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Nuclear Measurements

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Acting Director General
Joint Research Centre, European Commission, Brussels
http://www.jrc.cec.eu.int

IAEA SCIENTIFIC FORUM 2005
Nuclear Science: Physics Helping the World
27 - 28 September 2005
Vienna, Austria
Session: Developing advanced materials and technologies

JRC's Mission

… to provide customer-driven scientific and technical support for the conception, development, implementation and monitoring of EU policies …

…the JRC functions as a centre of science and technology reference for the EU, independent of special interests, private and national

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The structure of the JRC

Joint Research Centre

7 Institutes in 5 Member States ≅ 2300 staff ≅ 300 M€/y budget + 40 M€ income

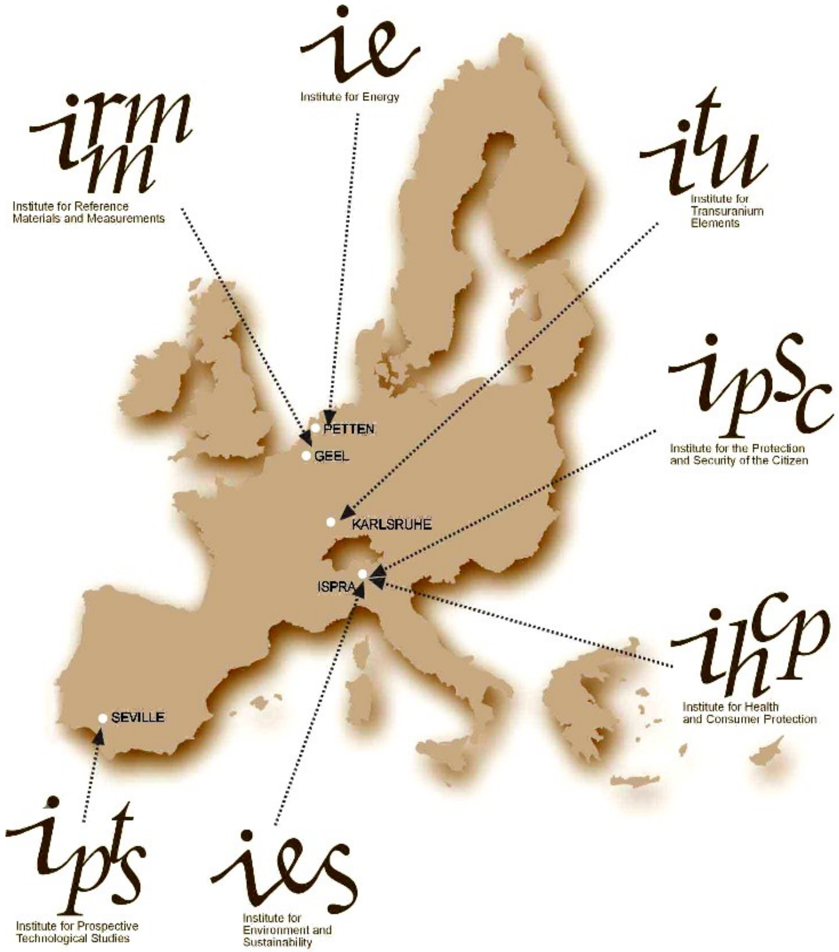
IE - Petten The Netherlands
-*Institute for Energy*

IRMM - Geel Belgium
- *Institute for Reference Materials and Measurements*

ITU - Karlsruhe Germany
- *Institute for Transuranium Elements*

IPSC - IHCP - IES - Ispra Italy
- *Institute for the Protection and Security of the Citizen*
- *Institute for Health and Consumer Protection*
- *Institute for Environment and Sustainability*

IPTS - Seville Spain
- *Institute for Prospective Technological Studies*



2. Challenges and opportunities in nuclear science and measurements

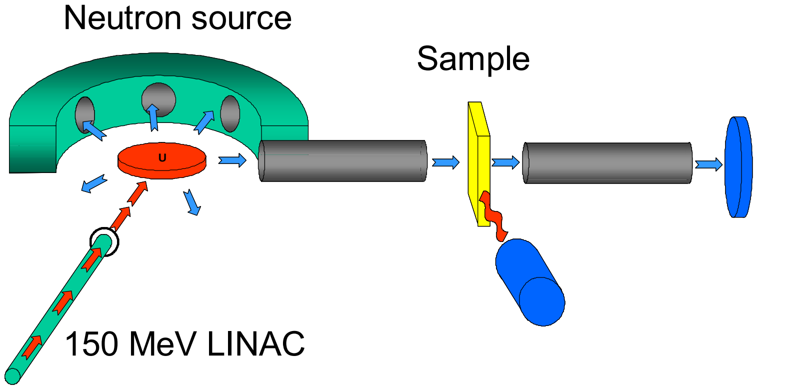
2.1 Basic Nuclear Physics

2.2 Basic Materials Science

2.3 Examples of nuclear measurements for different applications

Facilities for nuclear data measurements

GELINA:
neutron time-of-flight facility
very high energy resolution
energy range 10 meV – 20 MeV



total-absorption	σ_{abs}
capture	(n,γ)
inelastic scattering	(n,n') , (n,n'g)
neutron emission	(n,2n) , (n,2ng)
light charged-particles	(n,a) , (n,p)
fission	(n,f)

Cooperations: CEA (Cadarache, Saclay), CNRS-IN2P3 (Orsay, Bordeaux, IReS Strasbourg), INFN (Trieste, Bari), CIEMAT, Oak Ridge Ntl. Lab., Universities Bucharest, Delft, Gent, Sofia, Torino, Uppsala, Valencia, Vienna

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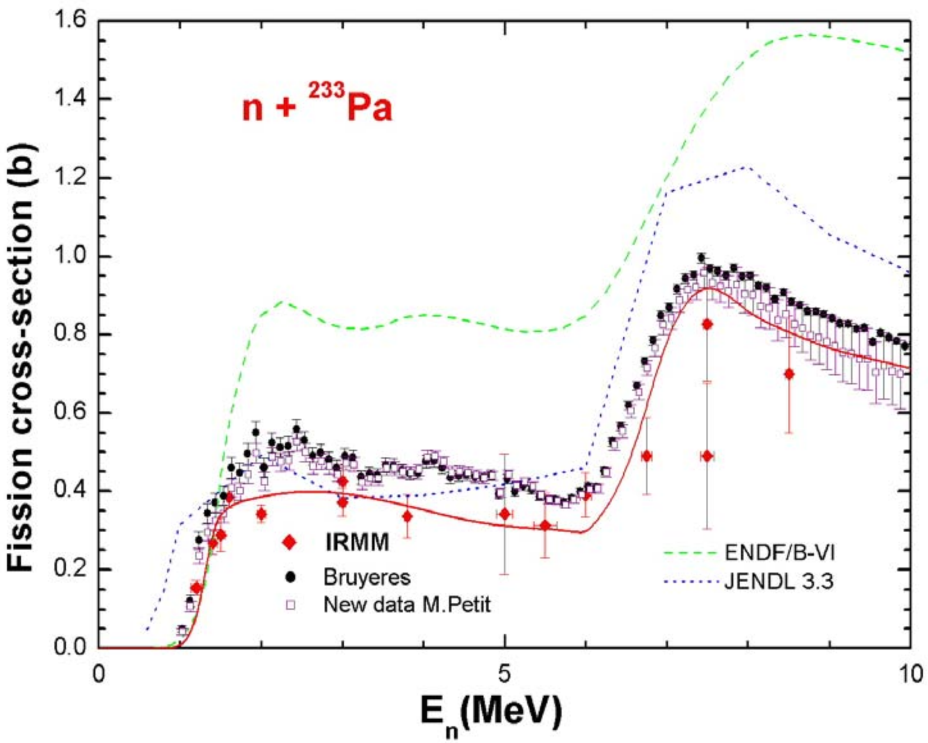
Th-U fuel cycle

first direct measurement of neutron-induced fission
of ^{233}Pa ($T_{1/2}$=27 d), collab U. Örebro

comparison of ^{233}Pa(n,f) reaction to surrogate reaction ^{232}Th(^{3}He,pf)
evaluated nuclear data files and a new calculation (red curve), collab. U. Bucharest



Data are being included
in IAEA-CRP F4.10.20
"Evaluated nuclear data
for the Th-U fuel cycle"

Phys. Rev. Lett. 88 (2002) 62502 ; Nucl. Phys. A733 (2004) 3
Phys. Rev. C69 (2004) 021604R ; Nucl. Phys. A740 (2004) 3

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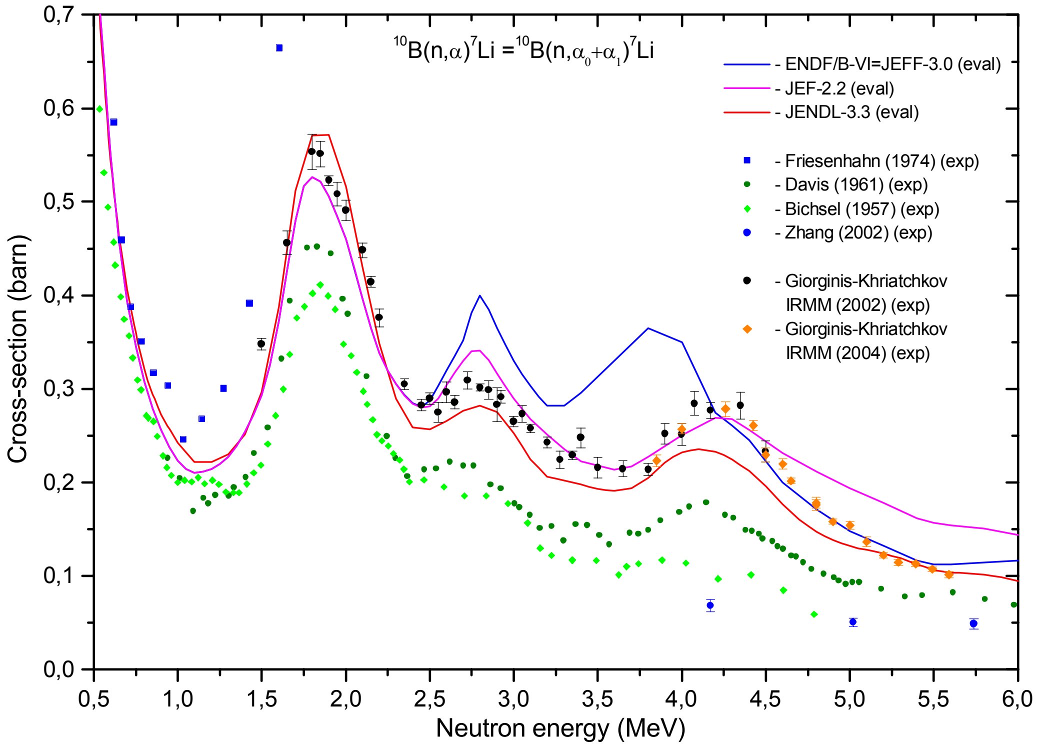
Accurate standards

$^{10}B(n,\alpha)$

comparison with previous measurements and evaluated data files
collaboration with IPPE, Obninsk



Data are being included in IAEA-CRP F4.10.19
"Improvement of the standard cross-sections for light elements"

