Partitioning Research at JRC – ITU
Recent Progress

J-P. Glatz, L. Casseyre, B. Christiansen, R. Malmbeck, M. Ougier, H. Ottmar, D. Magnusson, J. Serp, D. Serrano-Purroy, P. Soucek, T. Inoue, K. Uozumi

9th Information Exchange Meeting on Actinide and Fission Product Partitioning & Transmutation

26. – 28. September 2006
Nîmes, France

Partitioning activities at ITU

- ✓ Institutional program: Partitioning and Transmutation
  - activities in the field of hydro- and pyro-reprocessing
  - demonstration of separation schemes on simulated and irradiated materials
- ✓ Participation in European projects actually EUROPART
  - contributions to hydro- and pyro-metallurgy WPs
  - joint partnership with CRIEPI
- ✓ contractual collaboration with CRIEPI since 1991
  - reprocessing of the metallic fuels U,Pu,Zr with up to 5% An's and Ln's
  - study processes on calcined - chlorinated HLW from PUREX type waste
  - study of direct electroreduction processes for the head-end conversion of oxides to metal

ITU competences: P&T

Aqueous partitioning

- *demonstrate minor actinide separation schemes in view of an industrial implementation*

Pyrometallurgical reprocessing

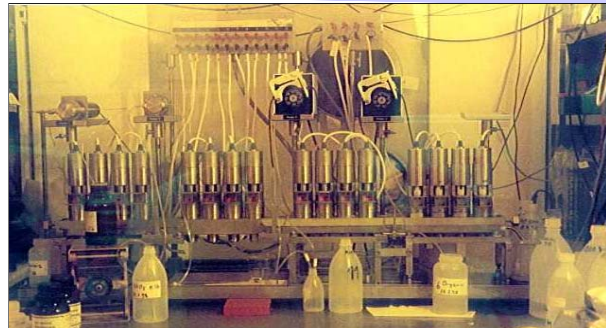
- *reprocessing of new types of fuels (e.g. metallic U,Pu,Zr,MA) and HLW*
- *electroreduction of oxide fuels*

