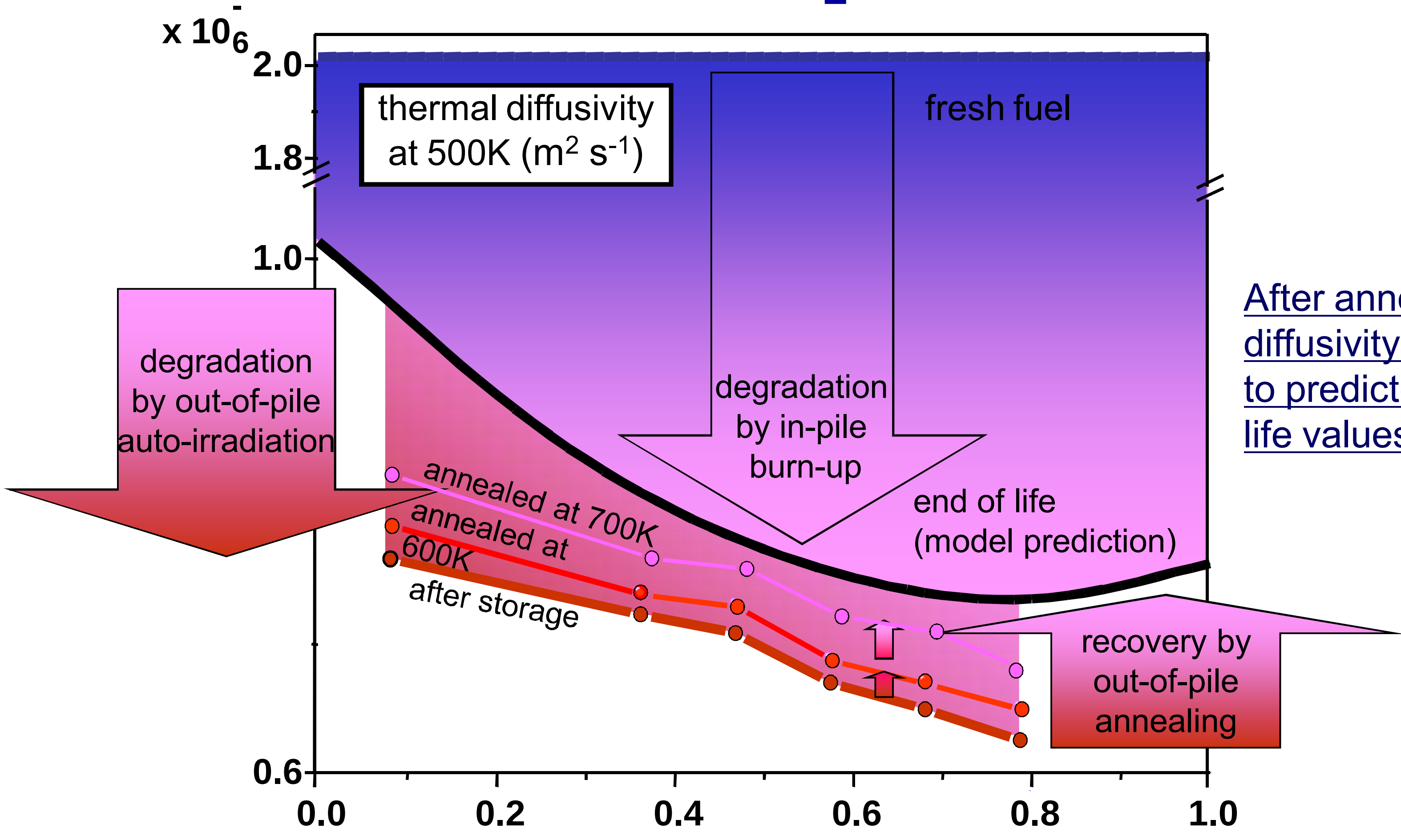
Commercial UO_2, 100 GWd/t



Radial position on the cross section of the irradiated fuel rod (r/r_0)

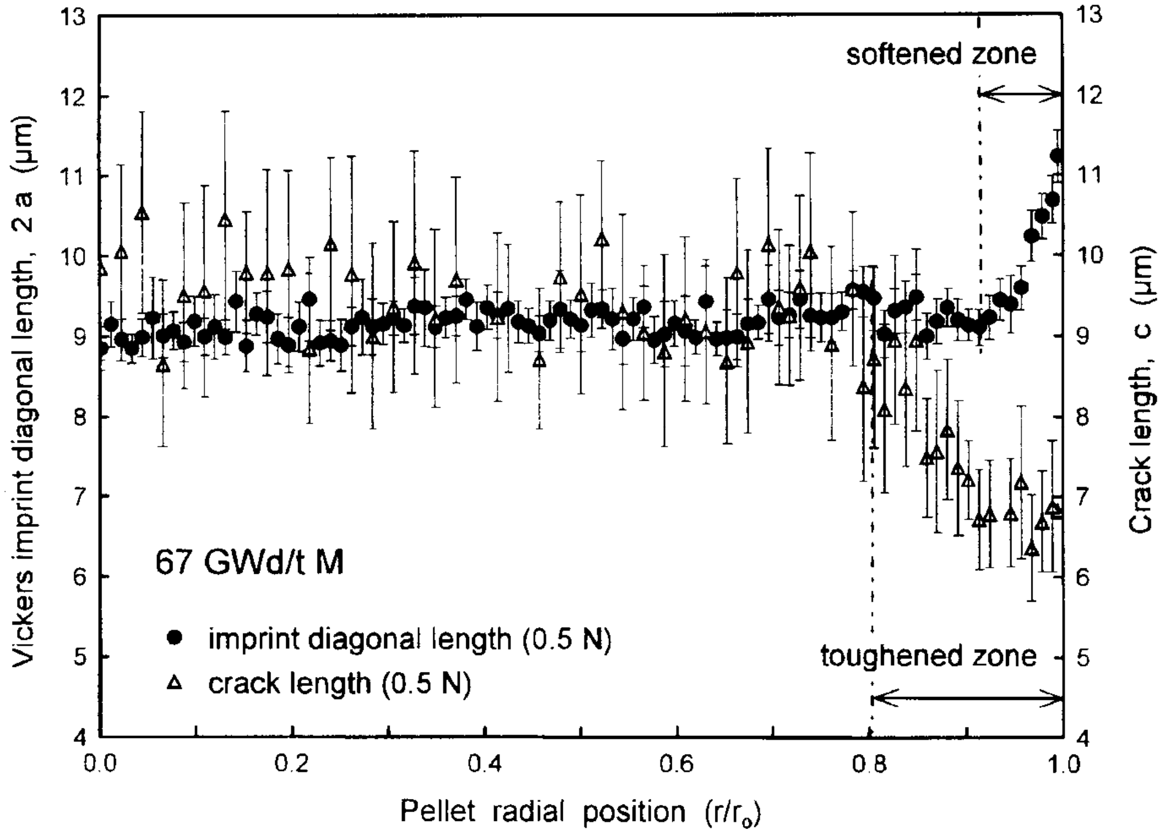
After annealing, diffusivity converges to predicted end-of-life values.

Mechanical behaviour of LWR fuel (microindentation)

Spino et al., 2003

LWR 67 GWd/tM,
indentation load 0.5 N

Radial profile of indentation:
Vickers print diagonal and crack length as a function of pellet radial position.



Softening at the rim corresponds to increased porosity due to the restructuring process

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Negative effects of radiation damage, He accumulation on waste forms

• **amorphization**:

corrosion in water 20-50 times faster

(conditioning matrices)

• **swelling**: pressurization of clad/container

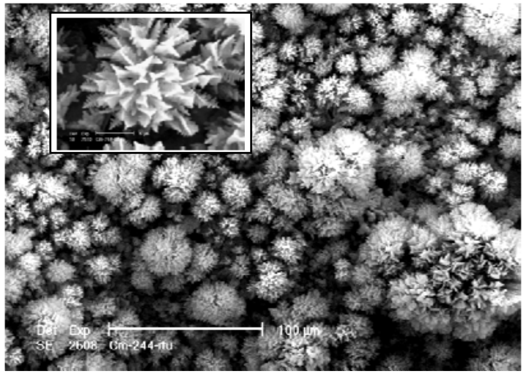
(spent fuel and conditioning matrices)

• **loss of mechanical integrity**:

surface area increase

(mainly spent fuel)

Surface structure

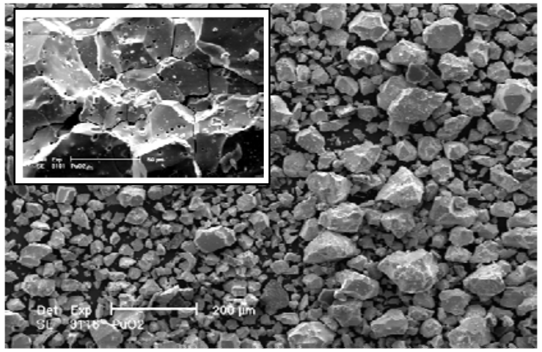


Sintered $^{244}Cm_2O_3$

extreme consequences

after 35y at RT (uncontrolled)