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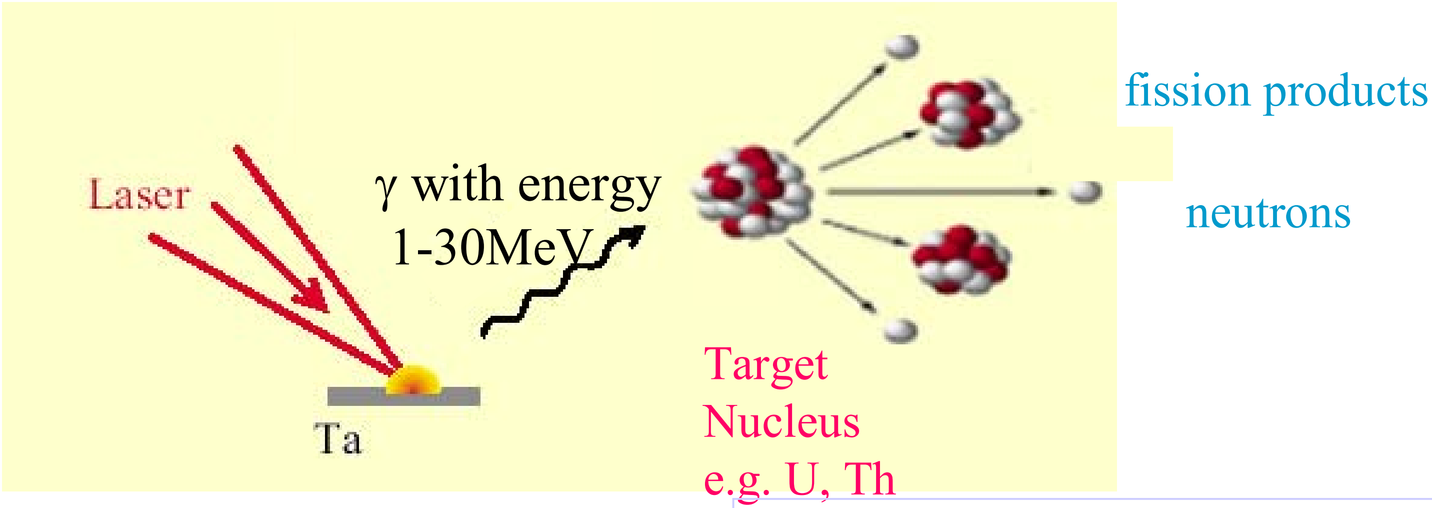
Laser induced nuclear reactions



Objectives

✓ photofission of U, Th, Np, Am, Pa, and Pu isotopes

✓ (γ, xn) reaction on long-lived fission products such as ^{99}Tc and ^{135}Cs, ^{129} I

Experiments performed

✓laser induced-photofission of ^{238}U and ^{232}Th. Photo-transmutation of ^{129}I.

(Appl. Phys. B. (2003) J. Magill et al. « Laser transmutation of ^{129}I»)

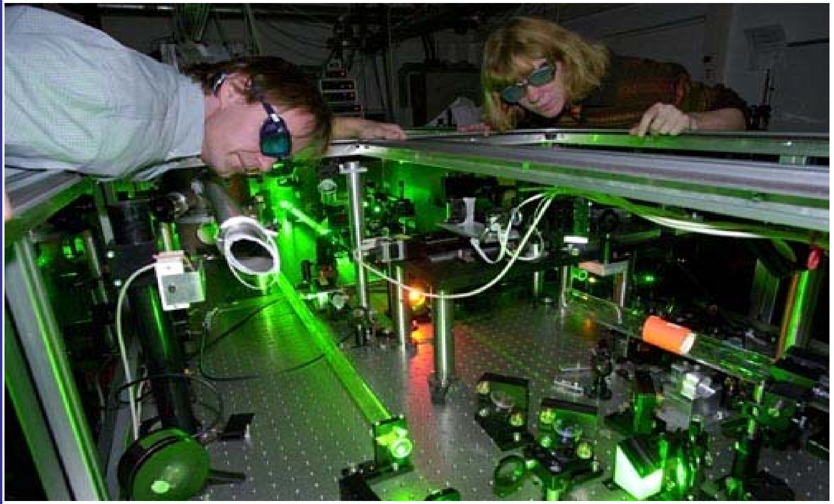
(J. Phys. D: Appl. Phys. **36** (2003) L79–L82. K.W.D. Ledingham et al. « Laser-driven photo-transmutation of ^{129}I - a long-lived nuclear waste product»)

(Europhys. Lett. 61 47 (2003) H. Schwörer et al. « Fission of actinides using a tabletop laser »)

✓first trials on ^{99}Tc

The Lasers

David vs. Goliath…..



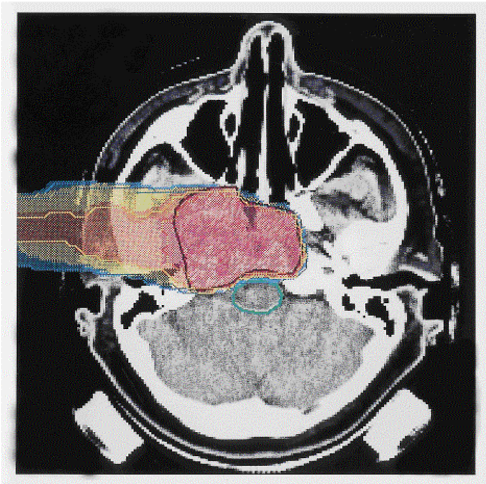
Tabletop laser at Jena

- 15 TW Ti:sapphire laser
- energy on target about 0.5 J, within a pulse duration of 80fs
- High repetition rate 10 Hz
- 10^{20} W.cm^{-2}

VULCAN giant pulse laser at Rutherford lab.

- Glass laser system operating at over 100 TW
- can deliver pulses with energy on target up to 100 J with pulse length about 1ps
- Repetition rate: one shot every 20 minutes
- 10^{20} W.cm^{-2}

Potential Medical Applications:

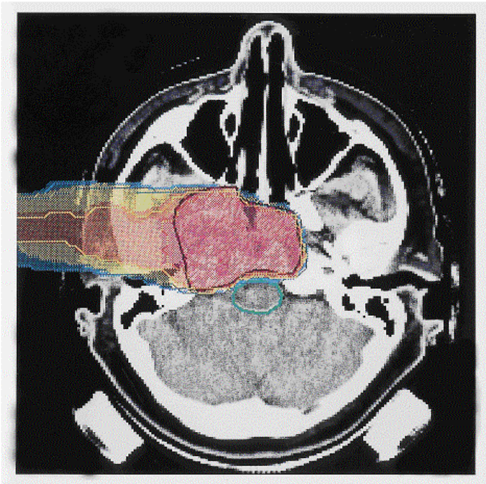


GSI heavy ion radiotherapy. The Tumour situated in the centre of the brain is treated directly by depositing the energy in this region

Ne19 17,3 s	Ne20 stable 90,48%	Ne21 stable 0,27%
F 18 1,83 h	F 19 stable 100%	F 20 11,16 s
O 17 stable 0,038%	O 18 stable 0,2%	O 19 26,46 s

Potential Medical Applications:

- Ion beam Therapy: Lead ions (430 MeV), C ions (80 MeV), protons (40 MeV) have been produced.



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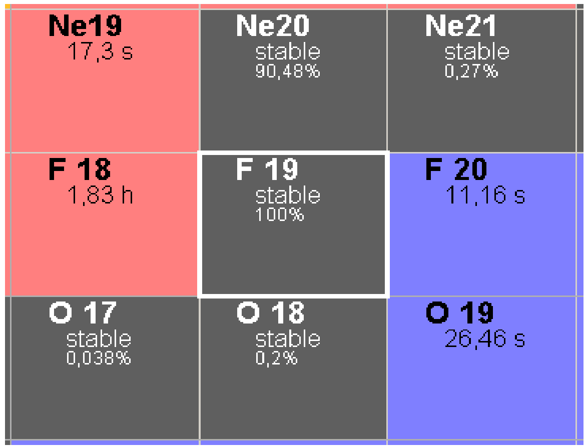

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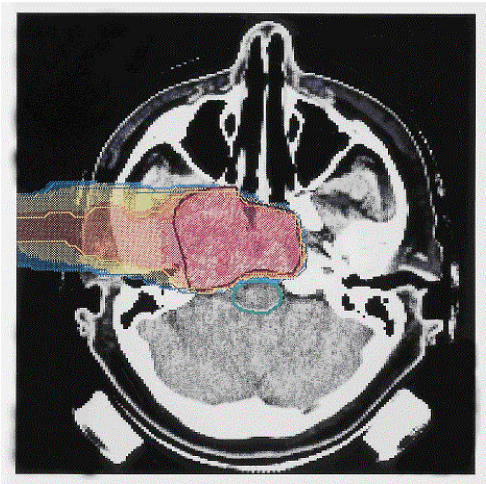

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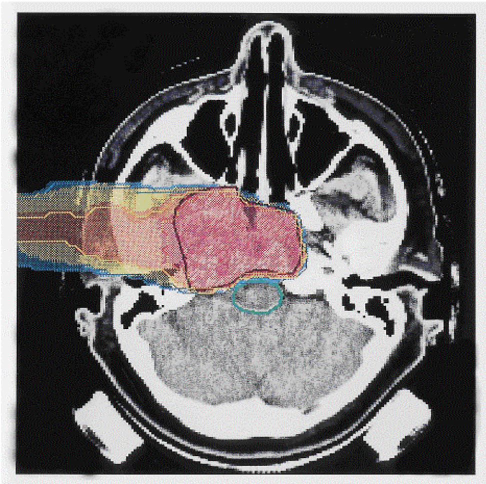

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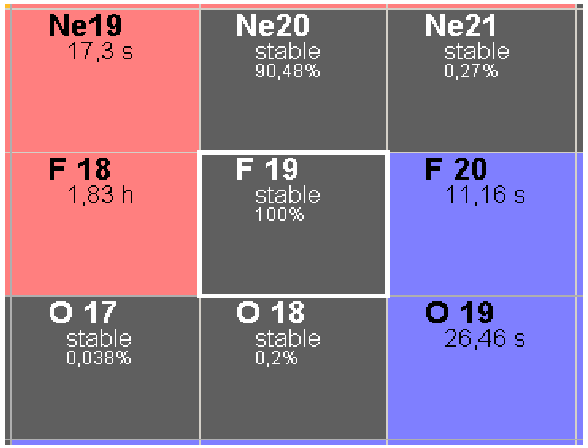

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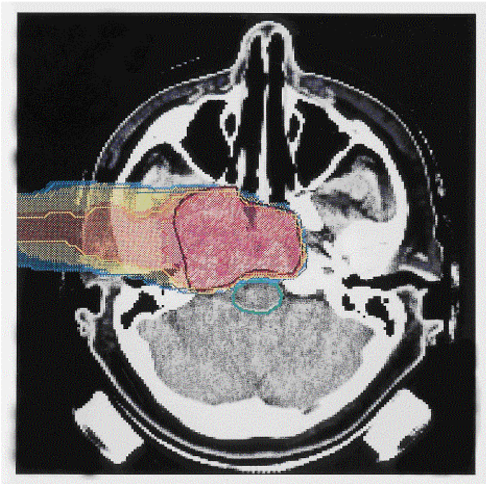

GSI heavy ion radiotherapy. The Tumour situated in the centre of the brain is treated directly by depositing the energy in this region

- Production of short-lived isotopes for PET (^{18}F) (need 1 kHz repetition rate and 1 J per pulse =>10^9 Bq)

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2. Challenges and opportunities in nuclear science and measurements