PIE of innovative fuels

- *CERMET, inert matrices, metallic fuels, ThO_2, nitrides, carbides etc.*

Minor Actinide Laboratory

- *fabrication of minor actinide containing fuels and targets for transmutation*



New Centrifugal Contactor equipment

Chlorination equipment

Pilot facility for pyrometallurgical reprocessing

Hot Cell Laboratory

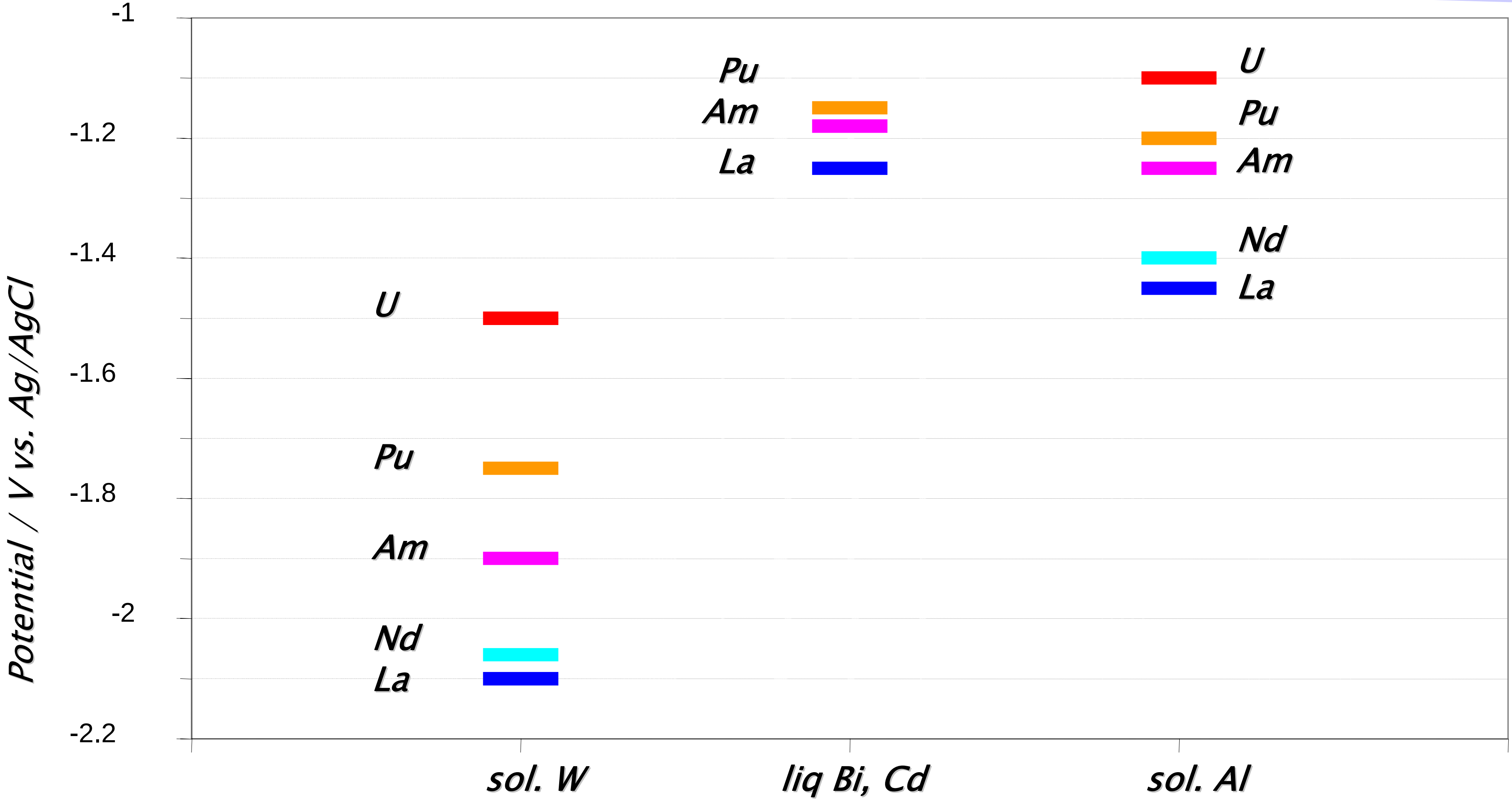
Minor Actinide Laboratory active operation started

Pyro-reprocessing objectives

- *develop reprocessing schemes for genuine fuels*
- *demonstrate co-recycling of **all** actinides (goal for new generation reactor systems)*
- *achieve an efficient separation of all actinides from lanthanide fission products*
- *set-up an efficient safeguarding technology*
- *investigate appropriate head-end conversion especially oxide to metal*
- *study the recovery and the recycling of actinides (fuel re-fabrication)*