Ceramographic



PuO_2 sintered pellet

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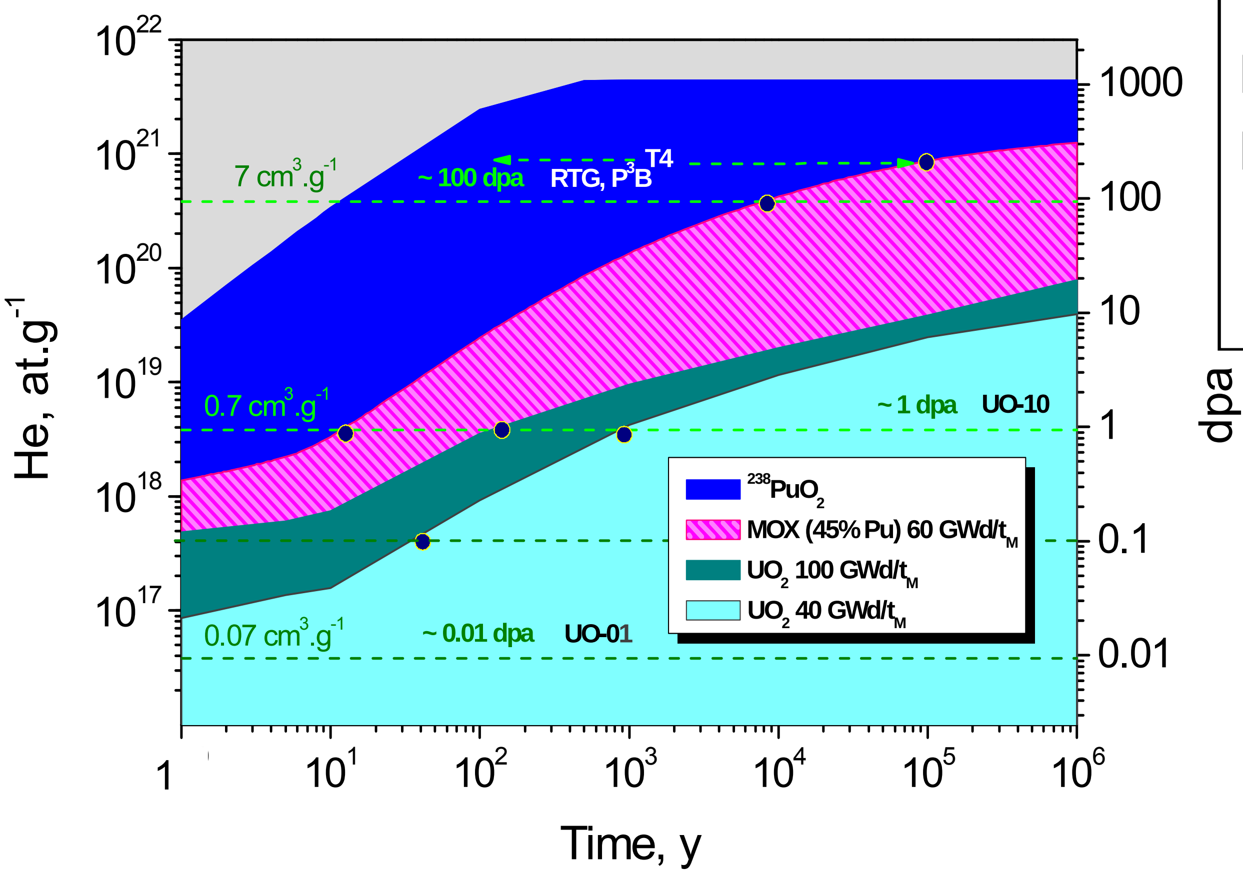
Helium formation in nuclear fuels



Free volume in a rod: 20 cm^3

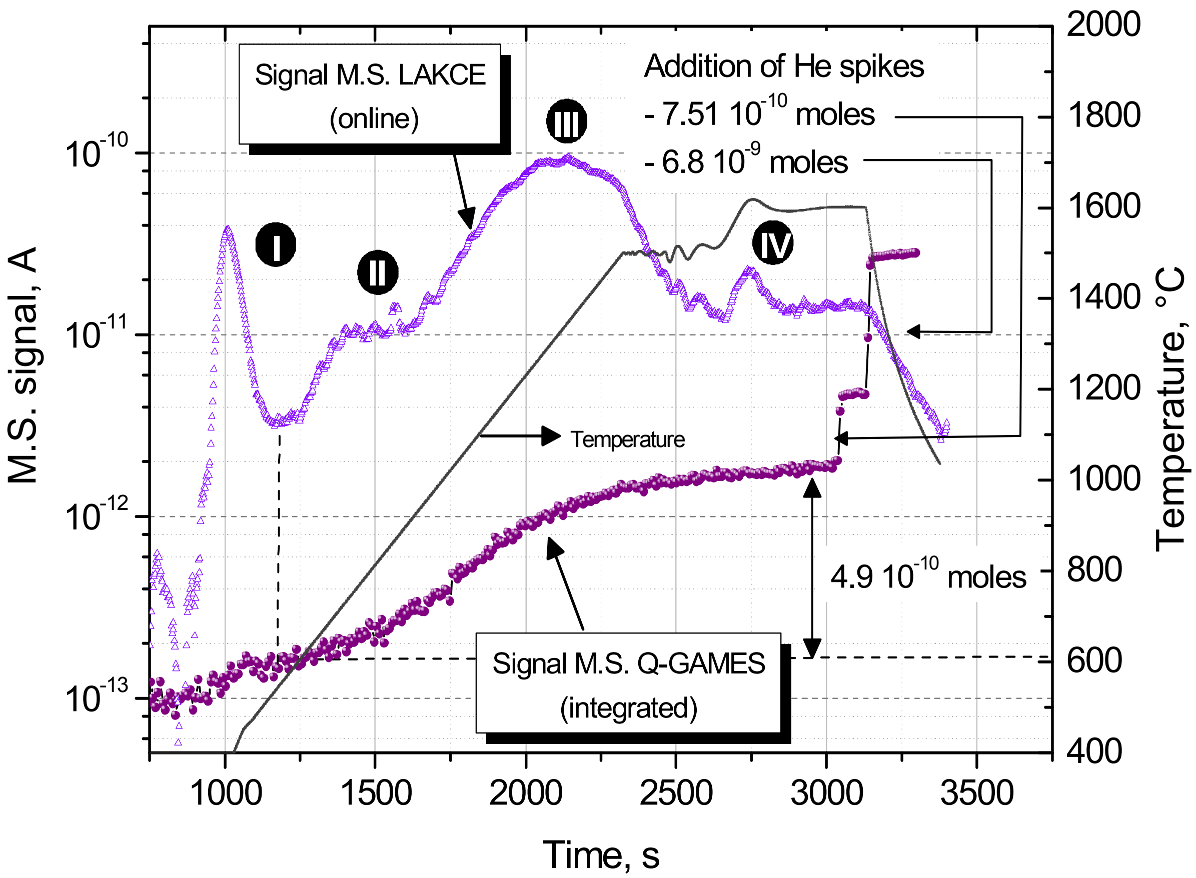
Pressure at EOI: 40 bar

Hydrostatic P disposal: 50 bar

Increase e.g. 50 bar

- Pacemaker = MOX 8000 y

Helium solubility

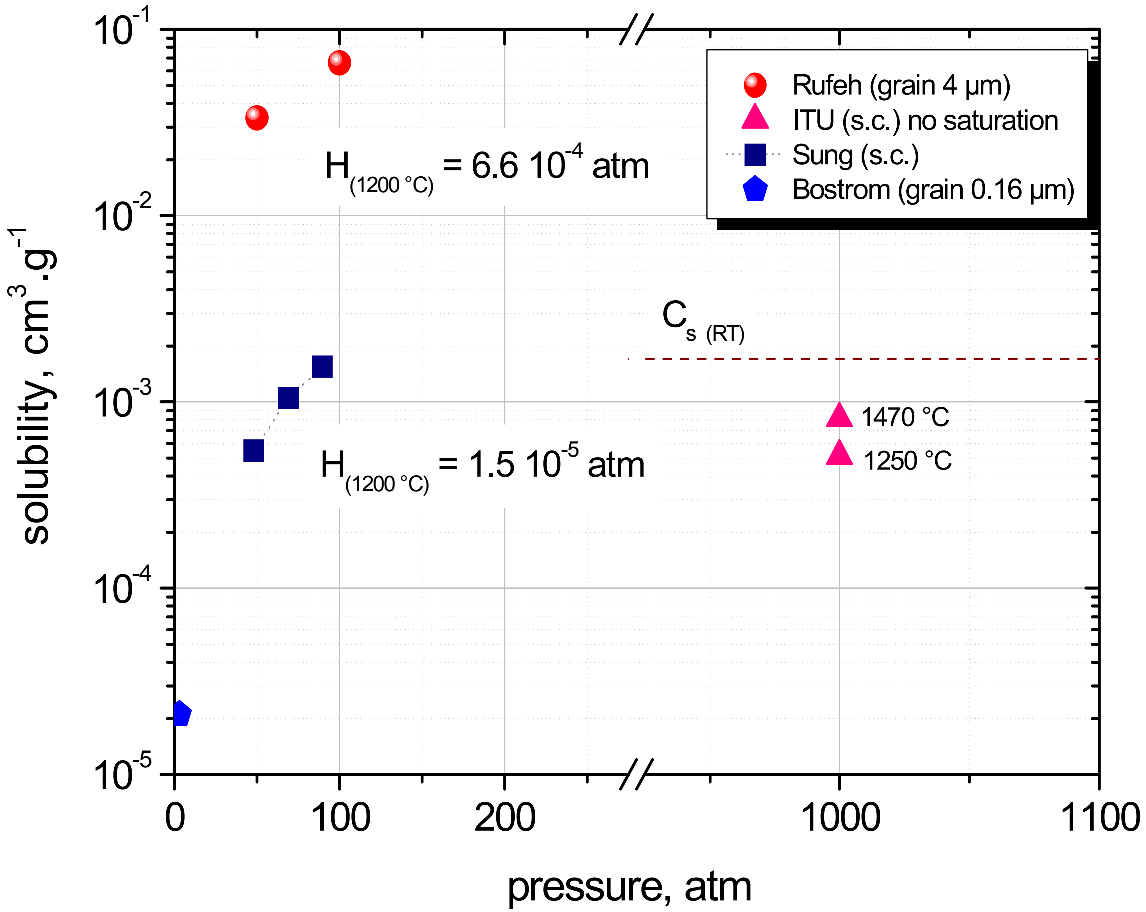


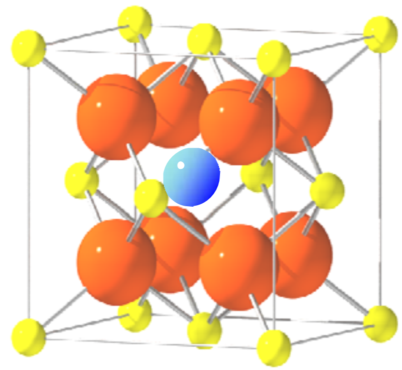
P = 1 kbar,
T = 1250 °C
t = 1 h

The solubility of He in UO_2 single crystal (for this operating conditions) is

$2.29 \cdot 10^{-8}$ mol.g^{-1} ($5.13 \cdot 10^{-4}$ cm^3.g^{-1}).

Helium solubility





Helium radius: 31 pm

Octahedral site: 37 pm

Solubility follows Henry's law (limit).

