

DE LA RECHERCHE À L'INDUSTRIE

**cea den**

CEADEN

# **REVIEW OF FOUR DECADES DEDICATED TO SPENT FUEL ANALYSES IN FRANCE: AN INCOMPARABLE DATABASE BUILT FOR CODES VALIDATION.**

S. Cathalau, J.F. Lebrat, L. San-Felice, B. Roque  
M. Guillo – P.M. Demy

PAGE 1

DE LA RECHERCHE À L'INDUSTRIE

cea den

CEADEN

## Content

1. Introduction
2. Experimental database for validation
3. Spent fuel analyses :
  1. Experimental processes
  2. Performed Measurements
  3. Uncertainties
  4. Example of (C-E)/E
4. Complementary Experiments
5. Conclusions

## o Almost 60 NPP France in operation in 2016

- ❖ More than 1200 t of new fuel loaded annually
- ❖ About 5000 MOX assemblies loaded in EDF plants since 1985 coming from irradiated fuel reprocessing

## o Improvement of Fuel use in reactor

- ❖ Increase of burn-up
- ❖ Reprocessing processes control and improvement
- ❖ Waste management

## o Calculation tool needs

- ❖ Increase of precision and predictability for isotopic inventory
- ❖ Experimental Validation for safety demonstrations

## o Experimental data required

- ❖ Several types of fuel (UO<sub>x</sub>, MO<sub>x</sub>, Enriched Retreated Uranium)
- ❖ Huge range of burn-up
- ❖ Any kind of NPP



In order to validate the French code systems for fuel inventory calculations, a large experimental program has been carried out in France since 1980's based on :

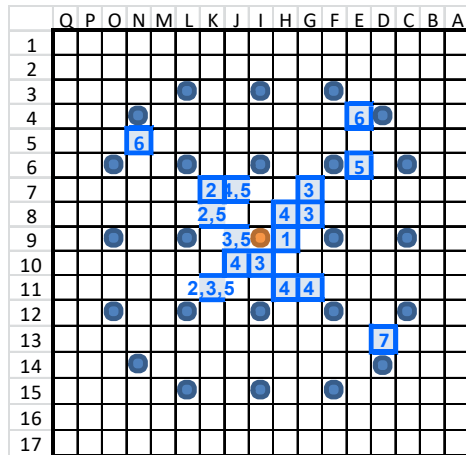
- ❖ Spent fuel chemical analyses (from fuel rod irradiated in PWR – UOX and MOX, but also for BWR-UOX) aimed at determining the isotopic inventory depending on the burn-up
- ❖ Dissolution aliquots of entire assembly sets obtained from La Hague reprocessing plant.
- ❖ Oscillations of irradiated fuel samples in the center of the MINERVE facility located at the Cadarache center for the total reactivity loss during cycles and burn-up credit studies

#### Isotopes :

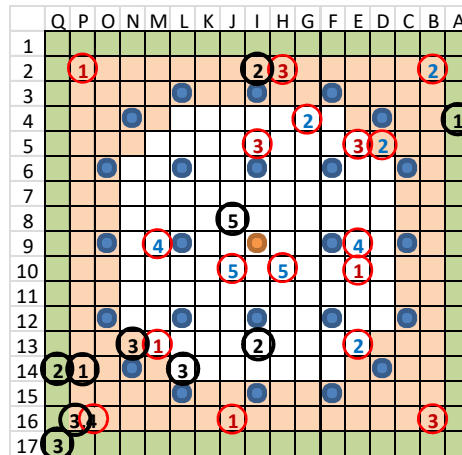
- Main Actinides : U, Np, Pu, Am, Cm.
- Important Fission Products including : FP involved in Burn Up Credit studies ( $^{143, 145}\text{Nd}$ ,  $^{147, 149, 150, 151, 152}\text{Sm}$ ,  $^{155}\text{Gd}$ ,  $^{153}\text{Eu}$ ,  $^{133}\text{Cs}$ ,  $^{95}\text{Mo}$ ,  $^{103}\text{Rh}$ ,  $^{109}\text{Ag}$ ,  $^{99}\text{Tc}$ ,  $^{101}\text{Ru}$ ) in decay heat ( $^{90}\text{Sr}$ ,  $^{144}\text{Ce}$ ,  $^{147}\text{Pm}$ ,  $^{101, 106}\text{Ru}$ ) or used to evaluate the burnup of the samples (Nd).