Pyroprocessing of metallic fuels

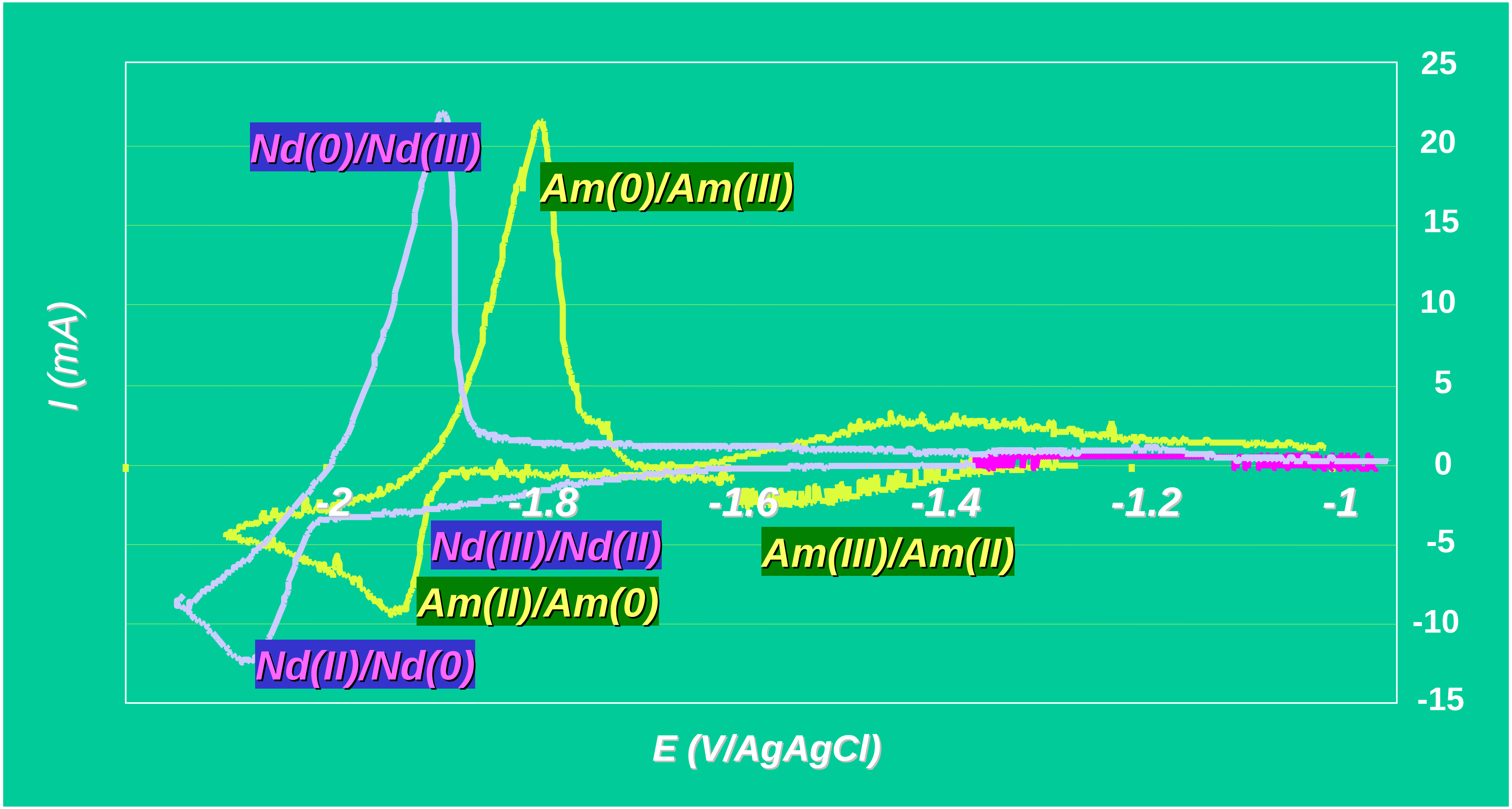
An reduction potentials on different cathodic materials



- more efficient An/Ln separation on solid cathodes (W and Al)
- alloying with An stabilizes the deposit (liquid Cd ,Bi and Al)
- Al unifies both advantages

Pyroprocessing of metallic fuels

Am and Nd valency in molten salt



cyclovoltamograms of Nd and Am in liquid Cd

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
Joint Research Centre

Electrolyses of U,Pu,Zr,Ln,MA alloy:
batch experiment involving 25 runs (~30g of fuel)