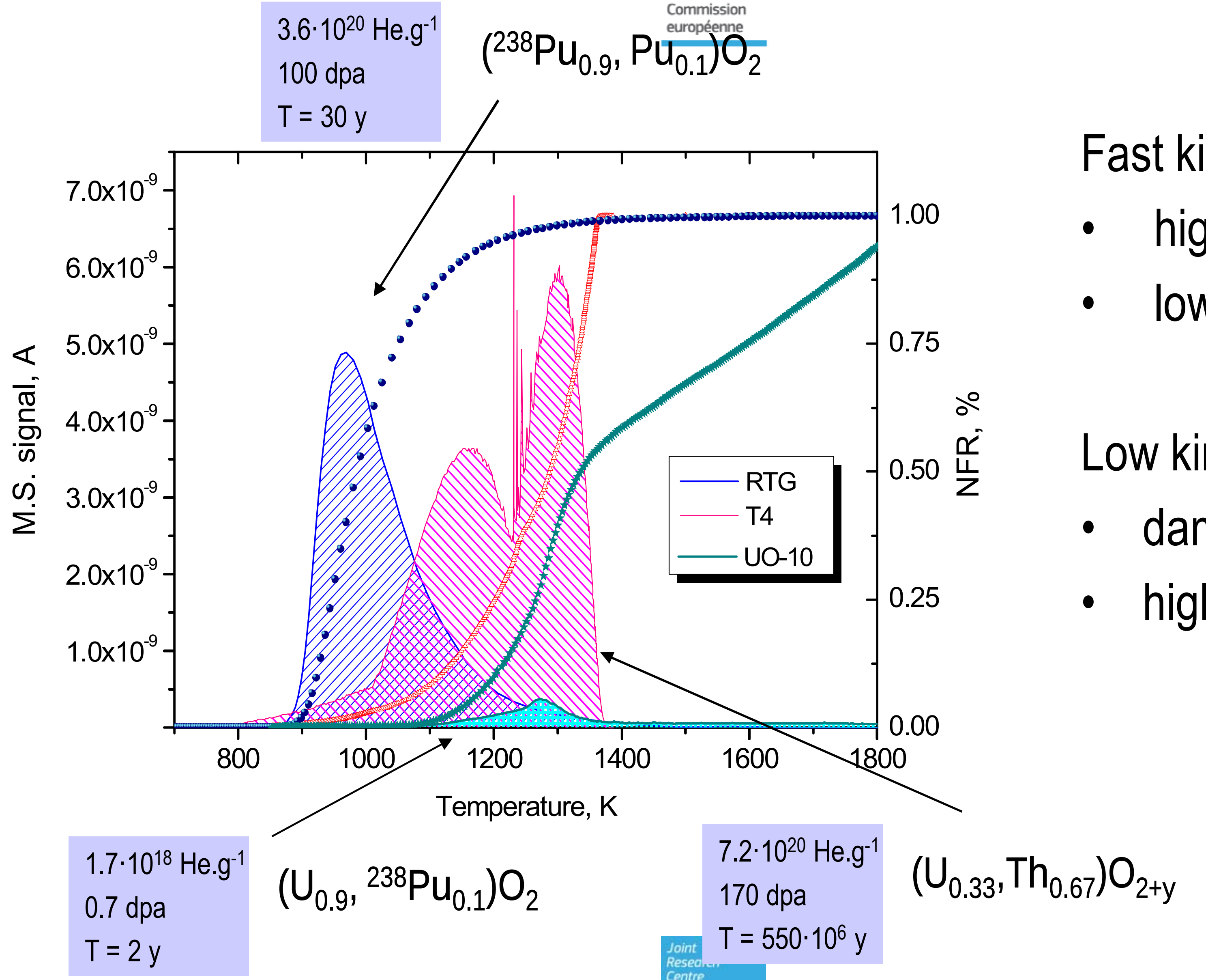
Helium occupies octahedral sites of the fluorite structure.

Sample description – Main results

sample	Original composition	Age, y	Damage, dpa	He, at.g^{-1}	Bubbles		Bubble pressure, MPa	Swelling, %	
					Average radius, nm	Conc., m^{-3}		lattice	From bubbles
UO233	$(U_{0.9}{}^{233}U_{0.1})O_2$	5	0.00001	$3.8x10^{14}$				**0.09**	
UO01	$(U_{0.999}{}^{238}Pu_{0.001})O_2$	9	0.028	$7.6x10^{16}$				**0.5**	
MOX40	$(U_{0.6}{}^{239}Pu_{0.4})O_2$	12	0.12	$4.7x10^{17}$				**0.7**	
UO10	$(U_{0.9}{}^{238}Pu_{0.1})O_2$	9	2.8	$7.6x10^{18}$	**1.2**	**$1.5x10^{22}$**		**1.3**	**0.01**
P^3B	$({}^{238}Pu_{0.9}, Pu_{0.1})O_2$	30	100	$3.6x10^{20}$	**2.5**	**$5x10^{23}$**	180	**2.2**	**3**
RTG	${}^{238}PuO_2$	36	110	$5.5x10^{20}$					
T4	$(U_{0.33},Th_{0.67})O_{2+y}$	$550x10^6$	170	$7.2x10^{20}$	**3**	**$8x10^{23}$**	320	1.5	**9**
U2	$(U_{0.92},Th_{0.08})O_{2+y}$	$220 x10^6$	130	$5.8x10^{20}$				1.5	

Nb: the van der Waals equation of state has not been used to calculate P_{bubble}

He desorption from different systems



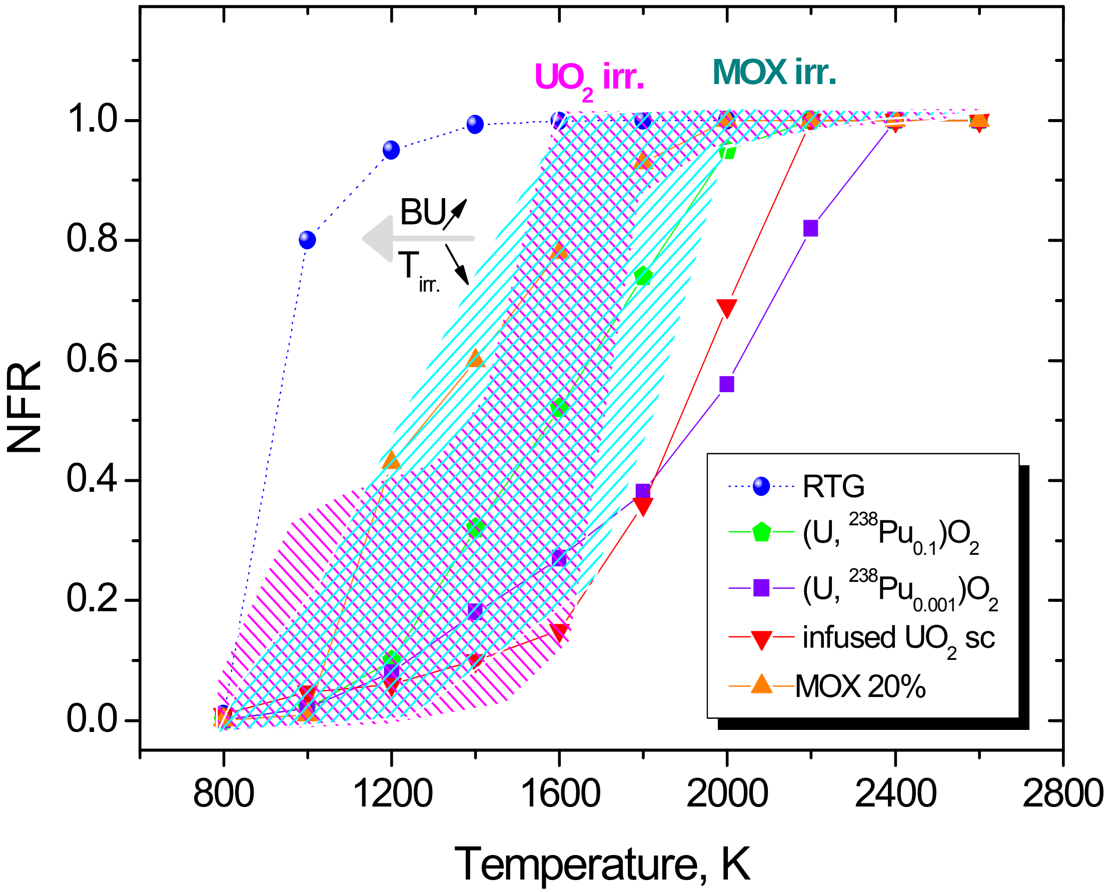
Fast kinetic :

- higher damage
- low release T

Low kinetic

- damage annealing
- high release T

He-release from different systems



Factors affecting the release temperature:

- Burnup
- Oxydation
- Irradiation temperature
- alpha-damage
- He content

Measurement of the Specific Heat by Differential Scanning Calorimetry (DSC)

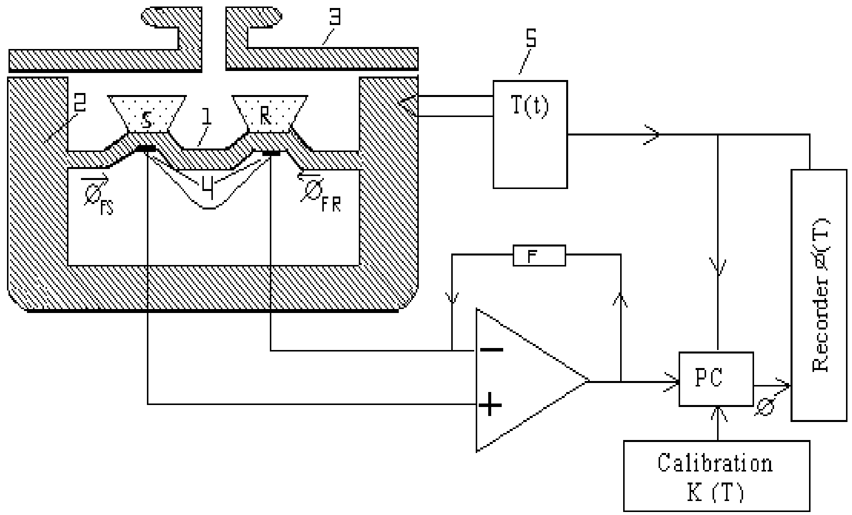
Thermal Analyzer **Netzsch STA 409**

Specific Heat for active samples

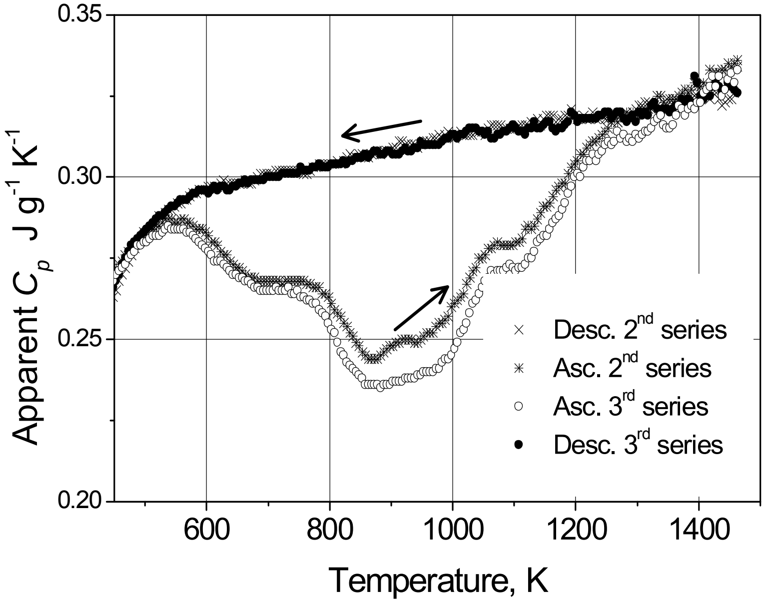
Heat effects associated to the annealing of radiation damage