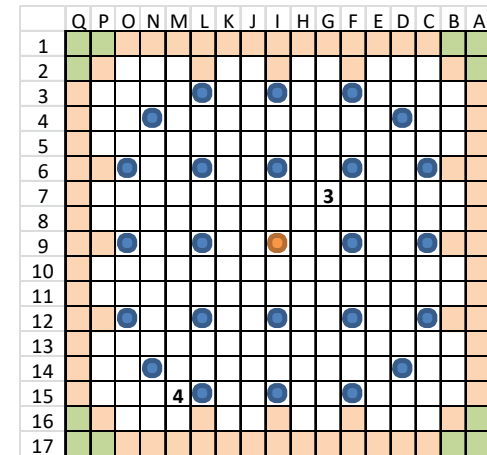
Fuel	Power Plant	Enrichment	Burn Up (GWd/t)	Isotopic Analyses	Burn-Up credit Fission product	Oscillations in MINERVE
UOX-Gd	« GEDEON »		2-12	U, Pu, MA, FP		
UOX	BUGEY & FESSENHEIM	2.1% & 3.1%	20 - 60	U, Pu, MA, FP	x	x
UOX	GRAVELINES	4.5%	25 - 60	U, Pu, Am, FP	x	x
ERU	CRUAS	3.5%	15 - 35	U, Pu, Am, FP		
UOX	BWR « GUNDREMINGEN »	4.0%	26 - 46	U, Pu, Am, FP	x	
UOX	GOSGEN « MALIBU »	4.3%	70	U, Pu, Am, FP	x	
UOX	GRAVELINES ALIX	4.5%	65 - 85	U, Pu, Am, FP		x
MOX	SLB1	2.9-5.6%	10 - 40	U, Pu, Am, FP		
MOX	DAMPIERRE	6.7%	10 - 58	U, Pu, Am, FP	x	x
MOX	TRICASTIN	9.8%	43 - 56	U, Pu, Am, FP	x	planned



UOX in BUGEY, FESSENHEIM and GRAVELINES

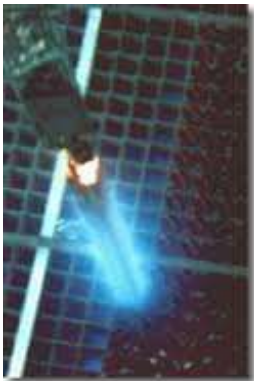


MOX in SLB1 and DAMPIERRE



MOX in TRICASTIN

From the power plant to non destructive examination labs



Extraction of the pin



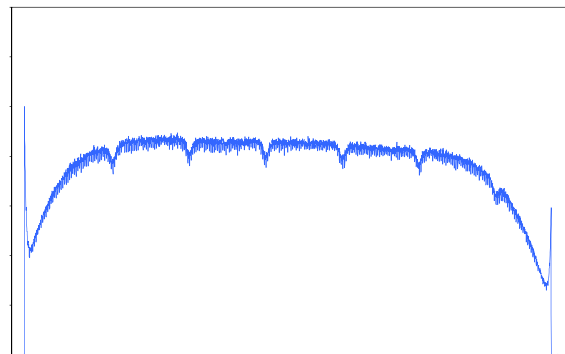
Shipping



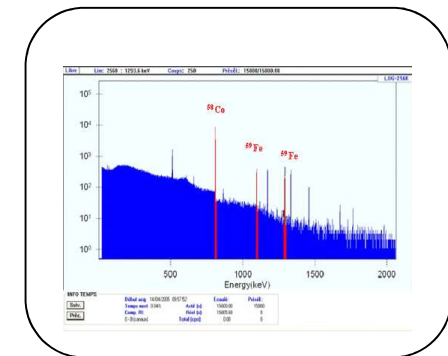
Introduction in hot cell for NDE



- Total  $\gamma$  spectrometry
- Estimation of Burn-up by  $^{134}\text{Cs}$  or  $^{137}\text{Cs}$  counting
- Axial Profile
- Determination of best location for cut-off and fuel sample to be analyzed.