	Wt % in salt						Composition of the dissolved deposit (mg)			
	Before Run 1	After Run 1	Before Run 11	After Run 11	Before Run 14	After Run 14	Run1	Run 7	Run 11	Run 14
U	2.55	2.14	0.25	0.30	0.20	0.30	936	512	223	176
Pu	0.96	1.58	0.65	0.50	0.32	0.27	94.2	243	203	146
Am	0.09	0.17	0.24	0.23	**0.19**	**0.19**	***4.4***	***13.5***	***9.95***	***8.8***
Zr	-	-	0.056	0.096	0.055	0.075	-	-	-	-
Y	0.016	0.048	0.074	0.075	0.075	0.078	0.039	0.018	0.015	0.008
Ce	0.021	0.047	0.078	0.085	0.089	0.091	0.041	0.026	0.024	0.011
Nd	0.177	0.403	0.59	0.61	0.65	0.65	0.374	0.244	0.183	0.092
Gd	0.025	0.048	0.069	0.073	**1.8***	**1.8***	0.042	0.025	**0.018**	**0.027**
Al	-	-	-	-	-	-	387	301	173	134
				Current density (mA/cm²)			17	9.3	8.2	6.1
				Charge (C)			1386	1193	674	598
				Faradic efficiency (%)			91	70	80	70
				$m_{An}/(m_{An} + m_{Ln})$ (%)			99.95	99.96	99.9	99.9
				m_{Am}/m_{Ln}			8.8	43	**42**	**26**

* *added to the melt as $GdCl_3$*

- *Gd has the lowest electrodeposition potential difference compared to Am*
- *at Gd conc. corresponding to ~250 runs (300g of fuel) Am/Ln sepation still efficient*

Joint Research Centre

actinide separation from U,Pu,Zr,MA,RE fuels

- ✓ Adherent and compact deposit are obtained on solid Al with a good faradic yield (~ 90%)
- ✓ Separation of MA from Ln: the cathodic potential can be chosen in order to collect selectively U, Pu, Am (Ec > -1. 25V vs. Ag/AgCl 1 wt%)
- ✓ Efficient An/Ln separation by electrolysis was demonstrated for almost 30 g of U,Pu,Zr,MA,RE fuels
- ✓ separation efficient after 25 electrolyses of the same salt mixture (more than 6 months of experiments)
- ✓ Selective grouped separation of actinides from "realistic" fuel demonstrated

Direct electroreduction of irradiated FR oxide fuel

Experiments ongoing using SUPERFACT irradiated fuel, fabricated by ITU:

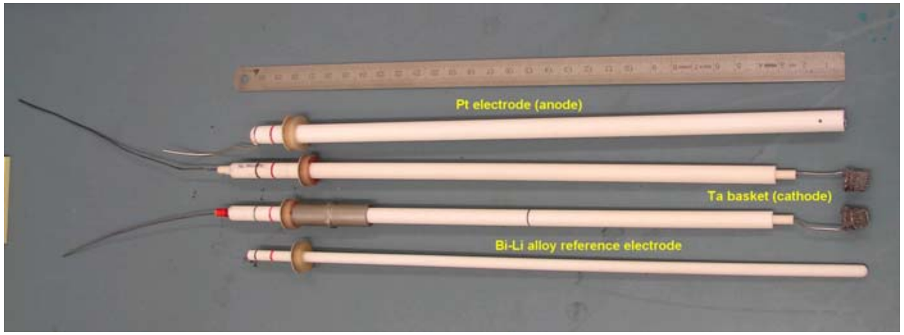
2 pieces of $(U_{0.74}Pu_{0.24}Am_{0.02})O_2$ and