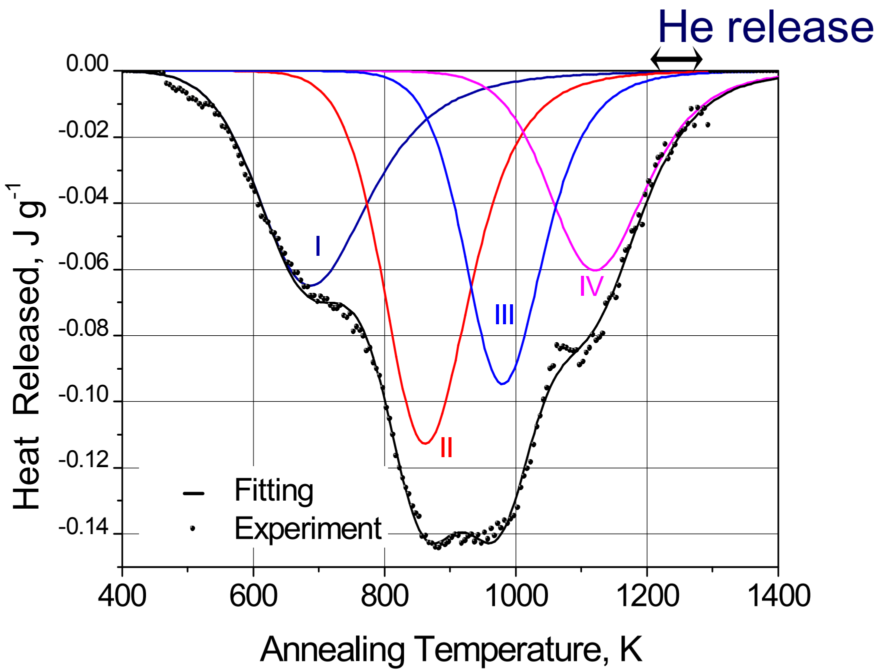
Sapphire reference sample (standard)

The characteristic feature of this measuring system is that the main heat flow from the furnace to the samples passes symmetrically through a disk of good thermal conductivity, (see Fig. 27).



Apparent C_p during annealing





Reproducible runs after 5 and 6 months storage (15 K min^{-1}).
The deviation of initial $C_p*(T)$ from annealed $C_p(T)$ is due to recovery of latent heat of lattice defects.
Curves corrected for the α-decay heat (~0.14 Jg^{-1}K^{-1})

Heat effects

I : O vacancy-interstitial recombination (14 J/g)

II : U vacancy-interstitial recombination (19 J/g)

III : Loop annealing (12 J/g)

IV : Void precipitation (15 J/g)

Thermal conductivity $\lambda = (A + BT)^{-1}$

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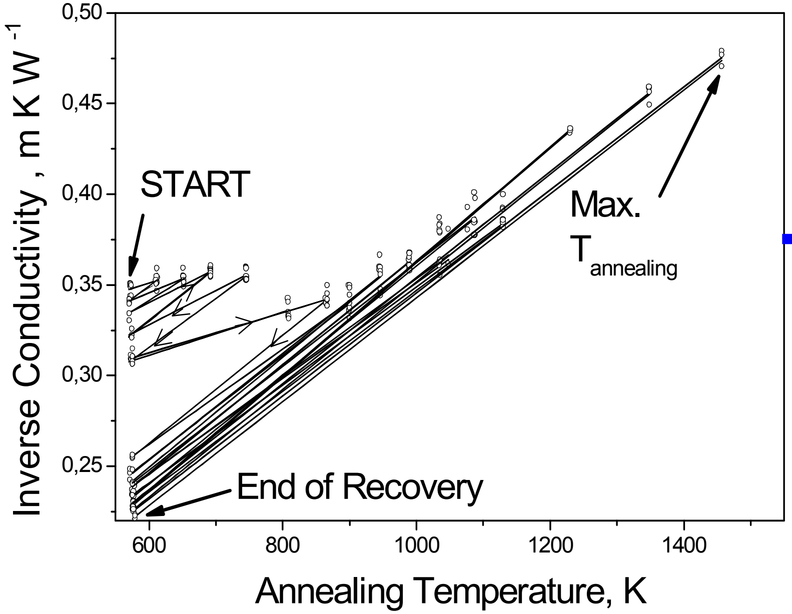
A **accounts for phonon-impurity scattering due to point defects** [f*(T), saturation]

B **should account for variation of the elastic properties with increasing damage:**

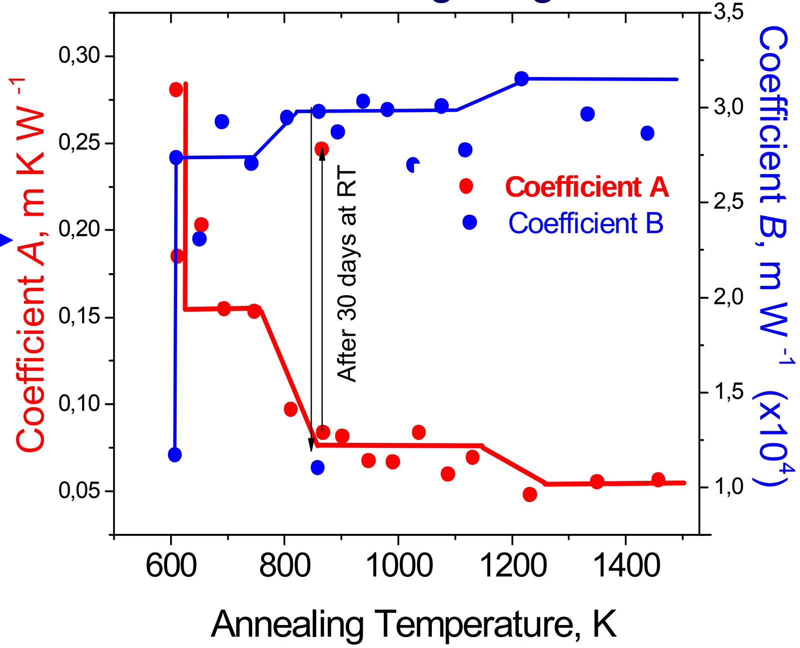
effect of thermal annealing: decrease of *A* and increase of *B*

λ degradation does not depend linearly on the concentration of defects, saturation occurs early; thermal recovery of λ can be used only to determine T ranges for healing.

α-doped UO_2 after 5 y storage



3 annealing stages



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Annealing of microstructural defects

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Combined analysis of independent recovery processes:

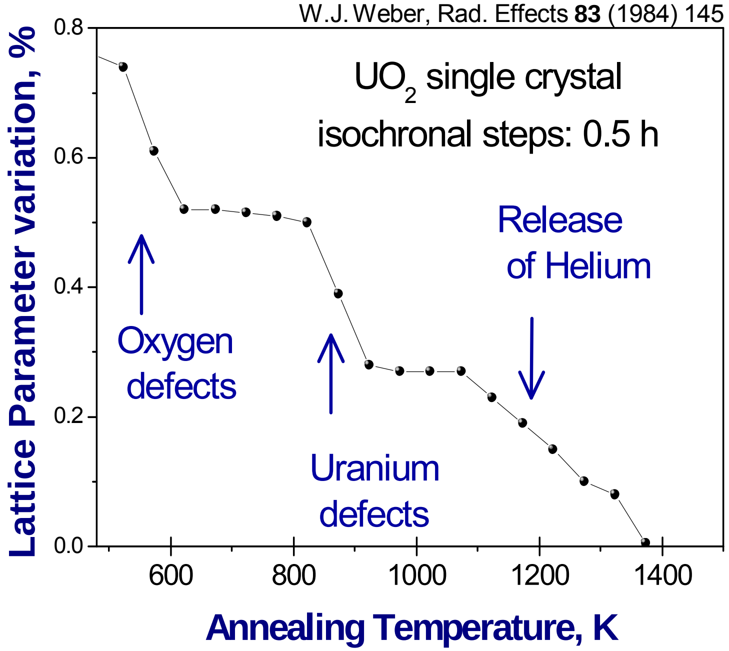
- microstructure (void/dislocation)
- calorimetry
- He release
- thermal conductivity
- lattice parameter

✓concentration of defects:
- microstructure examination
- lattice parameter changes

✓determination of temperature stages

✓energy associated with defects (apparent C_p)