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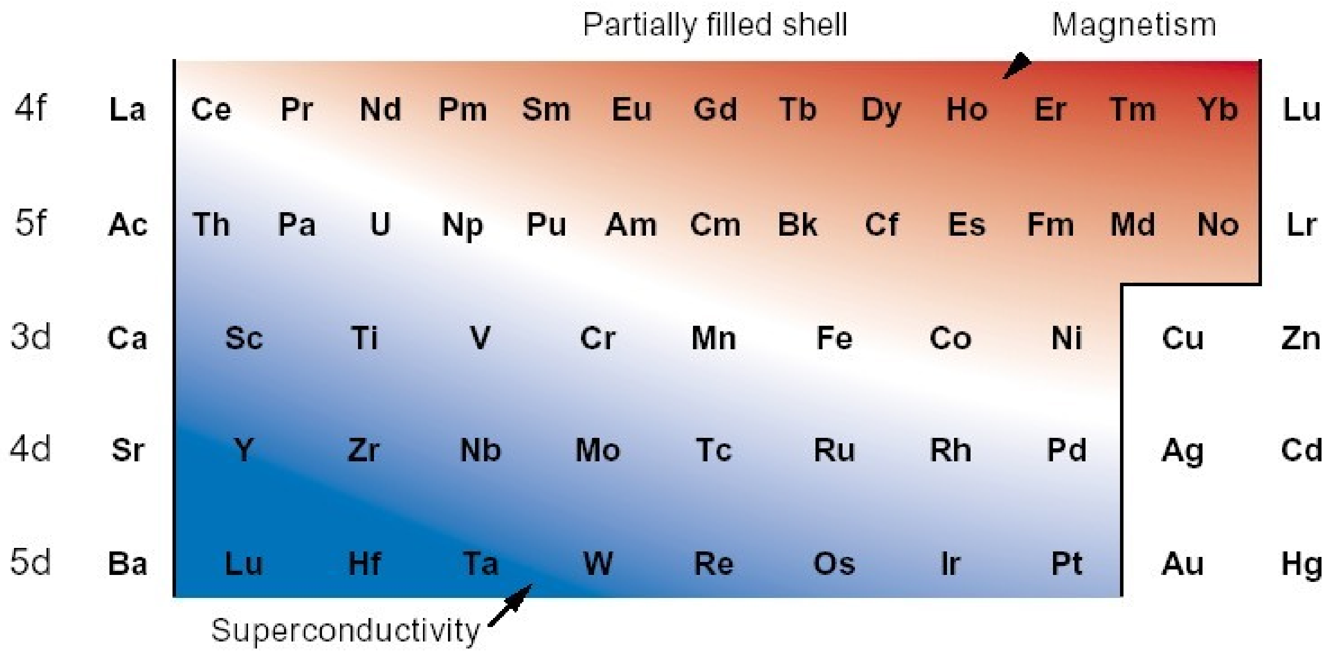
2.3 Examples of nuclear measurements for different applications

Magnetism versus superconductivity in groups of elements with partially-filled d- and f-shells

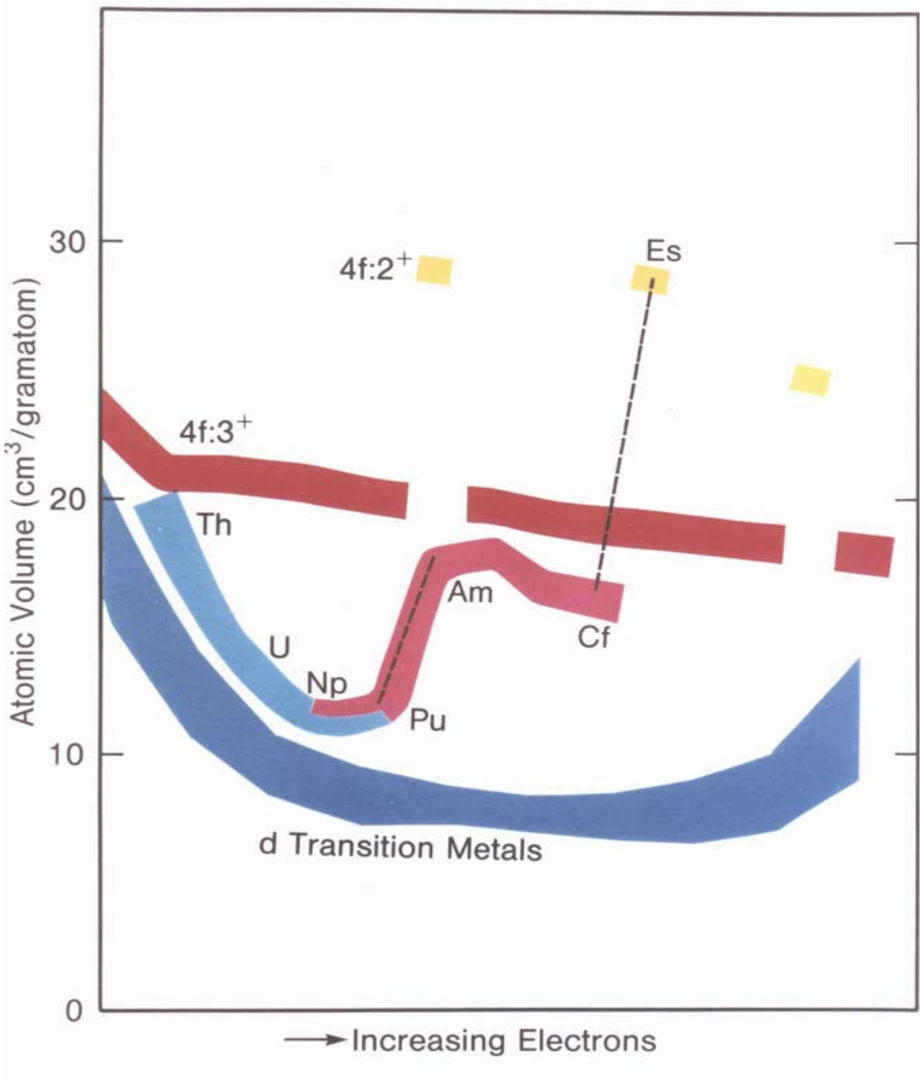


Courtesy: Smith, LANL

Atomic volume of *d* and *f* series

- **Notice how light actinides are smaller than rare-earths (4*f*) and follow a curve like the *d* series.**

- **The heavier actinides are more "rare-earth" like.**

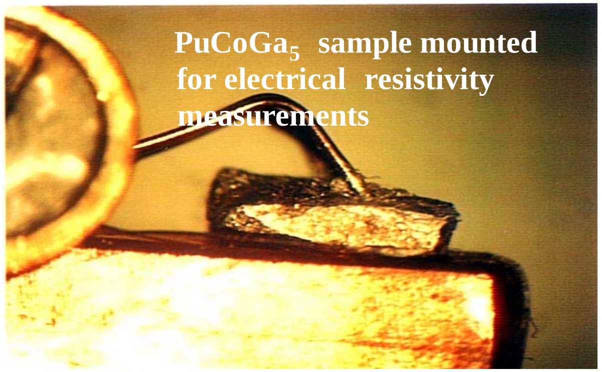


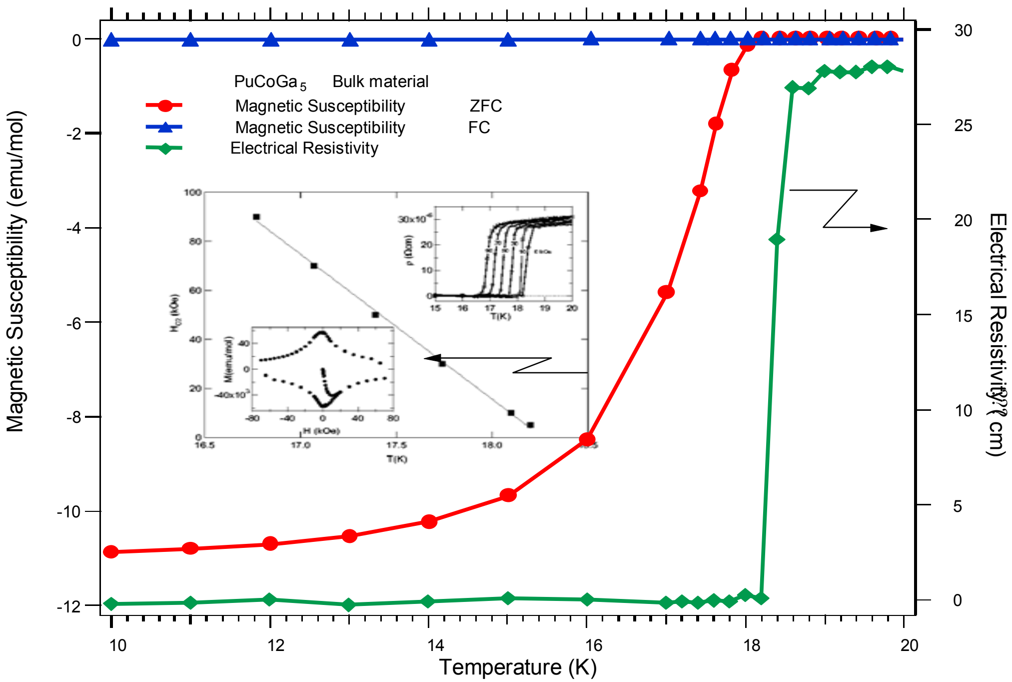
Discovery of Pu-based superconductivity above 18K in $PuCoGa_5$

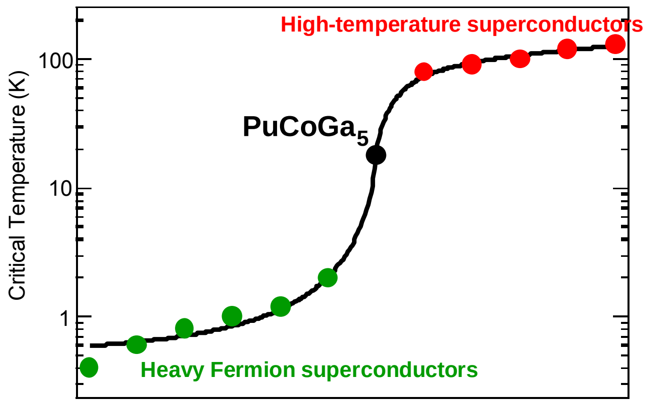
Sarrao et al., Nature 420 (2002) 297

Why Pu ?

- Plutonium is at a crossover in electronic properties
- Question: Is the superconductivity in this system magnetically mediated?







$PuCoGa_5$ as the missing step between magnetically mediated/heavy- fermion superconductors and high-temperature copper-oxide superconductors ?

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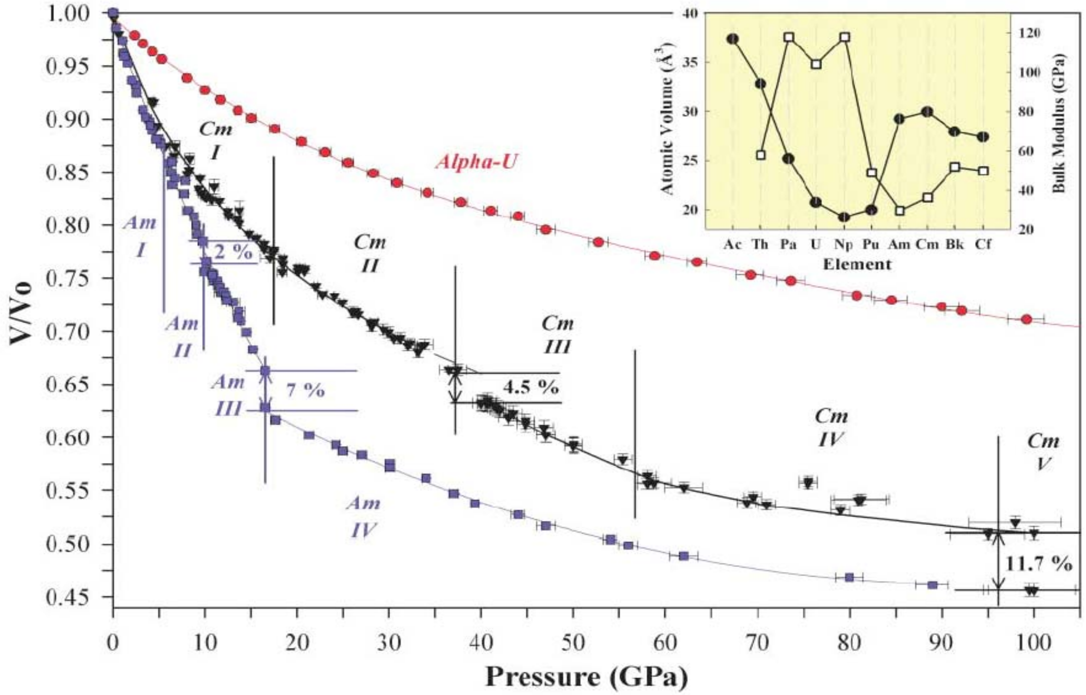
Compressibility of actinide elements – new Cm (III) phase

- *Note major difference between uranium and Cm and Am, especially at low P*
- *Experiments done on a few μg at world's most advanced synchrotron in Grenoble, France*
- *it was demonstrated that this new phase (Cm (III)) is stabilized by magnetism, as has been found previously in only two other cases (Fe, Co)*