2 pieces of $(U_{0.74}Pu_{0.24}Np_{0.02})O_2$

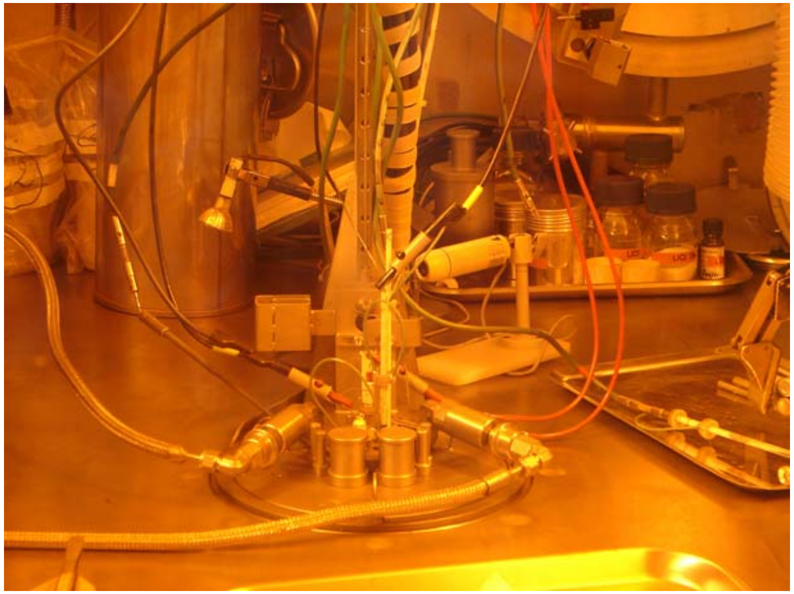
Crucible material: MgO

Electrolyte: LiCl + Li_2O (T=650°C)