Lattice parameter annealing:3 stages



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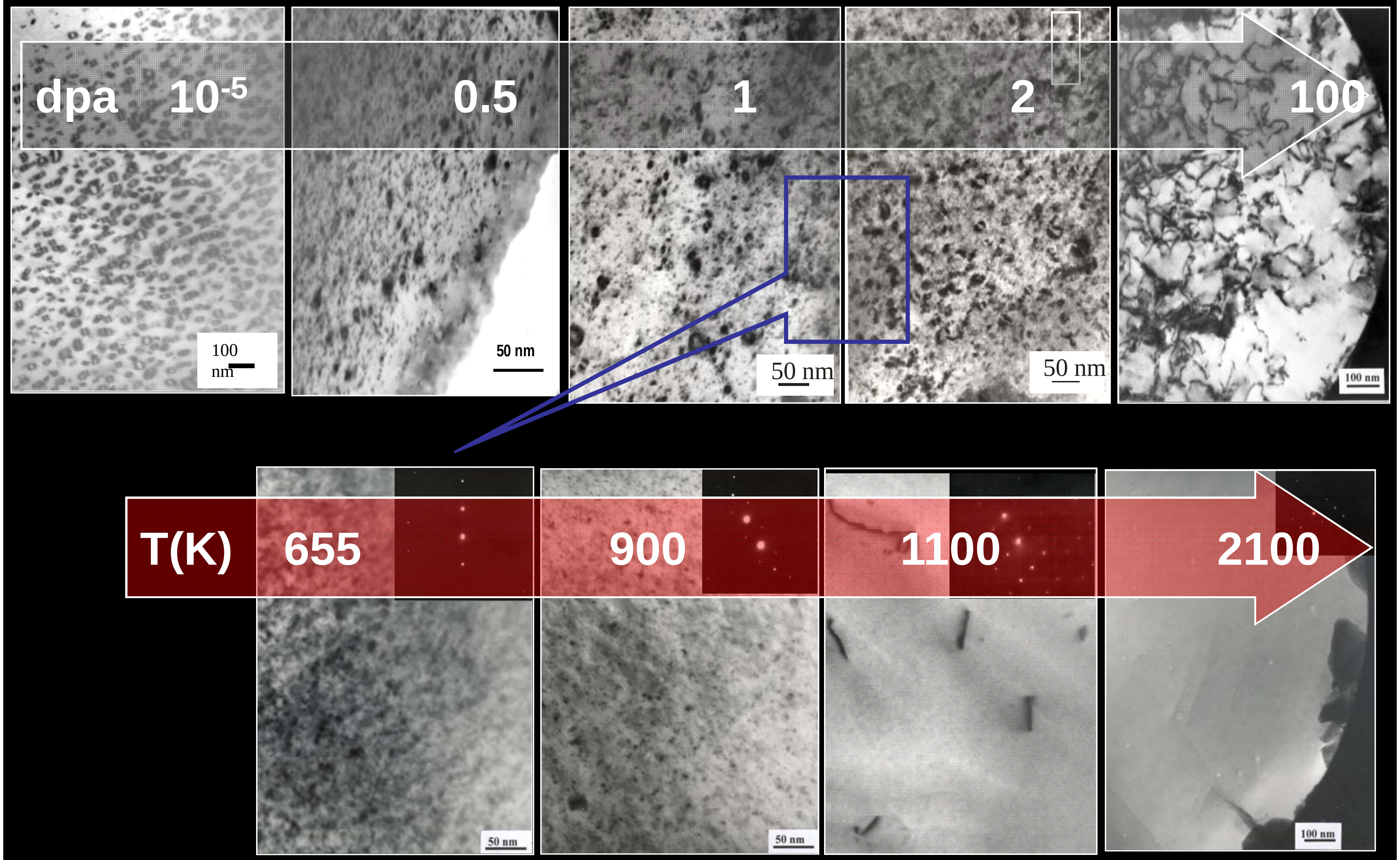
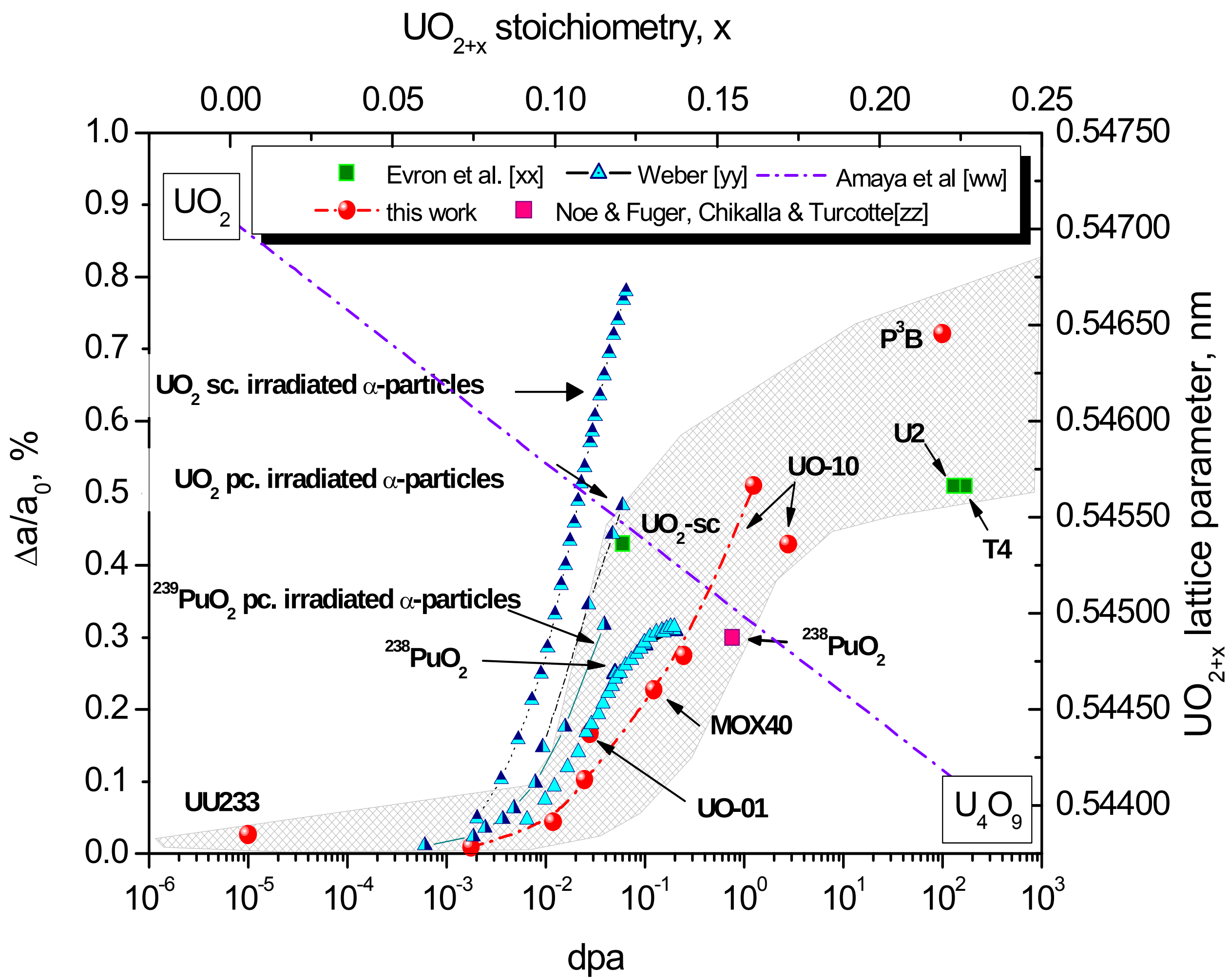
TEM alpha-damaged $(U_x,Pu_{1-x})O_2$

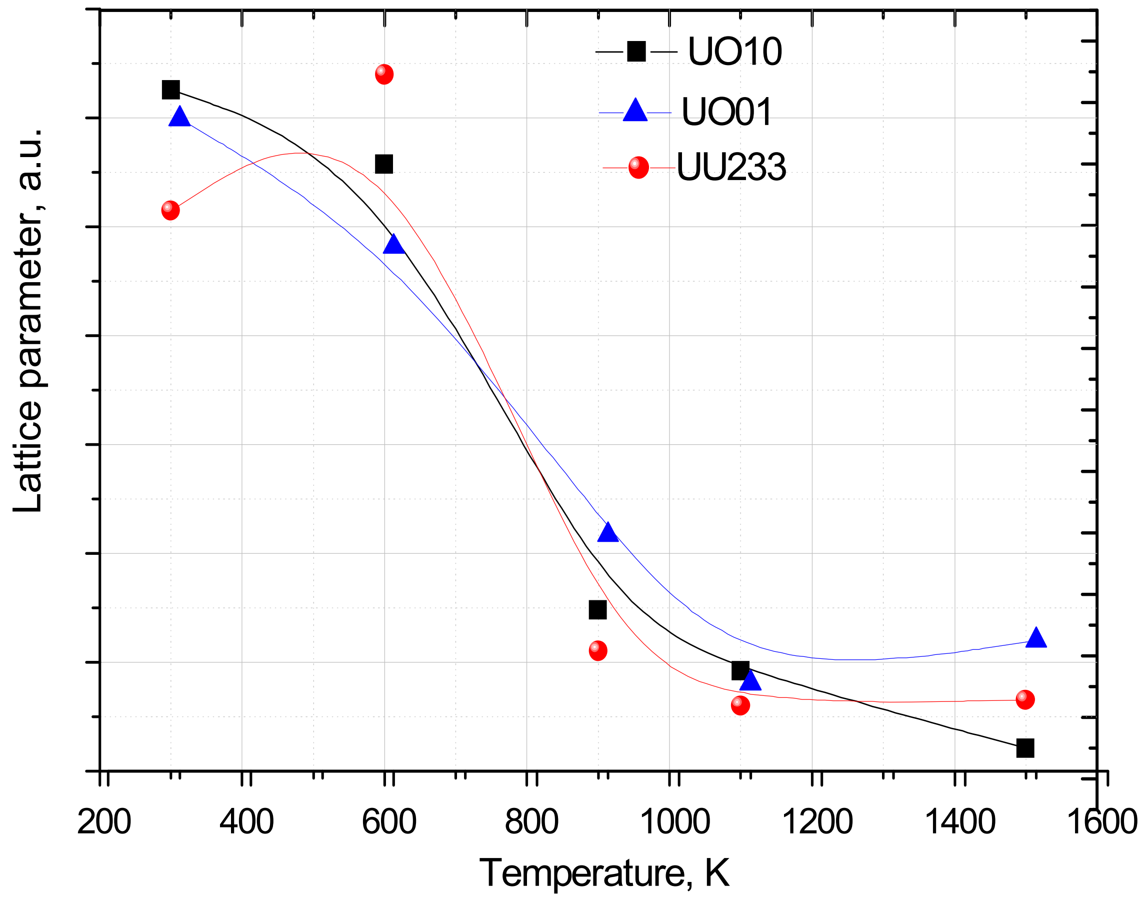


XRD analysis of α-damaged samples



- Oxydation a↓
- Damage a↑
- Kinetic effects
- Saturation

XRD annealing of alpha-doped UO_2



Representativeness of accelerated damage

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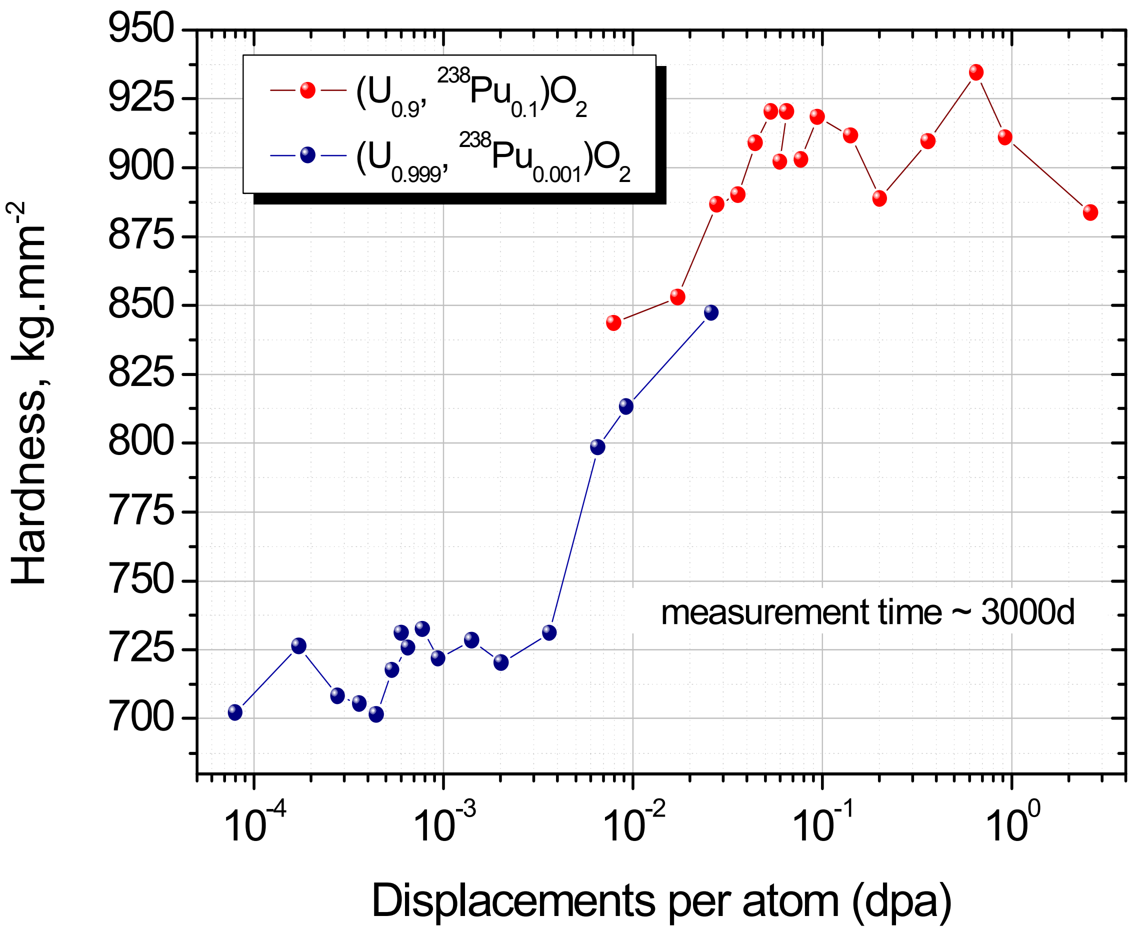
hardness *vs.* damage

The evolution of hardness for these materials is determined by accumulated damage and not affected by composition.