Axial burn-up profile



$\gamma$  spectrometry

From non destructive examination labs to the dissolution facility then chemical analyses



Cut-off of a ~2 cm sample

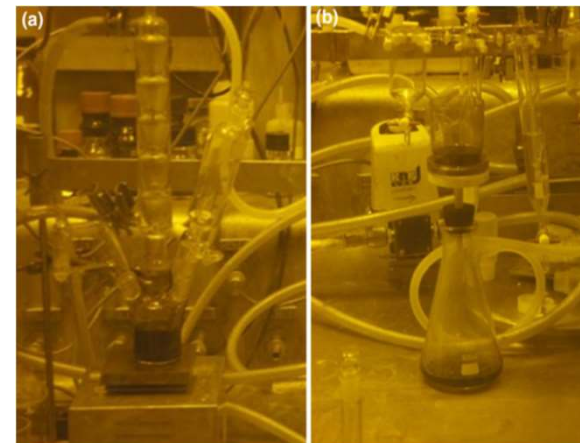


ATALANTE Facility

- Cut-off of the sample
- Gaseous FP measurements
- Shipping to the ATALANTE facility
- Characterization of sample (weight)
- Dissolution processes
- Shipping of aliquots to radiochemical analyses facility



Analyses



Dissolution in nitric acid, filtration

Simple Dissolution allows to determine :

❖ Heavy nuclide measurements :

- Isotopic ratios for uranium, plutonium, americium and curium
- $^{239}\text{Pu}/^{238}\text{U}$ ,  $^{237}\text{Np}/^{238}\text{U}$ ,  $^{241}\text{Am}/^{238}\text{U}$ ,  $^{244}\text{Cm}/^{238}\text{U}$

❖ Fission products :

- Isotopic ratios for neodymium, cesium, gadolinium and samarium
- $^{145}\text{Nd}/^{238}\text{U}$ ,  $^{137}\text{Cs}/^{238}\text{U}$ ,  $^{155}\text{Gd}/^{238}\text{U}$ ,  $^{149}\text{Sm}/^{238}\text{U}$ ,  $^{144}\text{Ce}/^{238}\text{U}$ ,  $^{90}\text{Sr}/^{238}\text{U}$

Multiple Dissolutions focus on following quantities (in addition to above ones) :

❖ Heavy nuclide measurements in non dissolved material during the first dissolution

- Isotopic ratios for uranium, plutonium
- $^{239}\text{Pu}/^{238}\text{U}$ ,  $^{237}\text{Np}/^{238}\text{U}$

❖ Metallic Fission products :

- $^{95}\text{Mo}/^{238}\text{U}$ ,  $^{99}\text{Tc}/^{238}\text{U}$ ,  $^{109}\text{Ag}/^{238}\text{U}$ ,  $^{101}\text{Ru}/^{238}\text{U}$ ,  $^{103}\text{Rh}/^{238}\text{U}$



# SPENT FUEL ANALYSES : COMPARISON OF SINGLE AND MULTIPLE DISSOLUTIONS



Recently performed for a MOX fuel irradiated during 4 cycles in Tricastin Plant

A single dissolution was performed for “classical” isotopic ratios

Multiple dissolutions were done of a neighboring sample (same fuel pin) for classical ratios and metallic fission products.

Classical Nuclide Ratio	Chemical analyses Discrepancy (%)
$^{235}\text{U}/^{238}\text{U}$	$0.2 \pm 0.3$
$^{238}\text{U}/^{239}\text{Pu}$	$0.1 \pm 0.6$
$^{240}\text{Pu}/^{239}\text{Pu}$	$0.1 \pm 0.3$
$^{241}\text{Am}/^{238}\text{U}$	$0.4 \pm 0.8$
$^{148}\text{Nd}/^{238}\text{U}$	$0.1 \pm 1.2$
$^{135}\text{Cs}/^{238}\text{U}$	$0.2 \pm 1.1$