S. Heathman *et al.* Science 309 (2005) 110

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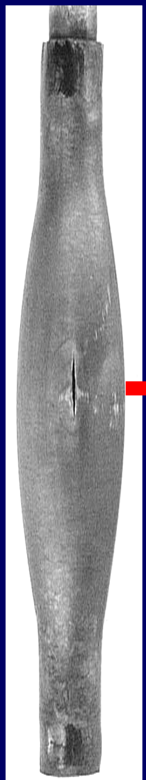
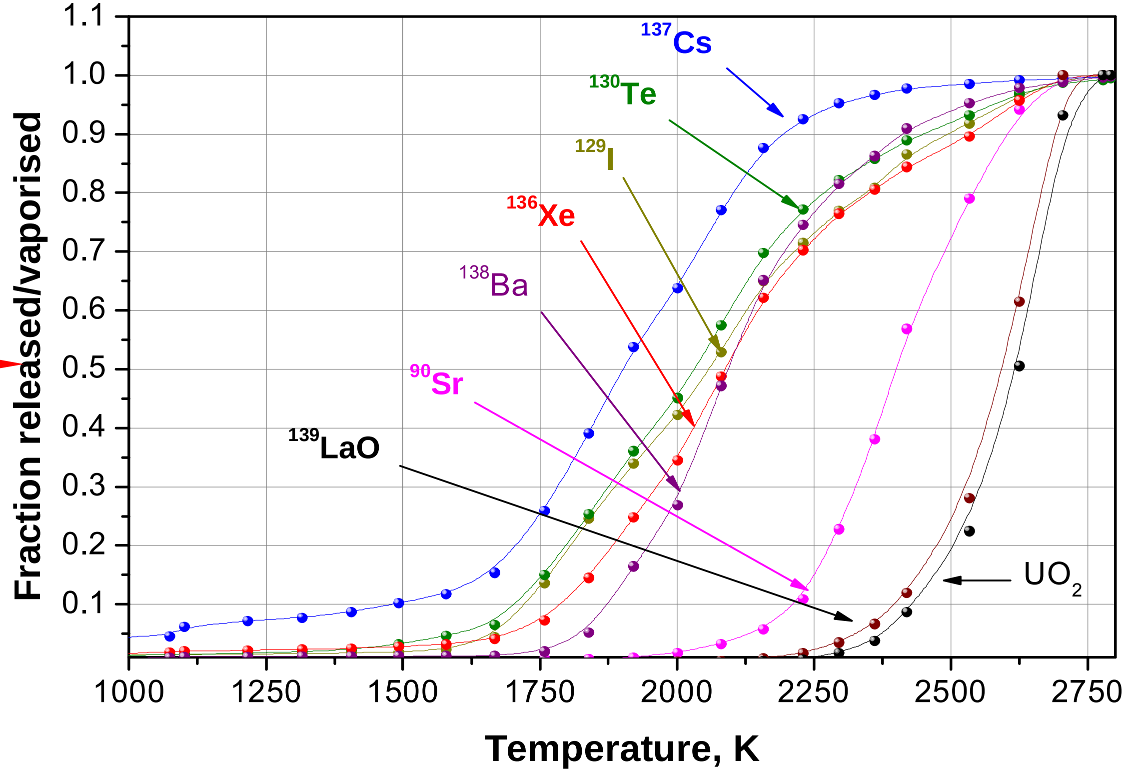
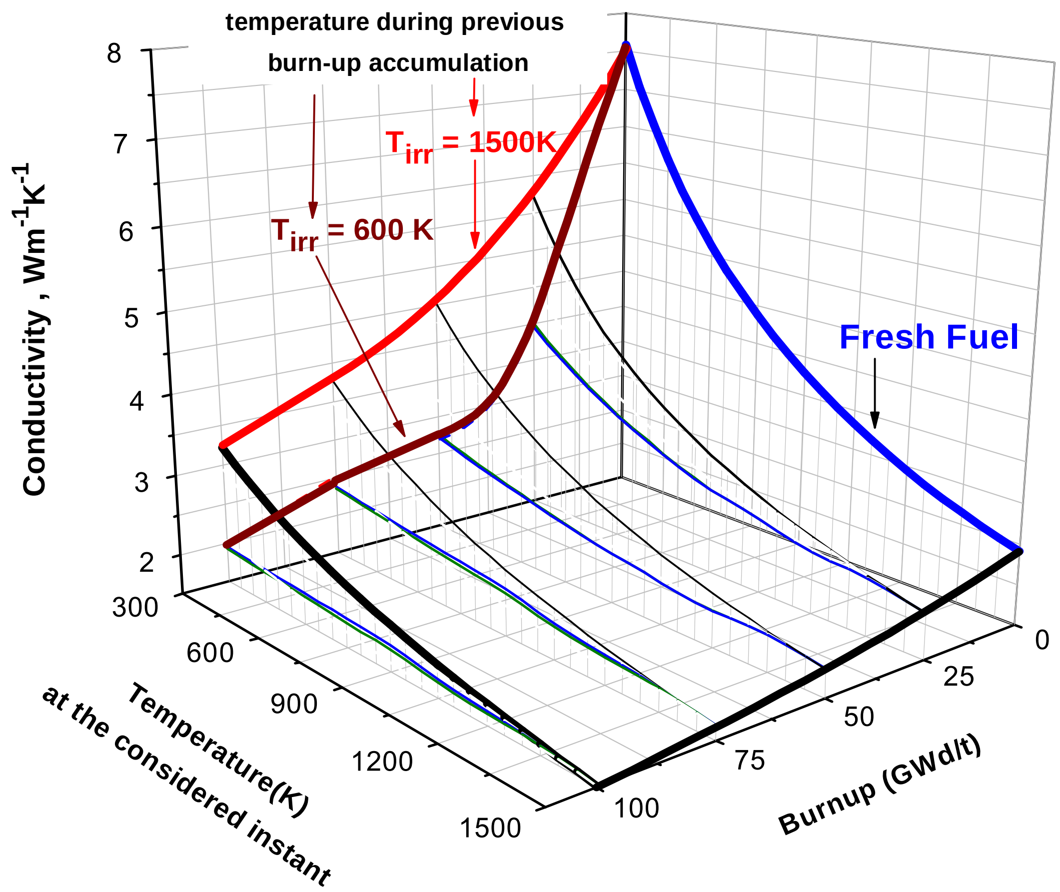
Fission products & uranium vapour release from irradiated UO_2 (65 GWd/t) during an accident





THERMAL CONDUCTIVITY OF IRRADIATED UO_2 AS A FUNCTION OF BURNUP AND TEMPERATURE

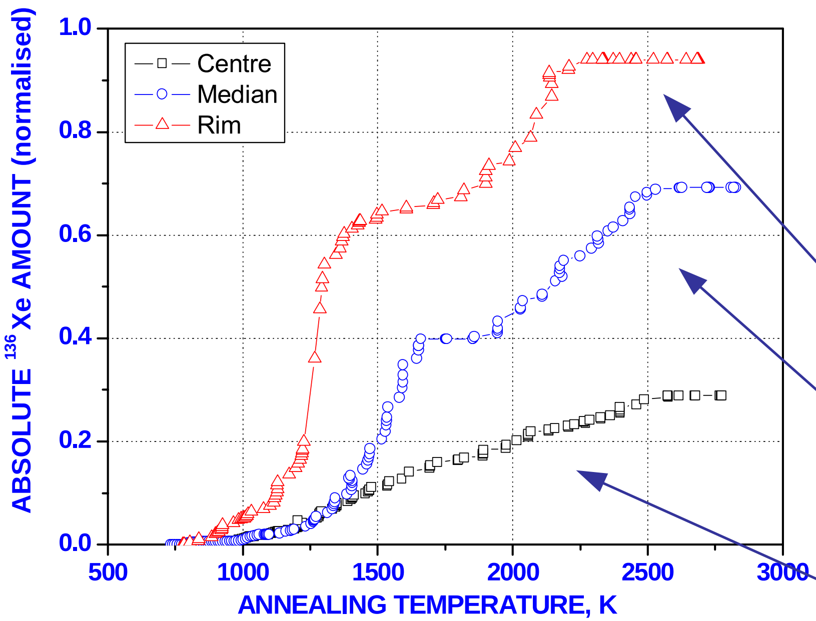


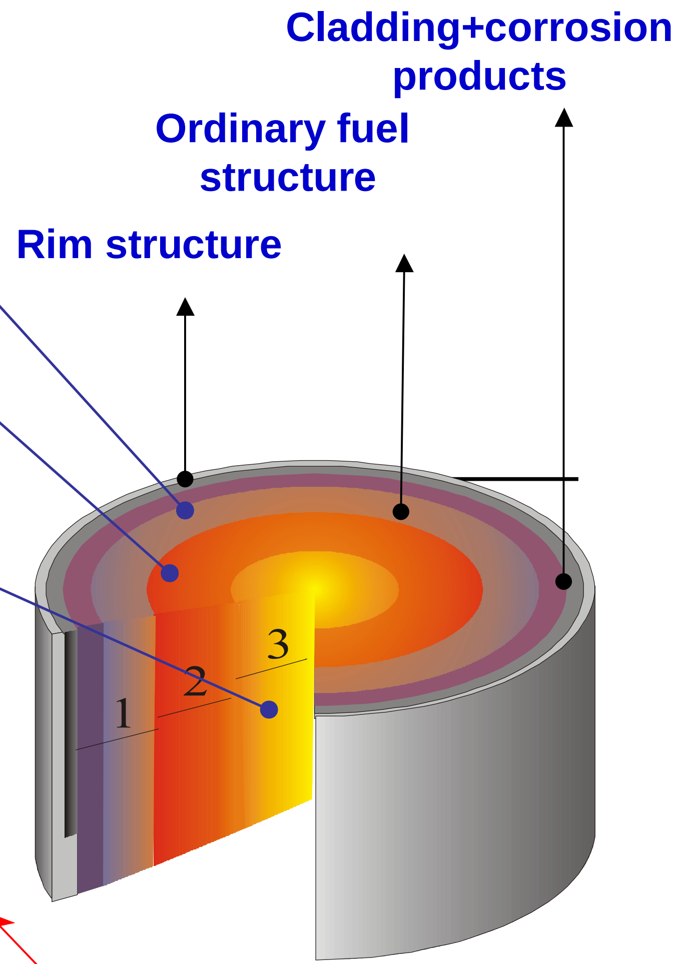
The fuel thermal conductivity dramatically decreases with burnup.

The effect is stronger if the fuel is irradiated at low temperature.