apparatus



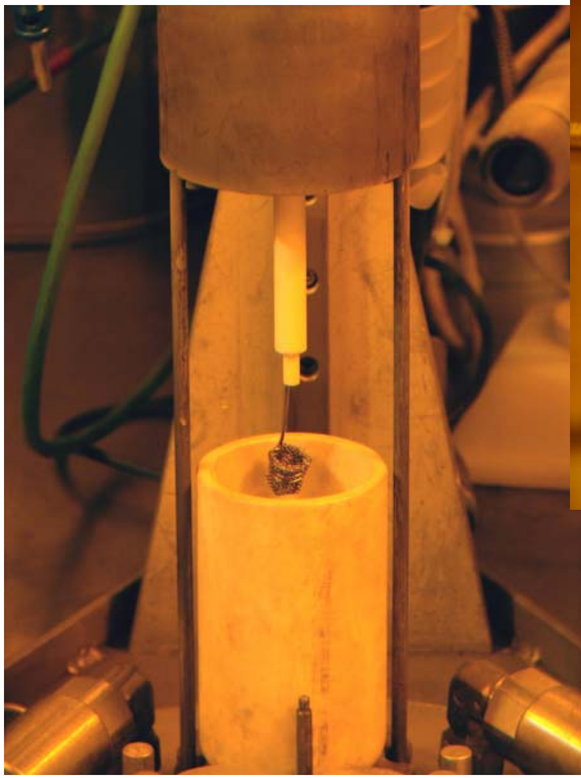
Set of electrodes



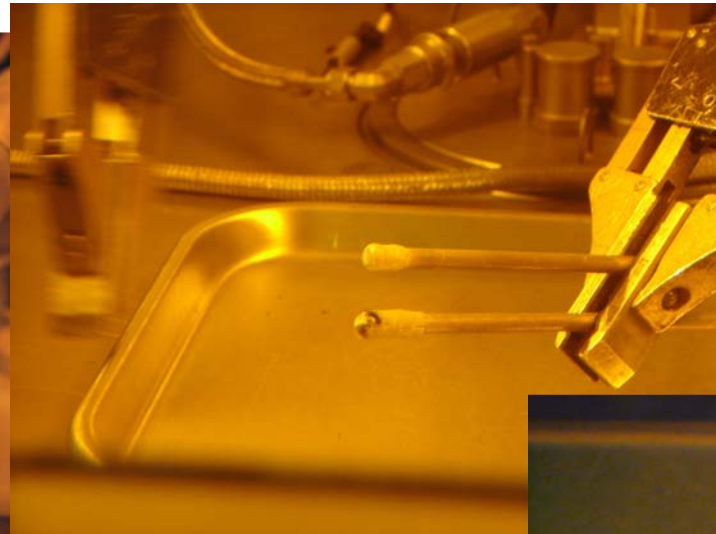
Setup inside the pyro caisson

Experimental set-up

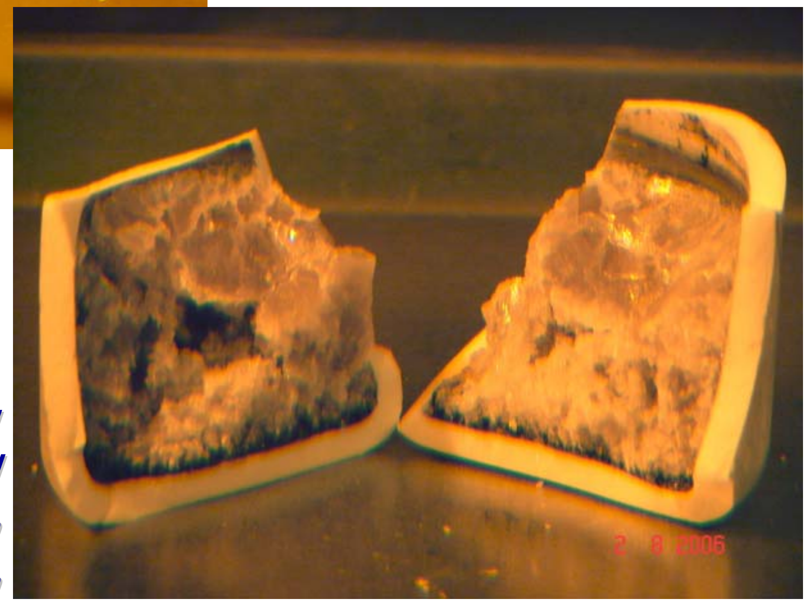
Direct electroreduction of irradiated FR oxide fuel



fuel loading



Salt samples taken from the top and bottom (incl. black fuel deposit) of the crucible

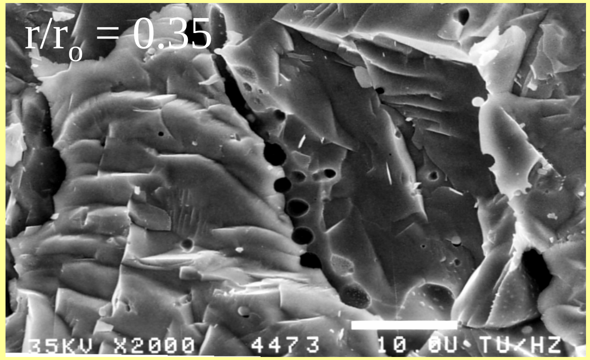


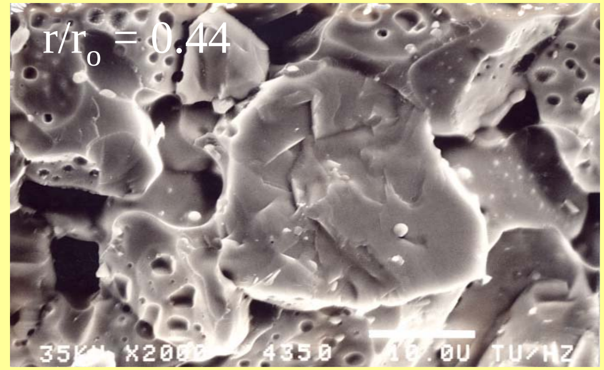
Crucible showing a black reduced fuel deposit on the bottom

preliminary results

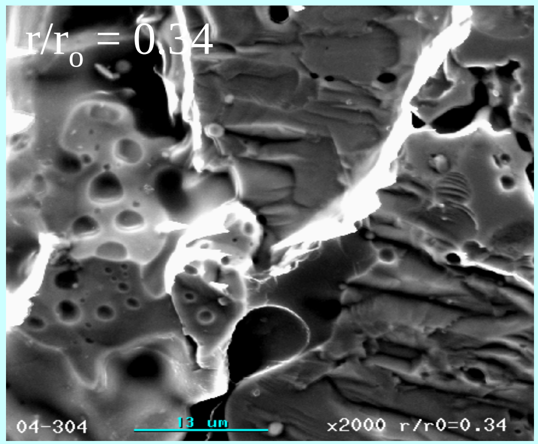
Direct electroreduction of irradiated FR oxide fuel