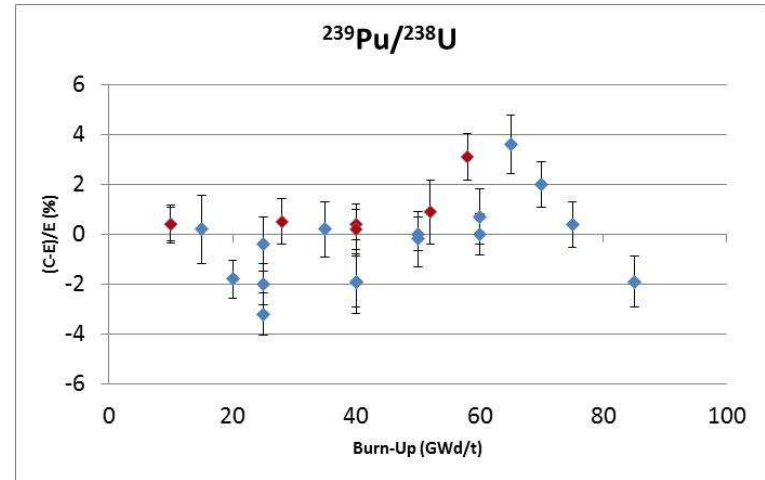
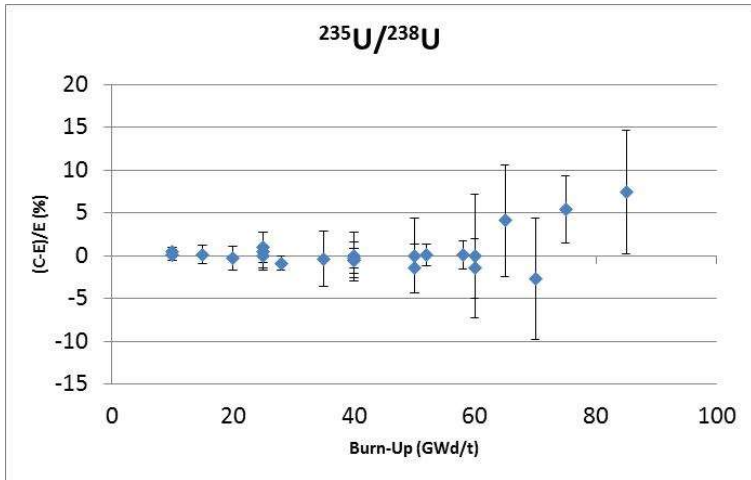
The observed discrepancies between the both processes are within the uncertainty margins (purely chemical uncertainties) -

o Each process described above generates its own uncertainty :

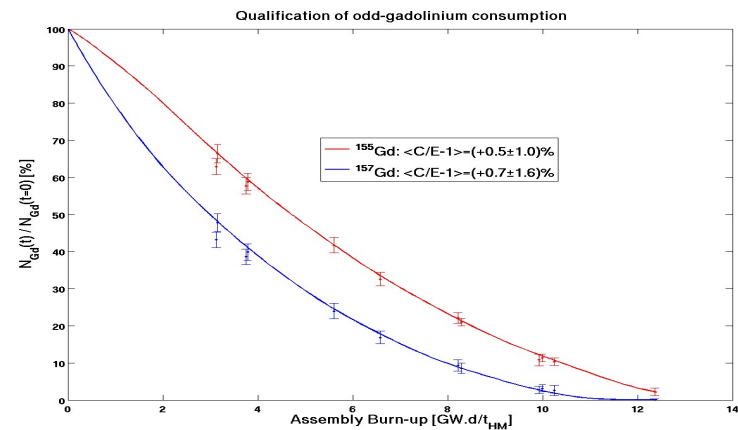
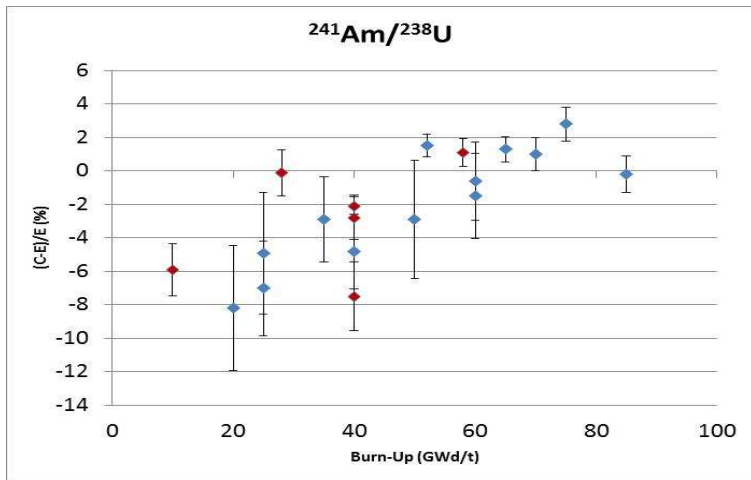
1. The material balance of the original fuel ( $\pm 0.2\%$  up to  $\pm 1.0\%$  -  $1\sigma$ )
2. The irradiation profile of the core ( $\pm 0.4\%$ )
3. Irradiated fuel sample : the precise location of the cutoff is known at  $\pm 5$  mm which induces uncertainty on local burn-up ( $\pm 2.0\%$  due to the yields for Nd isotopes), fuel temperature ( $\pm 50^\circ\text{C}$ ), moderator temperature ( $\pm 2.0^\circ\text{C}$ )
4. Dissolution processes (losses during transfers, non dissolved parts, pollutions, evaporation, residual drops in containers, ...) ( $\pm 0.2\%$  up  $\pm 0.4\%$ )
5. Isotopic analyses (final process) - (from  $\pm 0.1\%$  up  $1.4\%$ )