Most of this damage effect is permanent and only a small part of it is recovered by thermal annealing

Fission gas release effects during a fuel T-transient





Sample position	Burn-up (GWd/t)	~Irrad. Temp. (^{0}C)	ΔH_{diff} (eV/atom)
Periphery	88	~500	2.82 ± 0.2
Median	72	~700-1000	3.74 ± 0.3
Center	70	1000~1300	4.00 ± 0.4

Atomic migration energy

Safety of Nuclear Fuel

mechanical examination of irradiated fuel

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Safety of Nuclear Fuel

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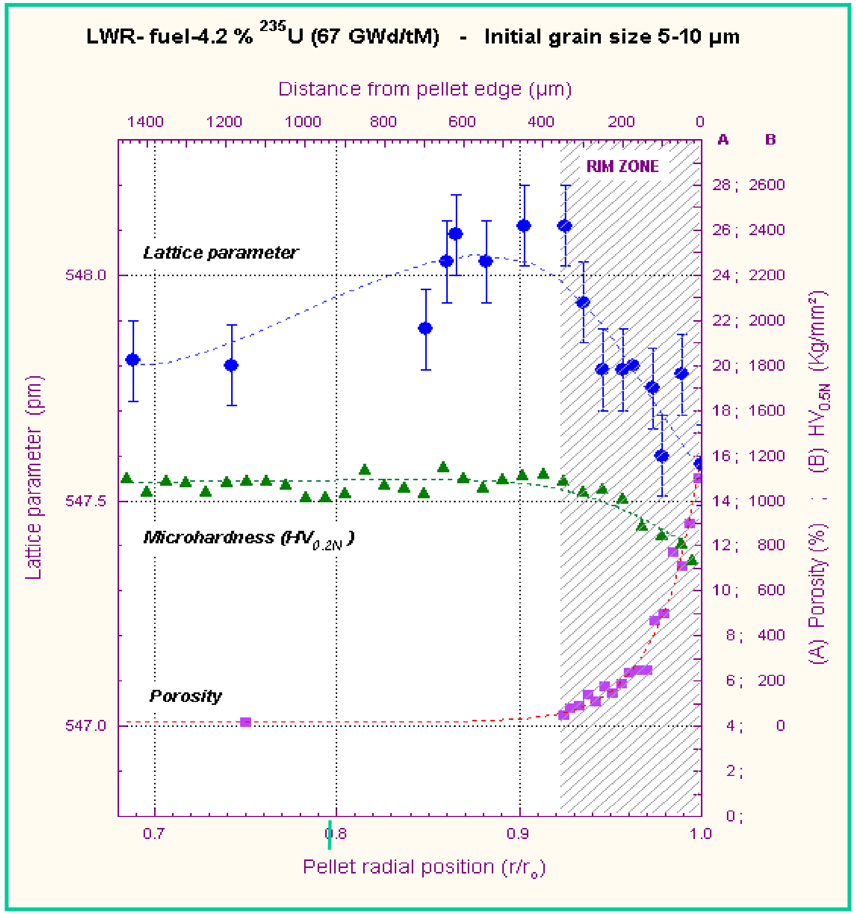

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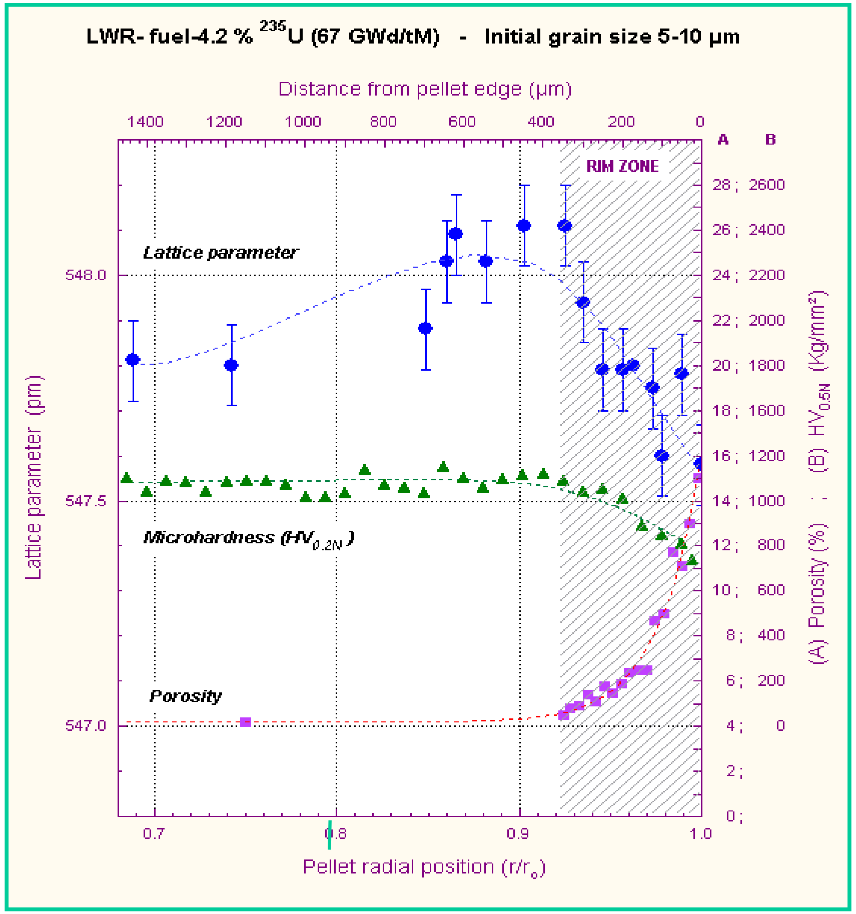

Microstructure, micro-hardness and lattice parameter variations at the fuel periphery (rim zone)

LWR-fuel (67 GWd/tM) - Initial enrichment: 4.2 % ^{235}U

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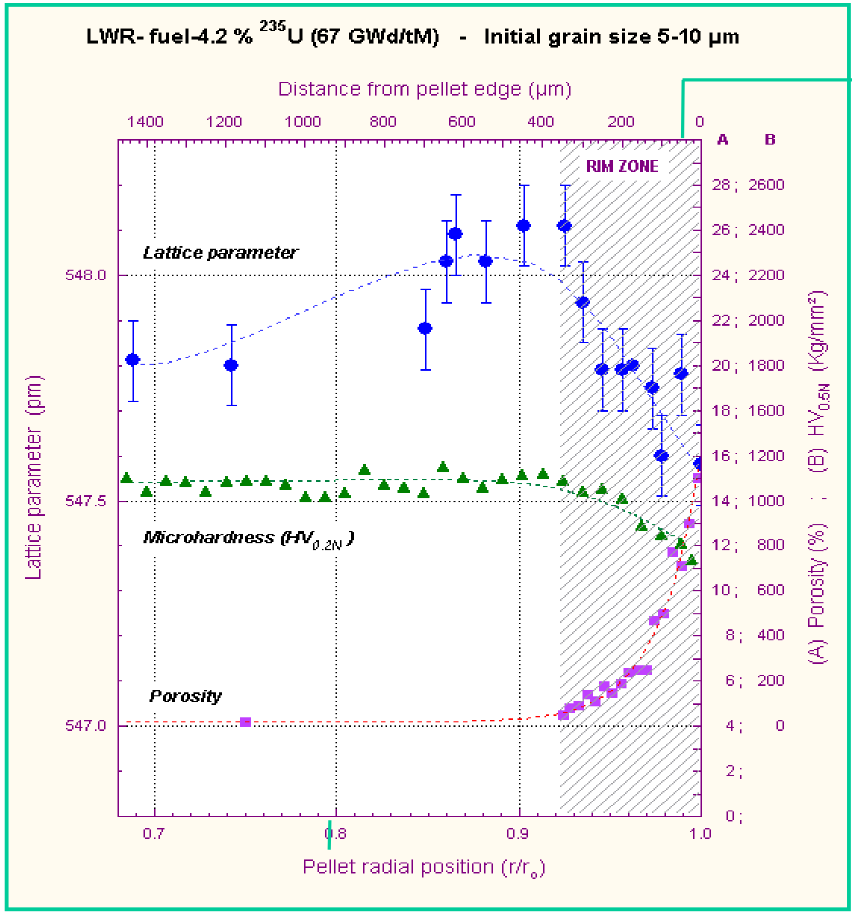
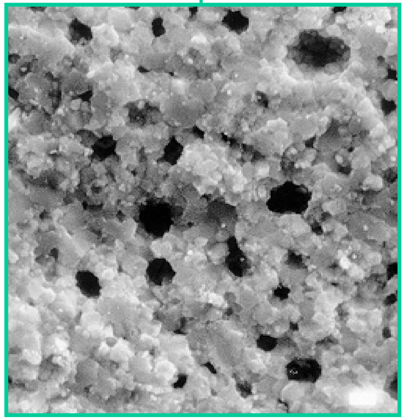
Safety of Nuclear Fuel

mechanical examination of irradiated fuel





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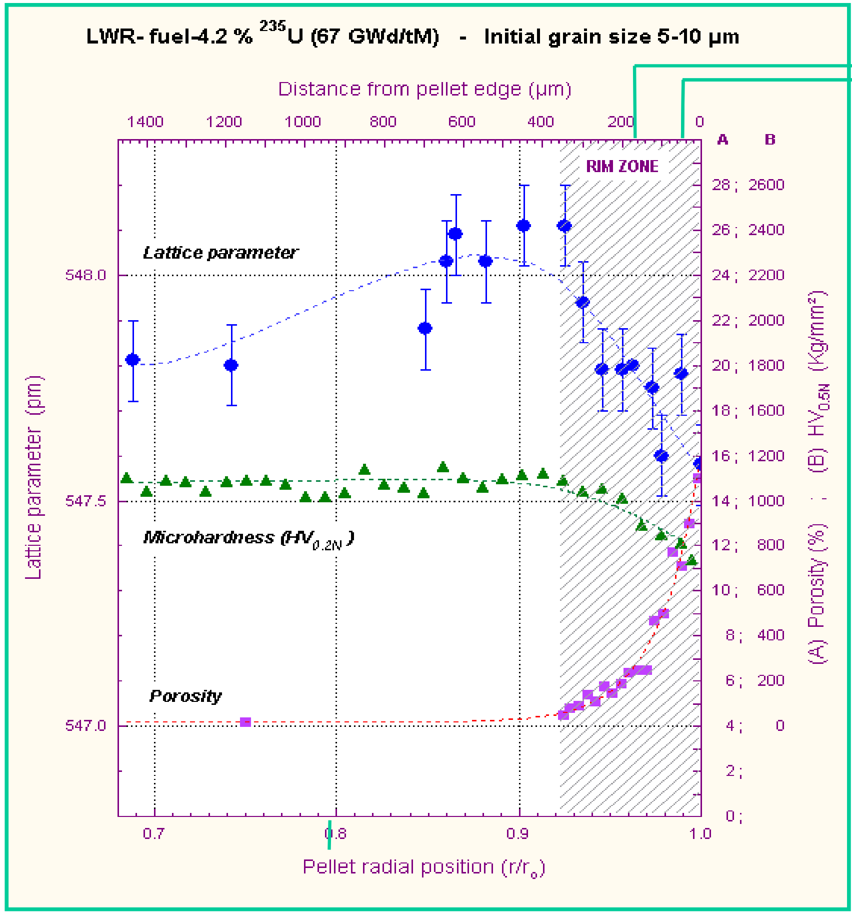

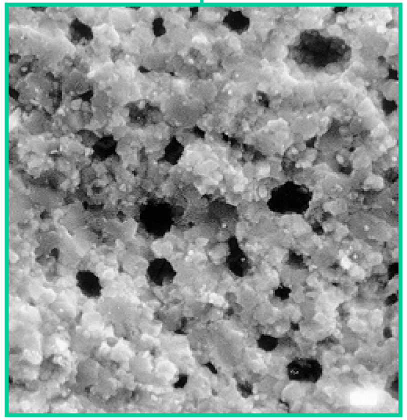

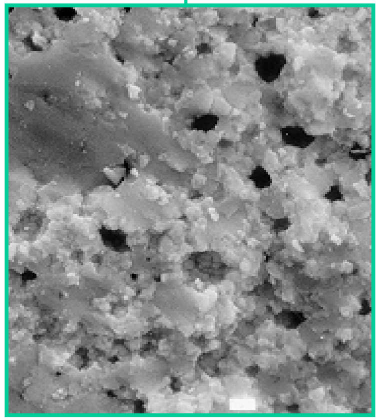

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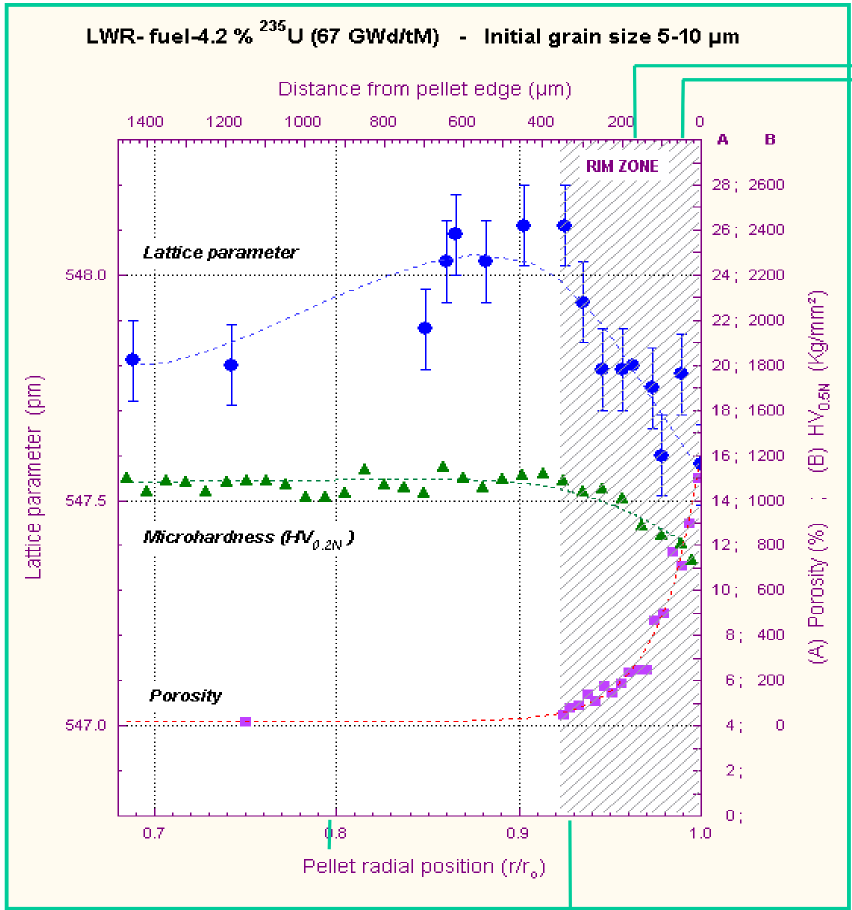