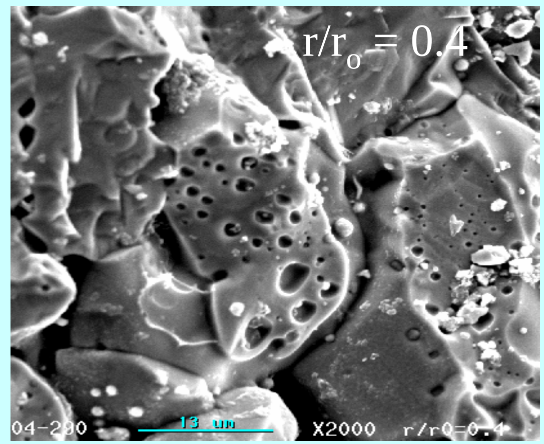
After irradiation 1991





After storage 2003





SEM examination of SUPERFACT fuel showing a strong intergranular porosity due to fission gas release

safeguards/non proliferation

- IAEA and EURATOM: effective flow sheet verification (FSV) at ITU to update the existing design for safeguarding U and Pu, and to provide additional information relevant to Np
- proliferation and safeguards issues are far less defined and investigated for pyroprocessing
- new schemes, approaches and analytical techniques under development
  - K- X ray Fluorescence (K-XRF)
  - Neutron Coincidence Counting (NCC)
  - High Resolution Gamma Spectrometry (HRGS)
  - Calorimetry

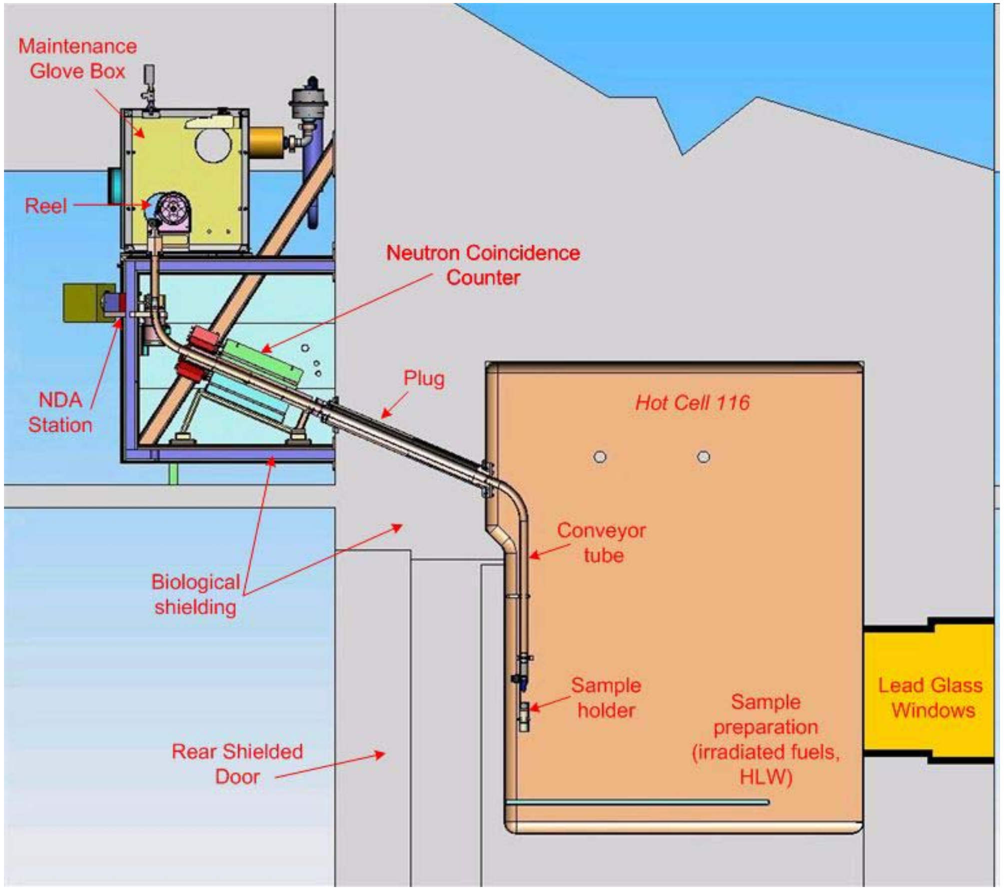
Application of radiometric techniques for MAs