o Globally the total uncertainty does not exceed  $\pm 5.0\%$  for classical isotopes and burn-up less than 60 GWd/t.

# SPENT FUEL ANALYSES : EXAMPLES OF (C-E)/E IN% OBTAINED WITH A TYPICAL DEPLETION CALCULATION ROUTE



Red points for MOX fuel



Gadolinium odd isotopes depletion

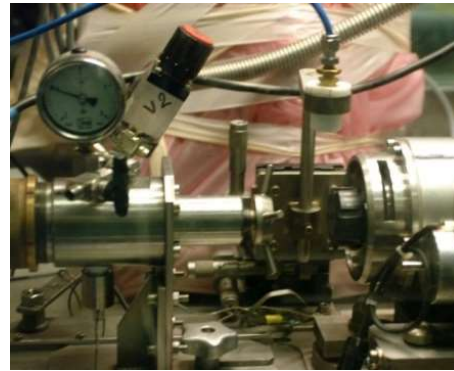
■ Preparation of samples of 10cm rod cut of irradiated fuel pins from NPP (PWR, BWR)



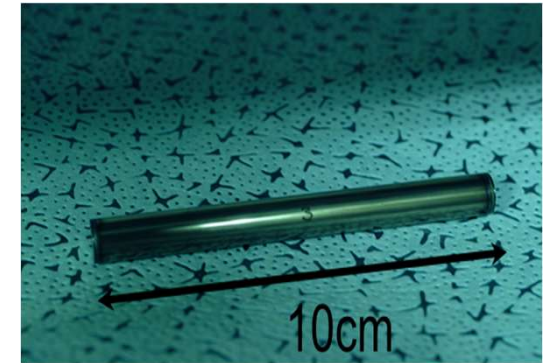
Spent fuel pin extraction



Cutting and reconditioning of a spent fuel sample



Double Zy4-cladding



Final Sample

→ Provide information on the overall reactivity worth due to FP and actinides

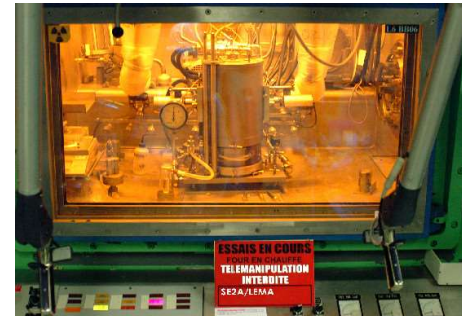
■ Manufacturing of special samples by the addition of separated isotopes of FP or actinides, homogeneously mixed with  $UO_2$  powders to prepare sintered pellets



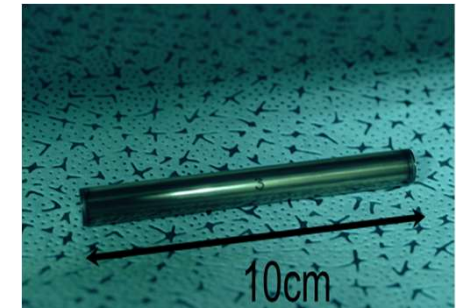
Mixing of  $UO_2$  powder with separated actinide