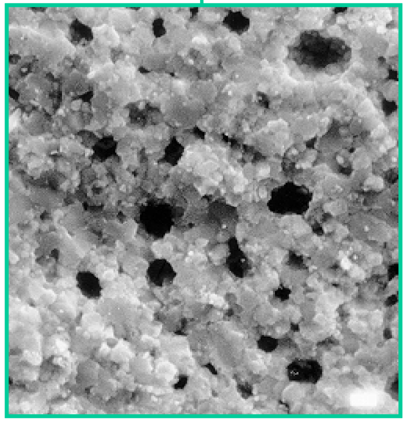

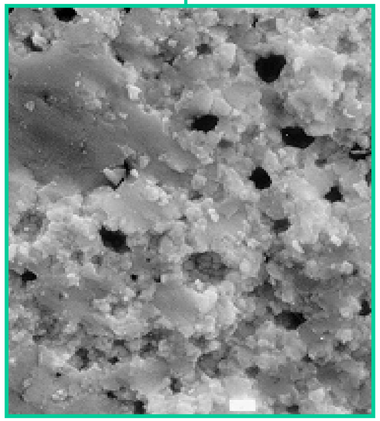

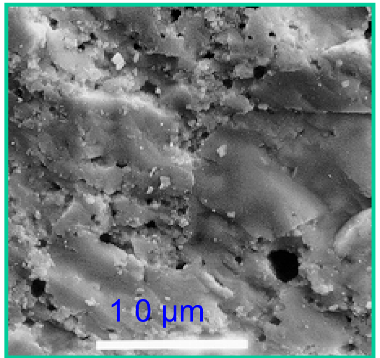

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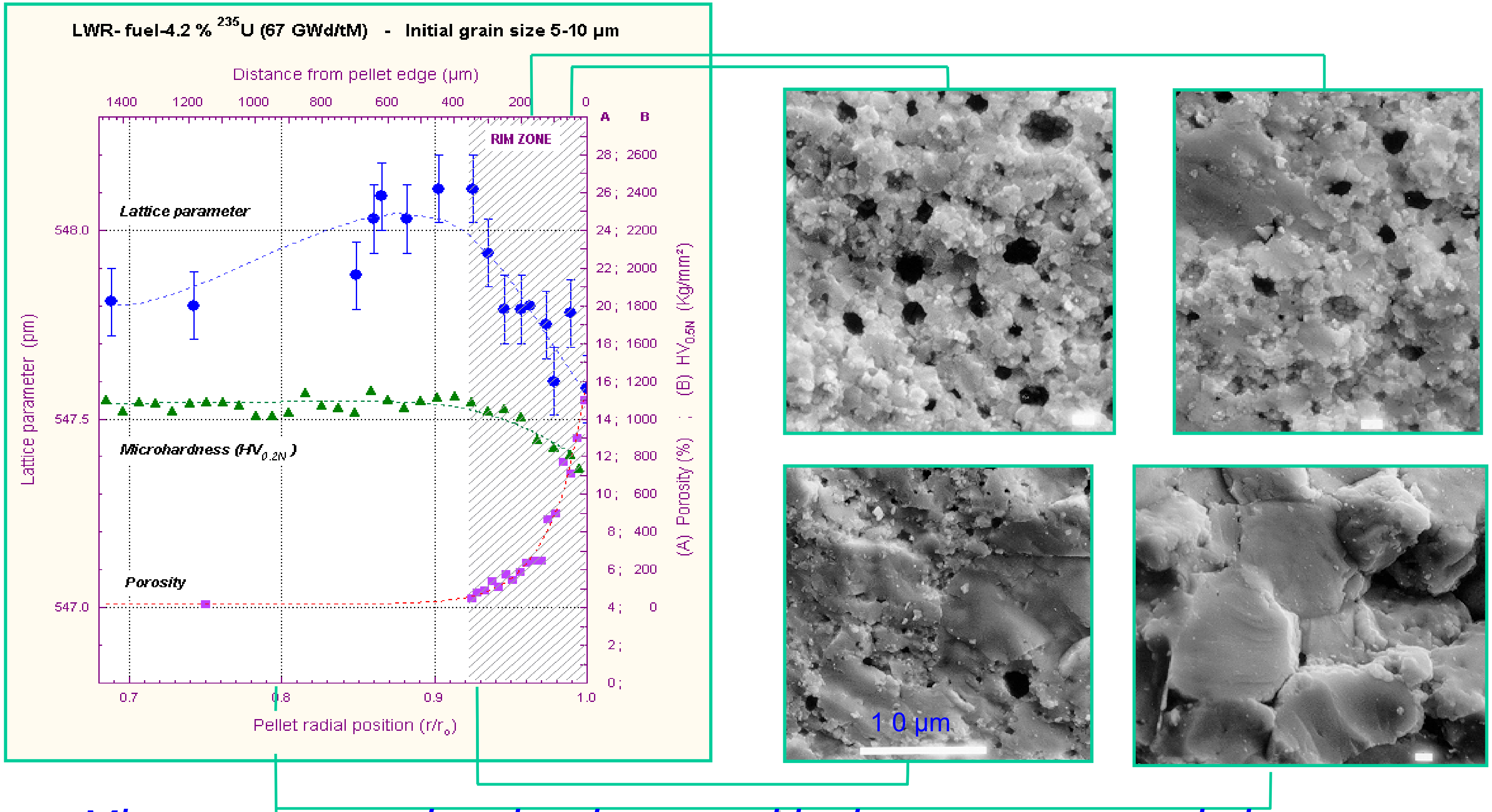

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2. Challenges and opportunities in nuclear science and measurements

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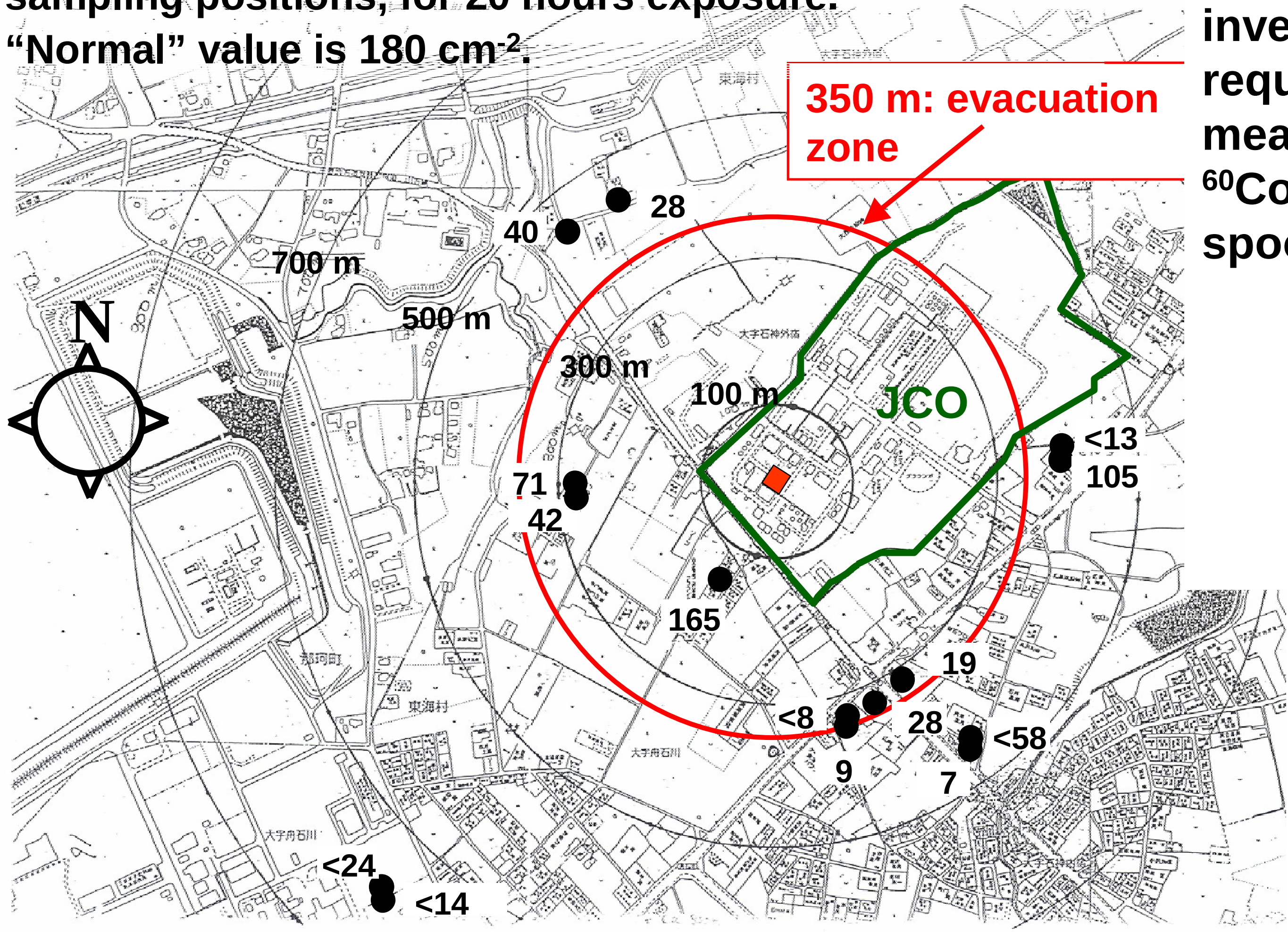
The criticality accident at the JCO nuclear fuel plant in Tokai-mura, 1999

Thermal neutrons fluence values (10^6 cm^{-2}) at the sampling positions, for 20 hours exposure. "Normal" value is 180 cm^{-2}.