The dose rate does not affect the property evolution in this range of alpha-activities.

Accelerated damage accumulation is representative of aging process



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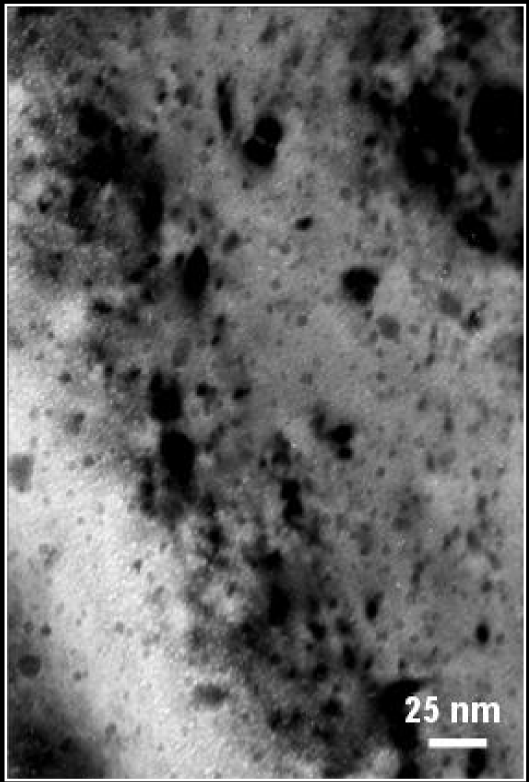
TEM analysis of $(U_x, Th_y, Pu_z)O_2$

$(U_{0.9}, {}^{238}Pu_{0.1})O_2$



$1.7 \cdot 10^{18}$ He.g^{-1}

0.7 dpa

T = 2 y

$({}^{238}Pu_{0.9}, Pu_{0.1})O_2$

$3.6 \cdot 10^{20}$ He.g^{-1}

100 dpa

T = 30 y

$(U_{0.33},Th_{0.67})O_{2+y}$

$7.2 \cdot 10^{20}$ He.g^{-1}

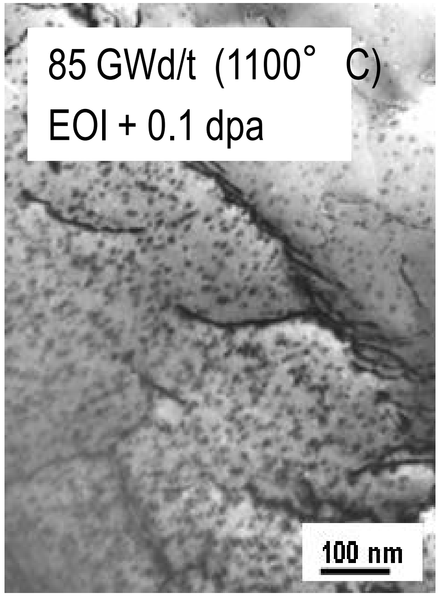
170 dpa

T = $550 \cdot 10^6$ y

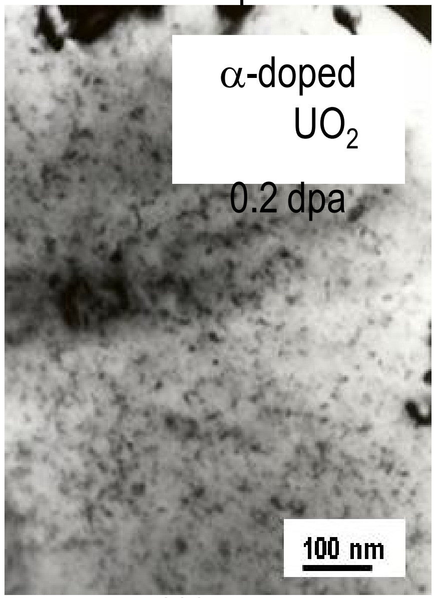
NF PRO

Alpha-damage in UO_2

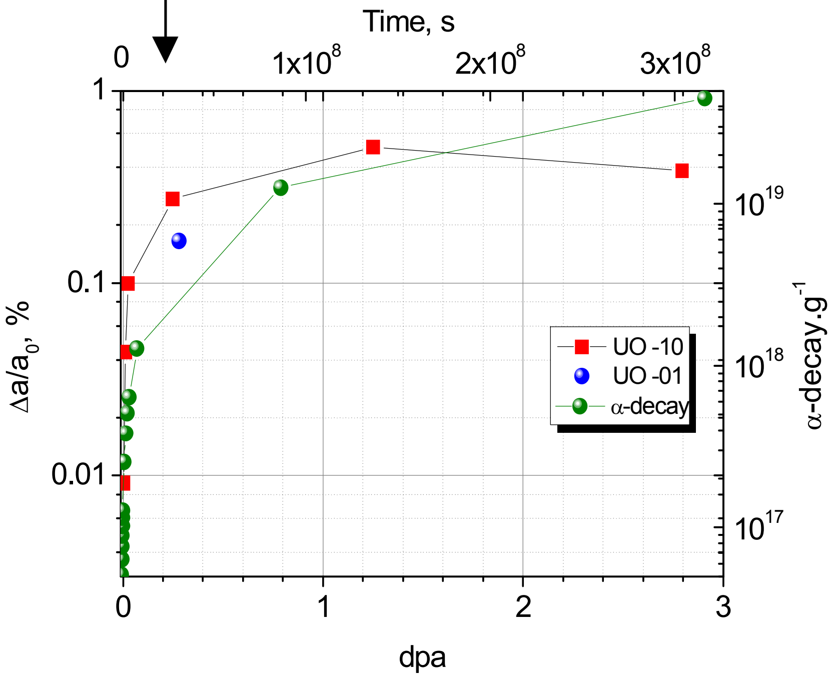
Large damage ingrowth



Irradiated fuel



$(U_{0.9}, {}^{238}Pu_{0.1})O_2$



PIE performed are affected by the growth of alpha-damage, even at low dose

e.g. lattice parameter, thermal conductivity, microstructure

Conclusion / perspectives

- Rapid lattice swelling and saturation at ~ 2%

  - consequence of extended defects ingrowth (polygonisation?)
- Low helium solubility (~ 2% MOX fuel)

  - verify the limit of Henry's low
- Gas swelling between 3 and 9 % !

  - assess the fraction retained (i.e. pressure in the bubbles using van der Waals equation of state)
- Probable embritlement/disintegration of the fuel

  - fracture stress and bubble pressure
- Extrapolation to behaviour of fuel during short time storage
- Basic aspects on helium behaviour – alpha-damage (GEN IV)

Conclusions

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- Combined analyses allowed quantification of damage and recovery process.

- Accelerated decay accumulation was validated as representative of long-term ageing of high-level waste forms.

- Comparison with irradiated fuels shows that damage effects and recovery processes during thermal annealing occur by similar mechanisms in α- and fission-damaged UO_2: → towards a unified understanding of radiation damage.

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The High Burnup Structure (HBS)

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- HBS (or RIM) structure is formed at high local burnup and low T_{irr}. It is characterized by grain subdivision, increased porosity, and evolves to an "ultimate" microstructure at very high burnup.
- No universal consensus on mechanisms and properties of HBS.
- However, it seems that HBS is not a negative feature of high burnup fuel:

  - *fg* is not released when HBS is formed

  - depletion of fission gases in the matrix, but almost complete retention in the fuel (rim porosity).

  - but release temperature decreases with decreasing $T_{irr.}$ and increasing burnup