Preparation of fuel pellets



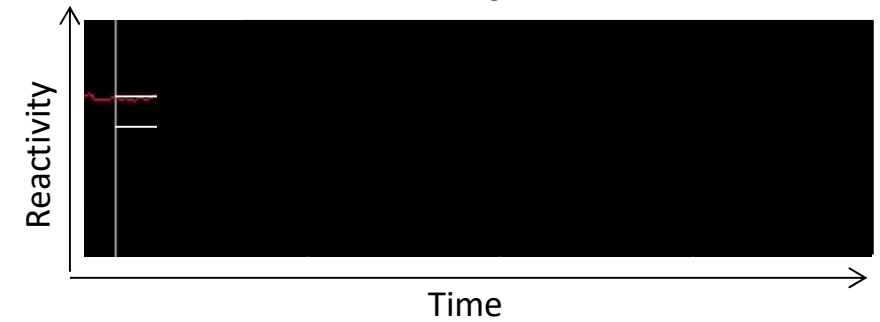
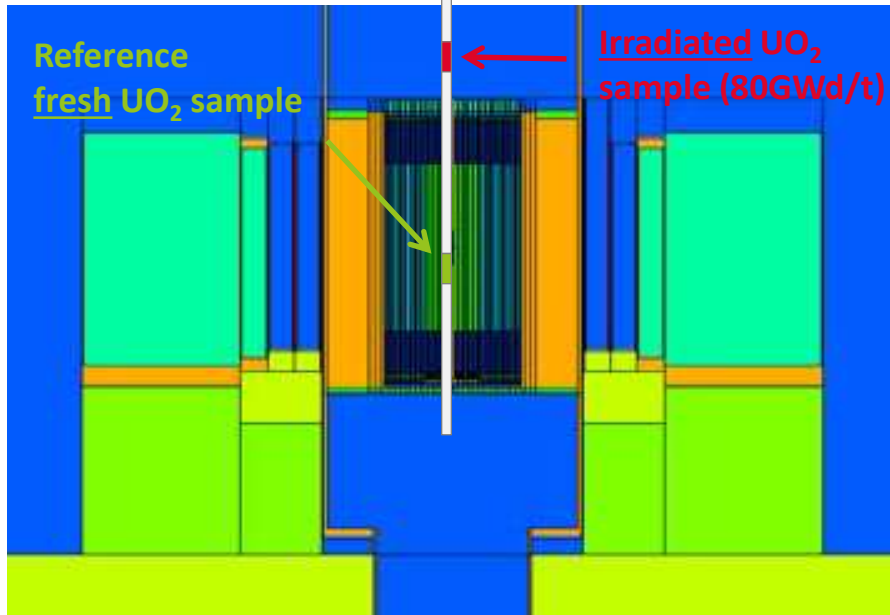
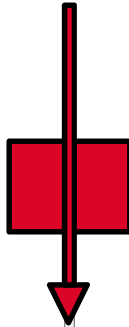
Sintering process



Final Sample

→ Provide information on the individual reactivity worth of FP and actinides

# COMPLEMENTARY MEASUREMENTS : OSCILLATION TECHNIQUE FOR ACCURATE RESULTS



- ⇒ High accuracy in the measurement of reactivity worth:  $\leq \pm 1\text{c}$  ( $\approx 2 \mu\text{g}$  of  $^{10}\text{B}$ )
- ⇒ Validation of the fuel reactivity loss with the burn-up within  $\pm 3\%$  ( $1\sigma$ )



- o From the 80', France has launched an ambitious experimental program aimed at validating depletion codes for LWRs
  1. More than 50 samples were analyzed (UOX, MOX) & (PWR, BWR)
  2. More than 1500 isotopic ratios.
  3. Range of burn-up between 2 and 85 GWd/t
  4. Heavy nuclides including minor actinides ; fission products
  5. Continuous Improvement of dissolution processes
  6. Advanced analyses techniques
  7. Total accuracy compatible with code validation requirements
- o This base is now available in France for the validation of current and future depletion codes used for any safety demonstration during irradiation cycles and cooling spent fuel in storage (pools or dry storage)