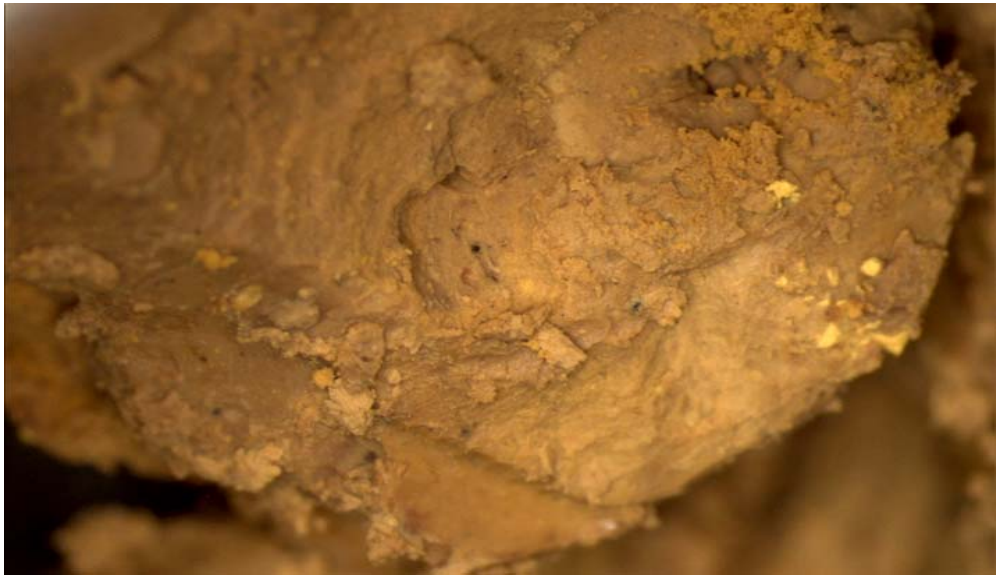


- **The Japanese investigation team requested support with measurements of ^{51}Cr, ^{60}Co and ^{59}Fe in table spoons.**

Case Study

Illicit Trafficking - Case Study : **Find 26 – Rotterdam**

On 16th Dec. 2003- 2-3 kg radioactive material was detected in a scrap metal shipment in Rotterdam harbour. The shipment arrived from a dealer in Jordan. Materials sent to JRC/ITU on 10th March 2004 and consisted of 2 bulk samples and 3 swipes.



Results - bulk

- **Gamma spectrometry**
  - U-235 enrichment 0,7 %
- **TIMS and MC-ICP-MS**
  - U-234 = 0,0052 %
  - U-235 = 0,712 %
  - U-238 = 99,283 %

  ⇒ **natural uranium, no indication of an enriched or irradiated uranium**
- **Titration**
  - U-content ~70 %

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Titration

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Lead isotopics

- Pb-204 = 1.47 %
- Pb-206 = 24.83 %
- Pb-207 = 21.47 %
- Pb-208 = 52.24 %

⇒ **resembles natural lead, no radiogenic lead**

Anions (qualitative)

- $(NO_3)^-$ and $(CO_3)^{2-}$

ICP-MS

- Main impurities (>1000 ppm): Al, Ca, Cr, Fe, Mg, Mo, Na, Ni, P

Find-26: Rotterdam

Find-26: Rotterdam

Bulk material:

- natural uranium oxides with ≈ 70% U
- presence of $(CO_3)^{2-}$
- main impurities: Al, Ca, Cr, Fe, Mg, Mo, Na, Ni, **P**
- lead isotopic composition

Swipes:

- natural uranium, Cs-137, Eu-154 and Am-241 (evidence of nuclear activities)

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What information did the analysis yield?

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Swipes:

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What information did the analysis yield?

- Intermediate product, possibly $(NH_4)_4(UO_2)(CO_3)_3$
- Impurities point to phosphate rich ores (North Africa, Middle East, USA, South Africa, Brazil)
- Pb isotopic composition (natural) indicates low uranium content in the ore, which is the case for P-rich ores
- evidence of nuclear activities
- corroborated intelligence information on the source of the material

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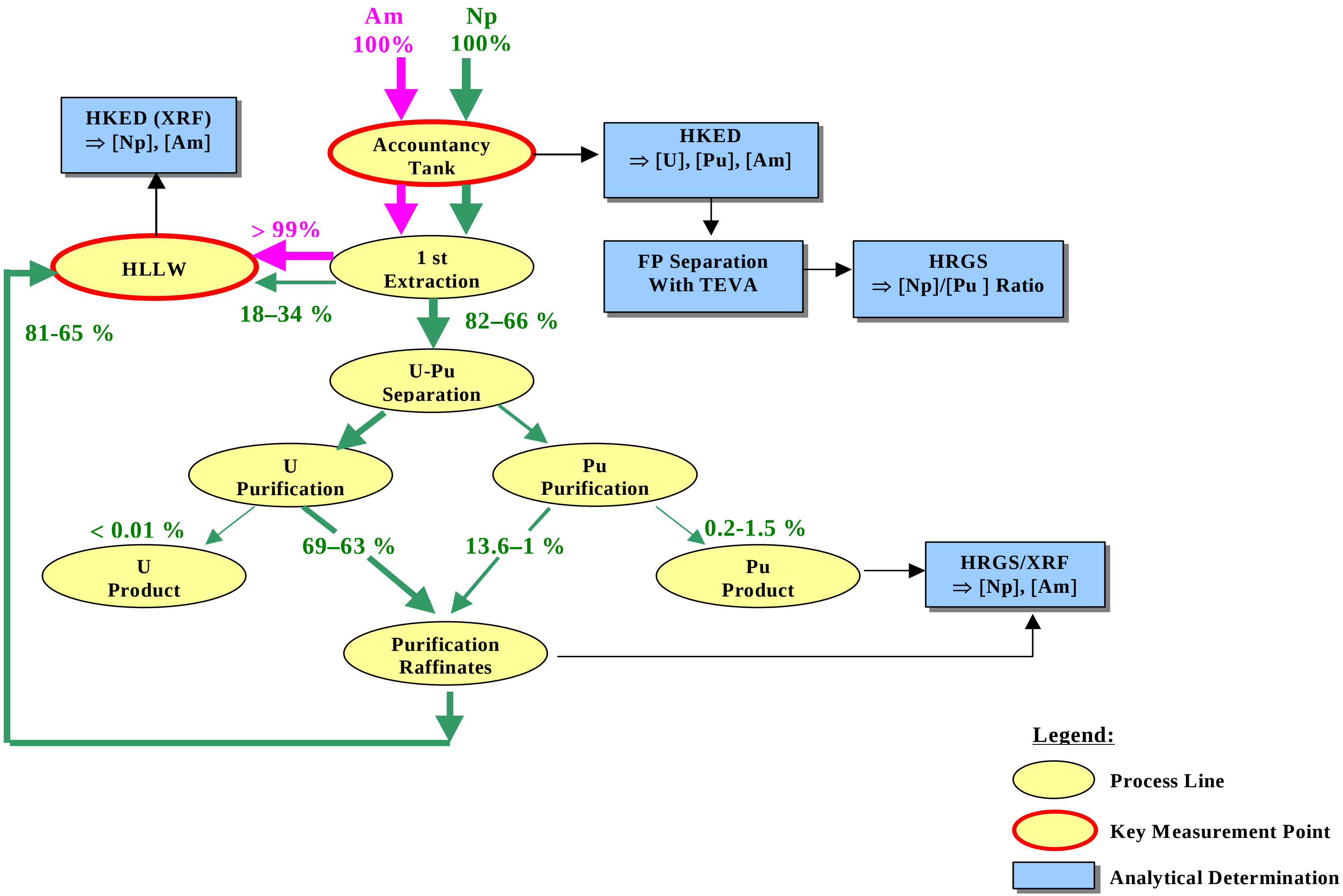
Neptunium Flow-Sheet Verification



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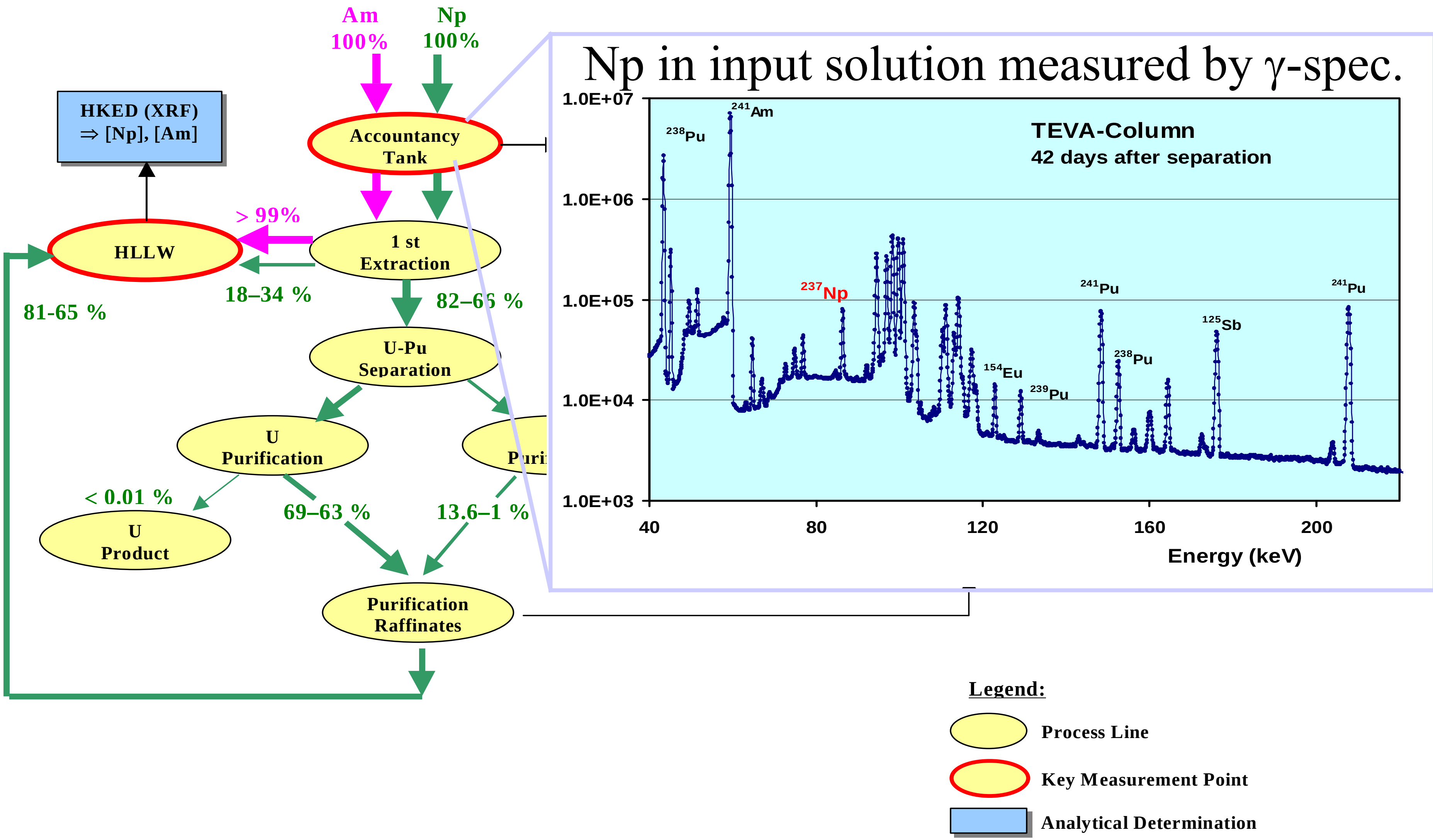
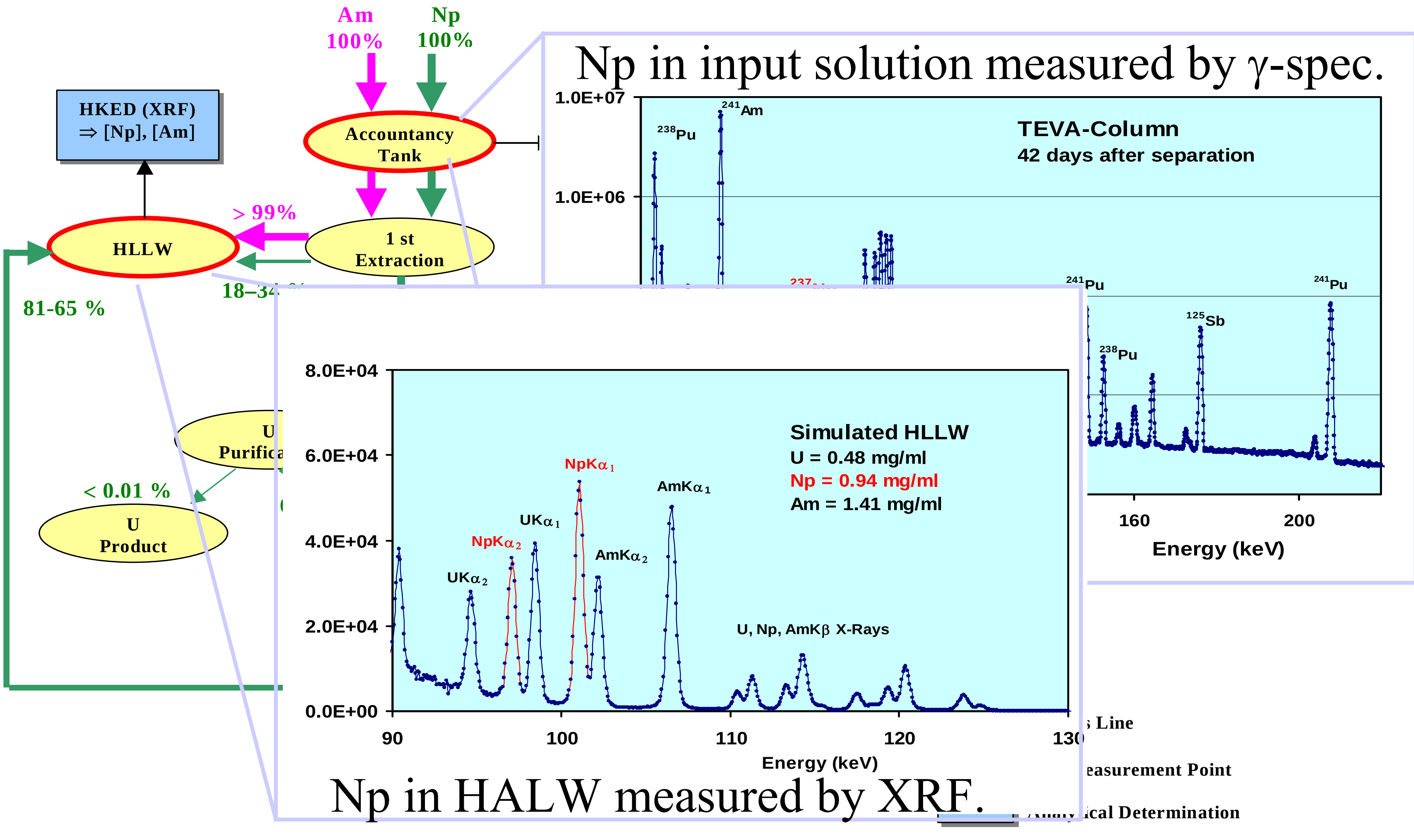