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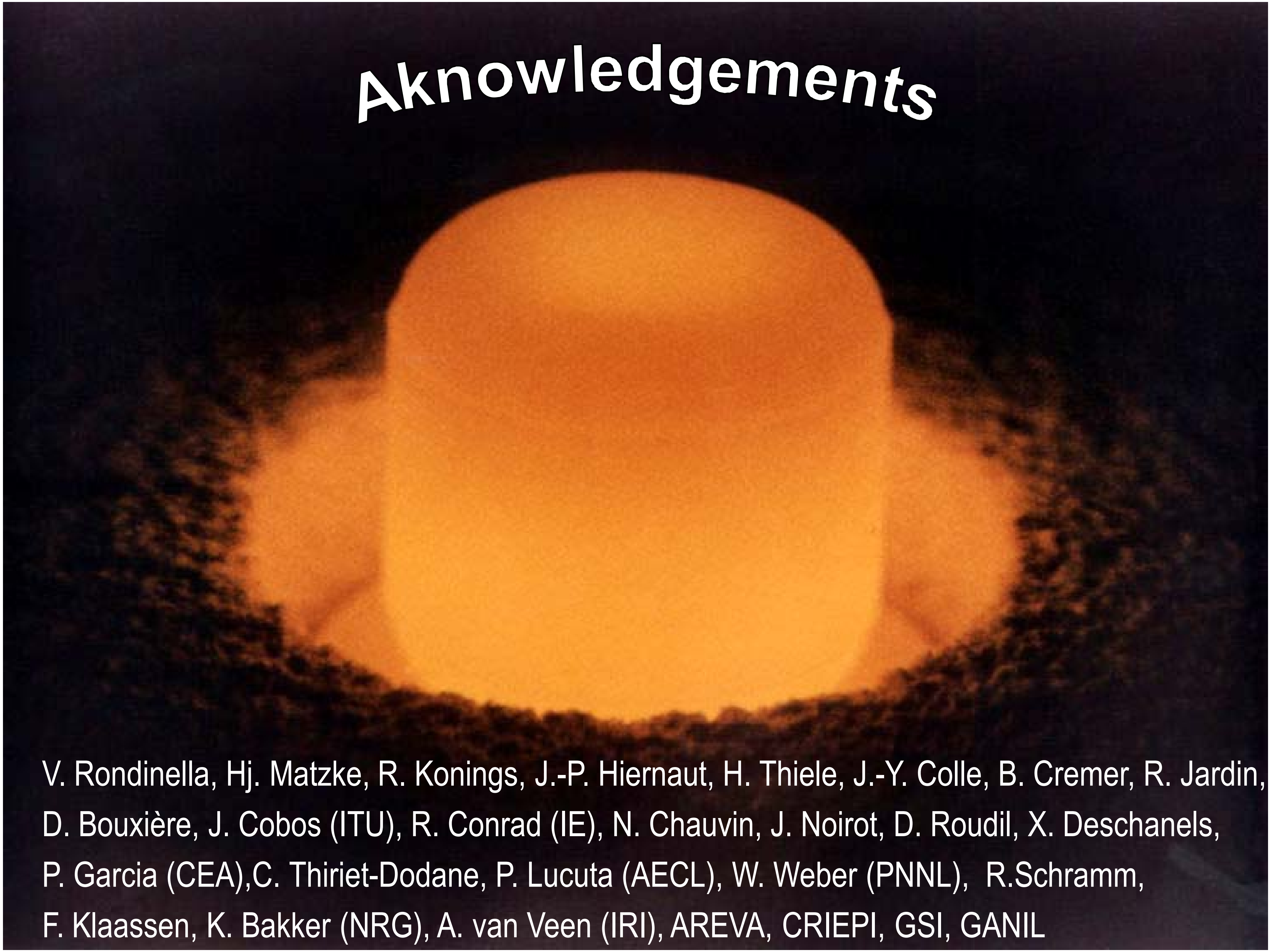

Aknowledgements

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Cladding

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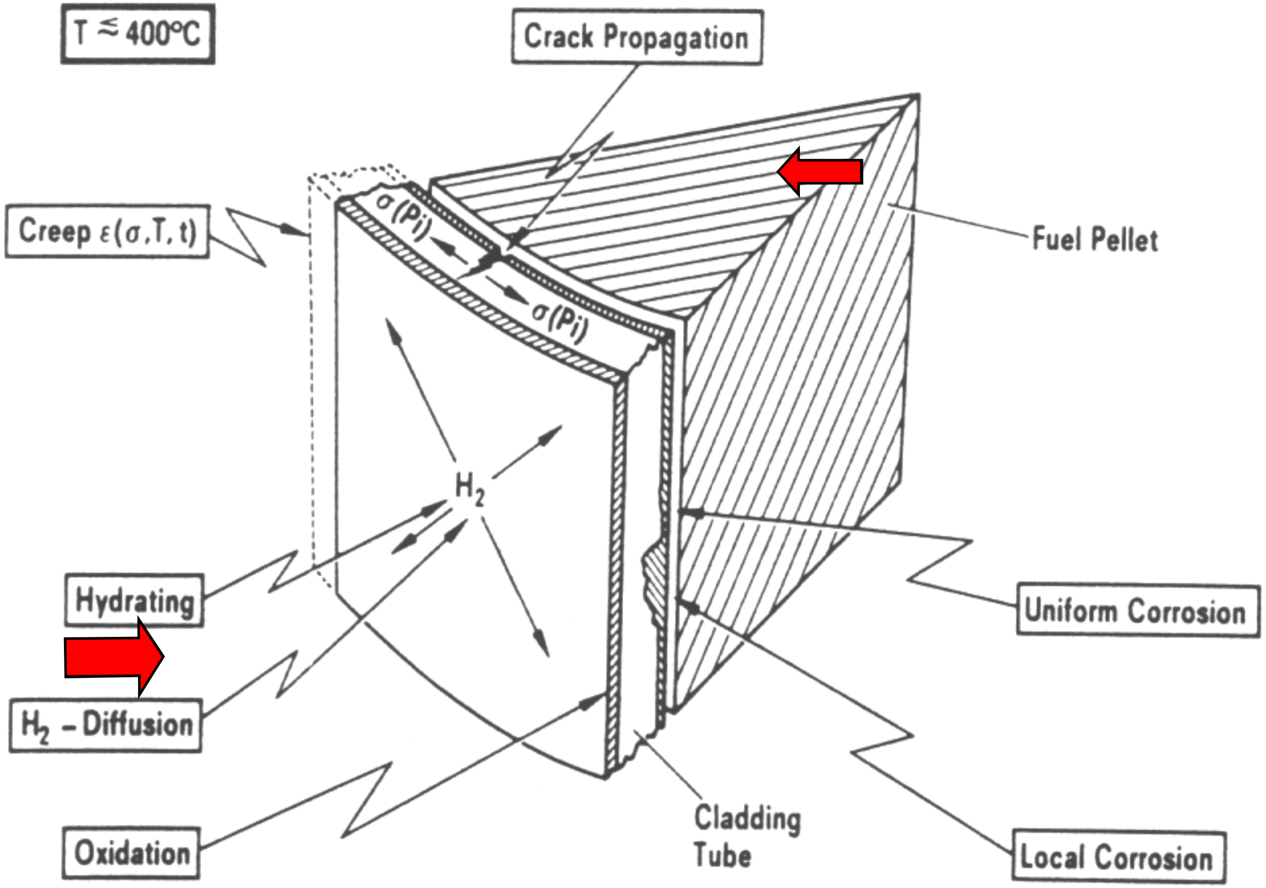
Cladding behaviour during storage (and transport)



Cladding

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Segmented Cone Mandrel Test

Displacement controlled
→ stable crack growth

“cold” development in IE

optimization and implementation in hot cell in ITU for application on irradiated cladding

extending existing creep test capabilities

Cladding

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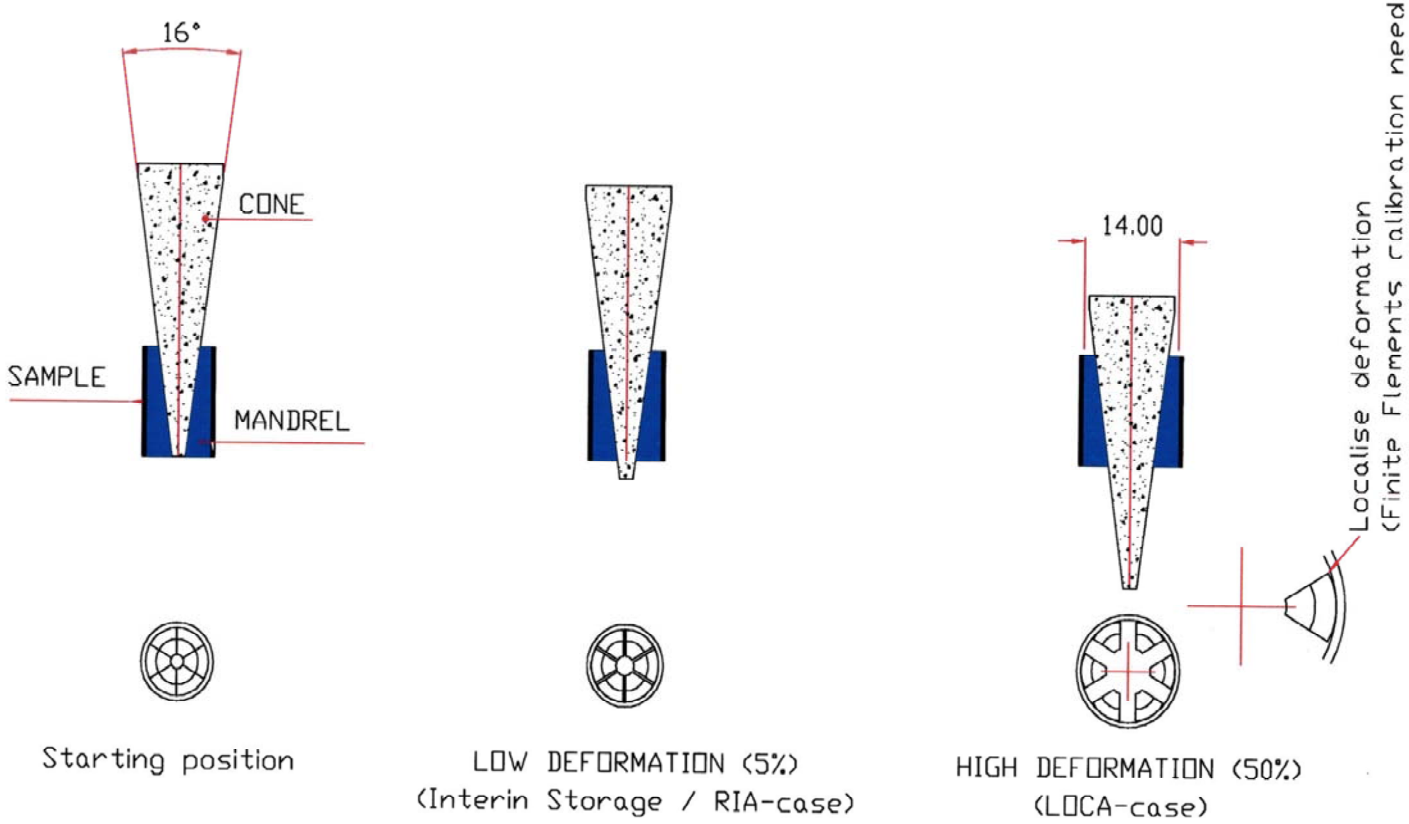
Schematics of SCMT Design

ITU - Prototype
(Cone/Mandrel - Concept)



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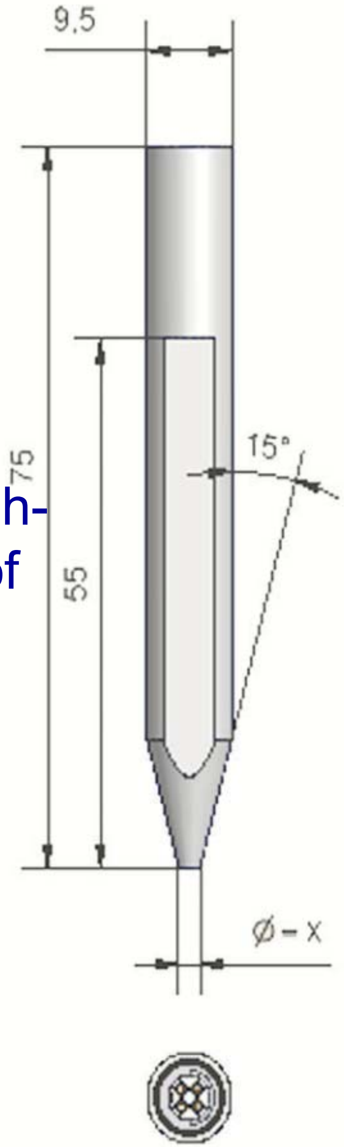
Crack detection measurement on fuel cladding based on eddy current

V.V. Rondinella, D. Papaioannou, J. Ejton,
*W. de Weerd, R. Nasyrow, H. Toscano, W. Goll**

AREVA NP GmbH, FDEEM, Erlangen, Germany



Manual (dynamic) crack detection; small surface cracks on high-alloyed in unspecified locations (independent of the direction of the inspection).