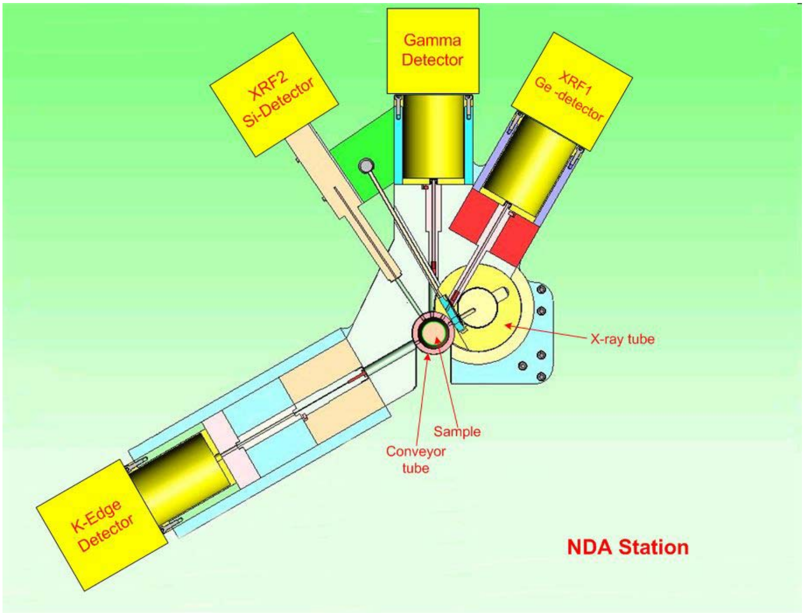
Technique	Element/ isotope measured	Isotope contribution to response*	Minimum amount for assay	Application
K-XRF	Np Am Cm	- - -	50 µg 70 µg 100 µg	Any sample type in liquid form mass fractions of analyte ≥ 0.02 %.
NCC	Cm	^{244}Cm. 90-95% ^{246}Cm: 5-10%	200 ng	For any type of Cm-containing samples (liquid 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

New analytical equipment for Pyrometallurgical Processes

Objective: to install NDA assay system for a direct non-destructive assay on irradiated samples





Status of the project

- Design completed
- Parts under manufacturing: Glove box, NCC, plug and supporting frame for shielding

Advanced NDA assay system

Joint Research Centre

Pyro-Partitioning: outlook

- ✓ reprocessing of irradiated Zr based metallic fuels; METAPHIX (collaboration with CRIEPI and CEA)
- ✓ TRU recovery from Al cathode: depending on the subsequent fuel type
- ✓ define limitations regarding the use of the salt bath
- ✓ determine recovery rate of actinides: use of graphite anodes
- ✓ clean-up of the salt and waste treatment
- ✓ study of alternative methods for oxide fuel reduction (collaboration with CRIEPI)
- ✓ basic thermodynamic data on Cm: collaboration with RIAR Dimitrovgrad in ISTC project
- ✓ integration study of the electrorefining process: collaboration CEA (S. Bourg, H. Boussier) -ITU

METAPHIX irradiation experiment

Irradiation of U,Pu,Zr,Ln,MA alloys

CENTRALE PHENIX
CEA-EDF

3 capsules;

3 different compositions

under irradiation since November 2003 in the PHENIX reactor in Marcoule

1. 90 PEPD 2.4 % burn-up
   unloaded: 8. August 2004
   transport to ITU 25. September 2006
2. 3 cycles of 120 PEPD 7 % burn-up
   unloaded: 20. July 2006
3. 5 cycles of 120 PEPD 11 % burn-up
   to be unloaded beg. 2009

work initiated by CRIEPI, Japan; now trilateral project CRIEPI, CEA, ITU

CRIEPI

Pyroprocessing: Tentative Test Plan