Neptunium Flow-Sheet Verification



Strengthened Safeguards: Particle Analysis

- Application of the method for Routine Safeguards High Performance Trace Analysis for the detection of U and Pu particles

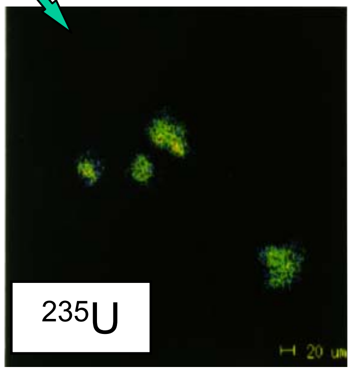
Find a Needle in the Hay Stack

"swipe" sample: billions of dust particles
with
low enriched uranium particles

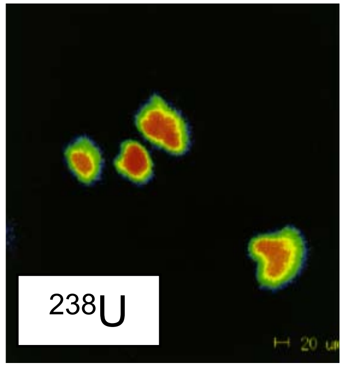
Natural U,
0.72 wt % of 235

Nuclear fuel,
0.72-5 wt% of 235

Weapons grade U,
20-90 wt % of 235



U-235 SIMS image



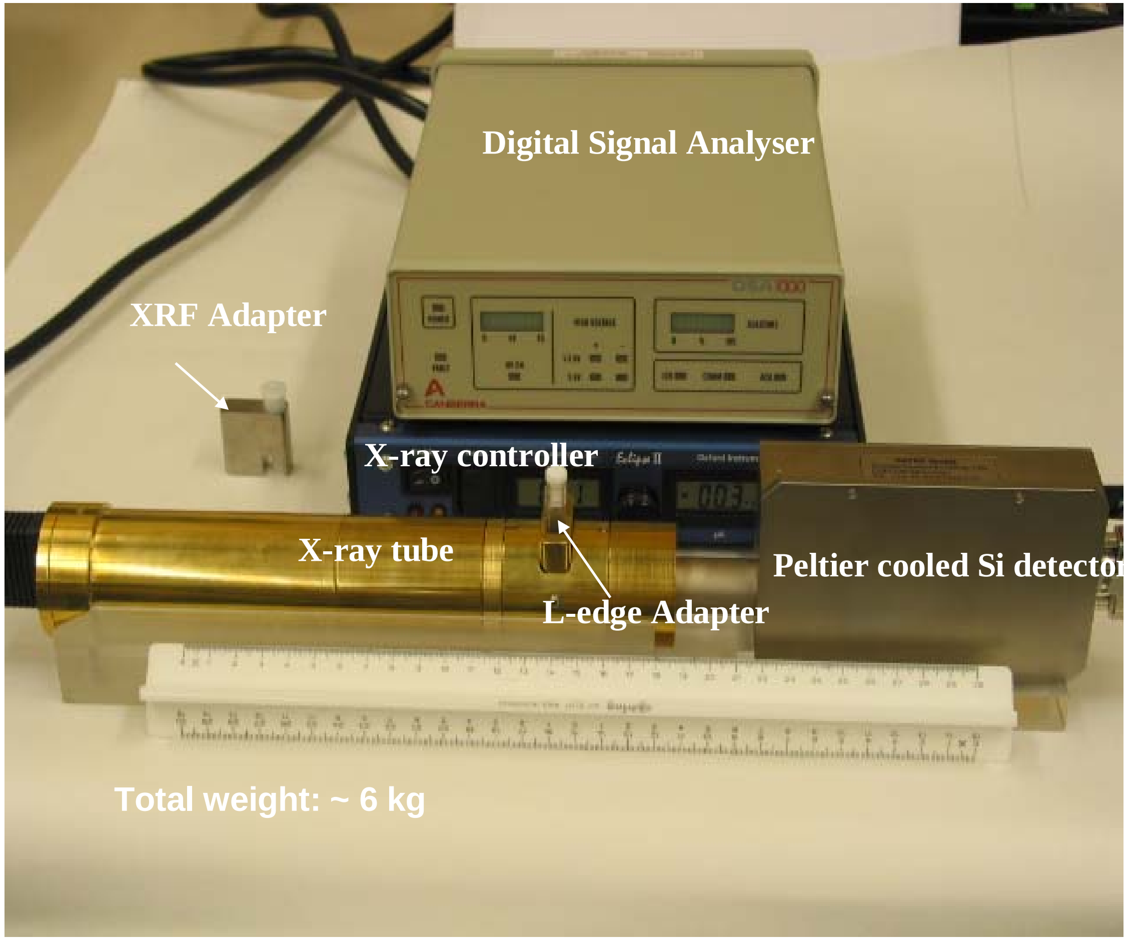
U-238 SIMS image

COMPUCEA 2nd generation : on-site U-enrichment measurements

(Under IAEA SP Task A1507)

Basic hardware components

- Mini X-ray system, 30 kV/100 µA (Amptek, Eclipse II)
- Si drift detector, Peltier-cooled, with integrated electronics (KETEK)
- Digital signal processor (Canberra DSA 1000)



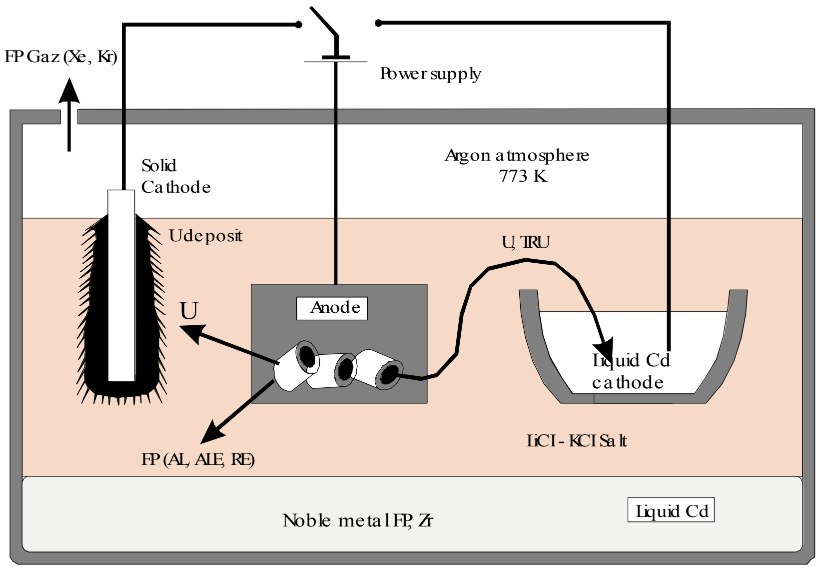
Control of Actinides in a Pyrochemical Partitioning Process

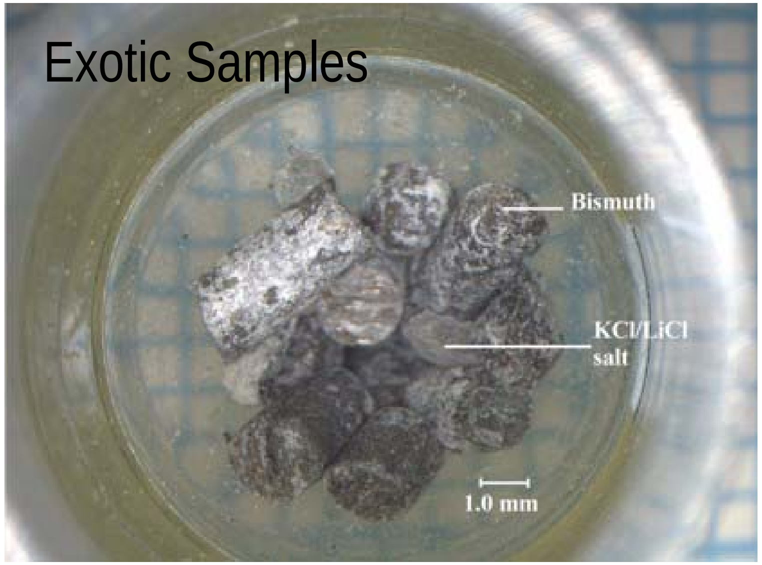
Quantitative analytical methods are required in order to establish a material balance:

1) for process development

and –at a later stage–

2) for accountancy and control purposes





Application of radiometric techniques for Minor Actinides

Technique	Element/ isotope measured	Isotope contribution to response*	Minimum amount for assay	Application
K-XRF	Np Am Cm	- - -	50 µg 70 µg 100 µg	Any sample type in liquid form mass fractions of analyte ≥ 0.02 %.
NCC	Cm	^{244}Cm. 90-95% ^{246}Cm: 5-10%	200 ng	For any type of Cm-containing samples (liquid or solid) with Pu/Cm ratios ≤ 1000
HRGS	^{237}Np ^{241}Am ^{243}Am	- - -	500 µg 10 ng 100 ng	Liquid samples for absolute measurements. Low FP content for ^{237}Np assay.
Calorimetry	Am Cm	^{241}Am: 98% ^{243}Am: 2% ^{244}Cm: 99% ^{243}Cm: 1%	5 mg** 200 µg**	Refractory MA fuels for transmutation. Combined with NCC/HRGS for interpretation.

* For typical MA isotopic composition in spent LWR/FBR fuels
** Can be lowered by factor of 10 when using microcalorimeters

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Conclusions

- Nuclear science and measurement technology have made considerable progress
- Remains an attractive field for young scientists and engineers
- Need to maintain competences and facilities: strengthen cooperation with universities
- Materials Science: trend goes to micro-measurements of axial/radial dependence of properties (10-20µm)
- Laser technology opens new and innovative areas of fundamental science and applications
- Nuclear measurements: trend goes towards higher detection efficiency and capability to measure short-lived isotopes in very low quantities

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