PROBE SYSTEM: Absolute, ferrite core, transformer

FREQUENCY RANGE: 100 kHz - 3 MHz

ACTIVE AREA: Approx. 1.0 mm

PENETRATION DEPTH: Low

CABLE: EK-X-HF/1, EK-X-007

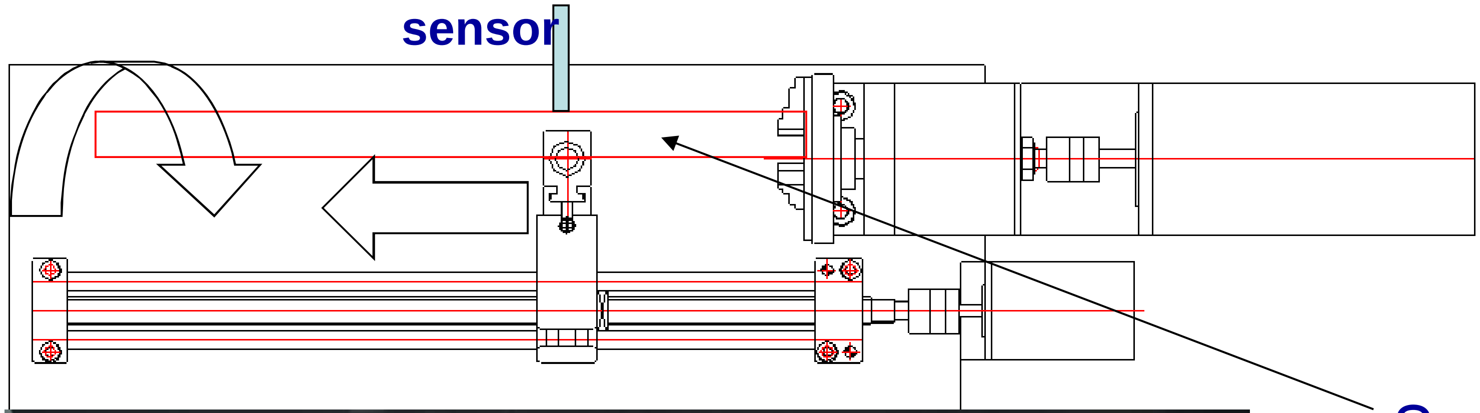
HOUSING: Plastics (Delrin); pencil housing # 2

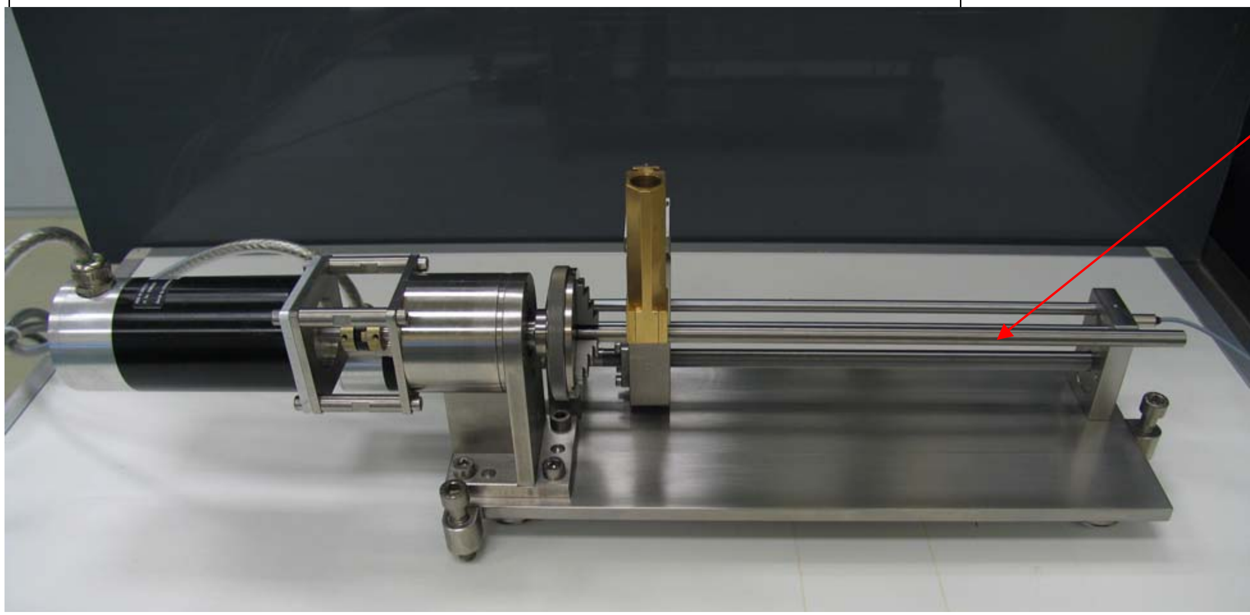
DIAMETER: 9.5 mm

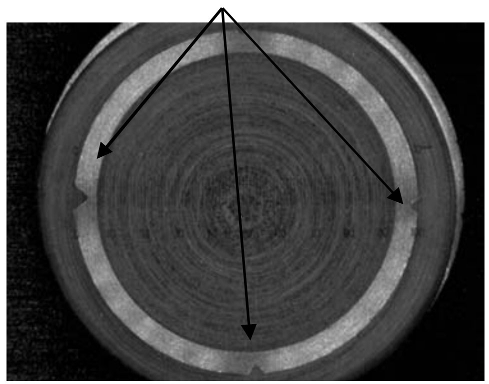
LENGTH: 75.0 mm

WEIGHT: 10 g

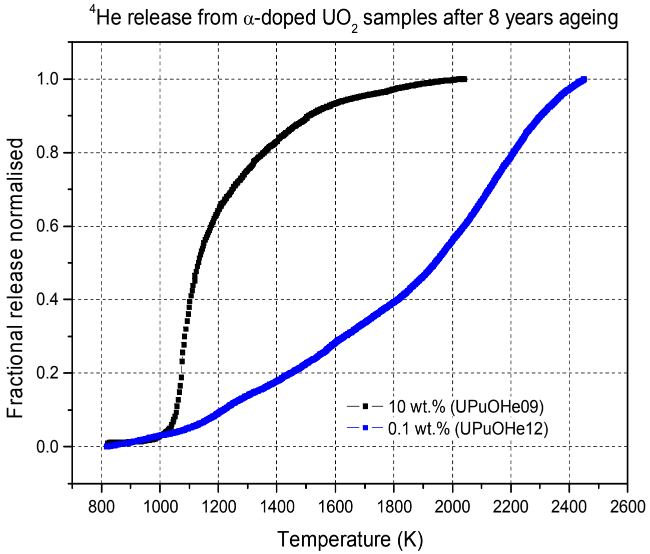
Crack detection device



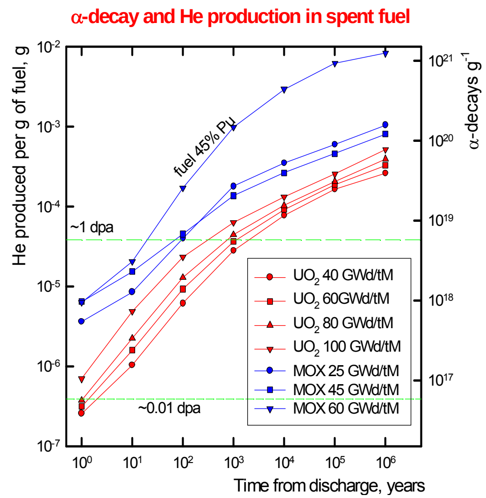


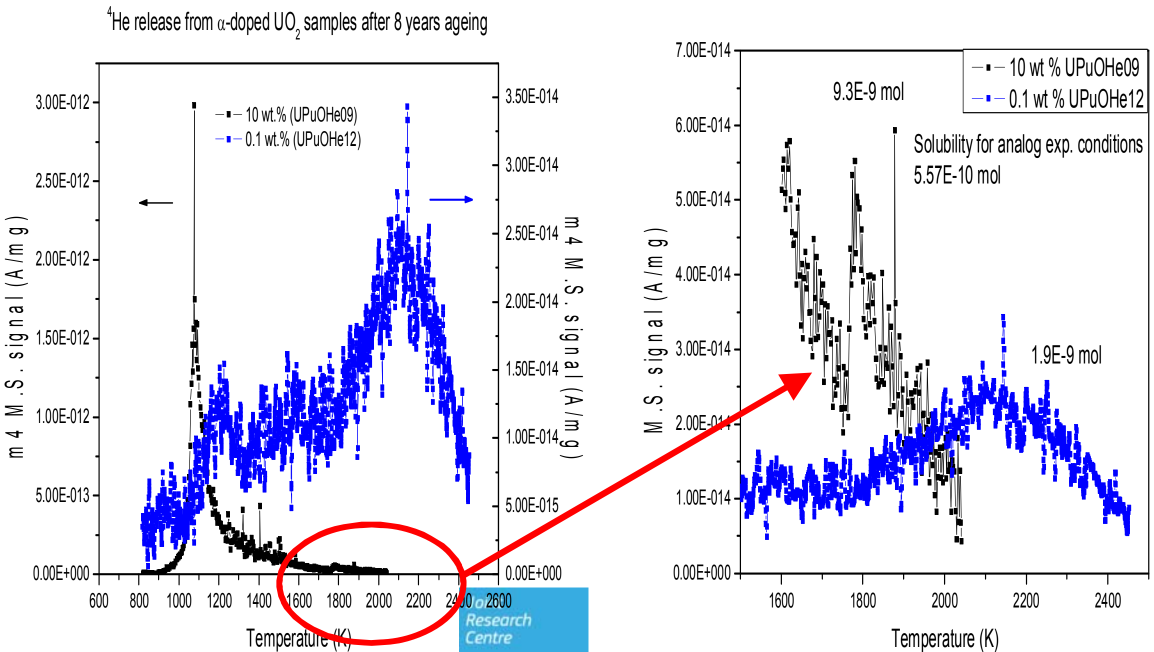


Helium release from 0.1 wt% ^{238}Pu-doped UO_2



The helium quantity released at higher temperature is close to the expected solubility.





He solubility in UO_2