

DE LA RECHERCHE À L'INDUSTRIE

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ANALYTICAL DEVELOPMENTS FOR HIGH PRECISION ISOTOPE MEASUREMENTS ON TRANSMUTATION TARGET IN THE PHENIX FAST NEUTRON REACTOR

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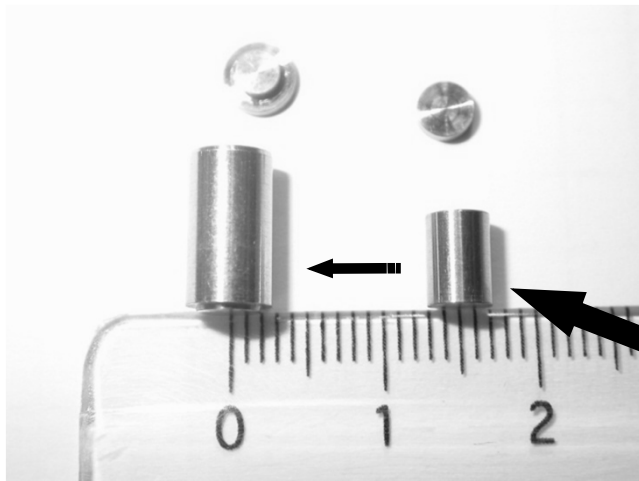
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- The **PROFIL-R** (fast spectrum) and **PROFIL-M** (moderated spectrum) experiments were performed between 2003 and 2008 in the **French fast neutron reactor Phénix**.
- The objectives are to measure the **total neutron capture integral cross sections** of the principal heavy isotopes and some important fission products **in the spectral range for fast reactors**.
- The method consists in the **irradiation of pure isotope samples** in a well-characterized flux and the **determination of the composition change** induced by irradiation.
- The experimental data are compared with **neutronic calculation code** for data determination or validation, or identification of bias.
- A unique neutronic data base is built for **neutronic calculation code** for **fast neutron reactor** and for **transmutation studies** (particularly for Am and Cm targets)





- Powders enclosed in very small **steel containers**

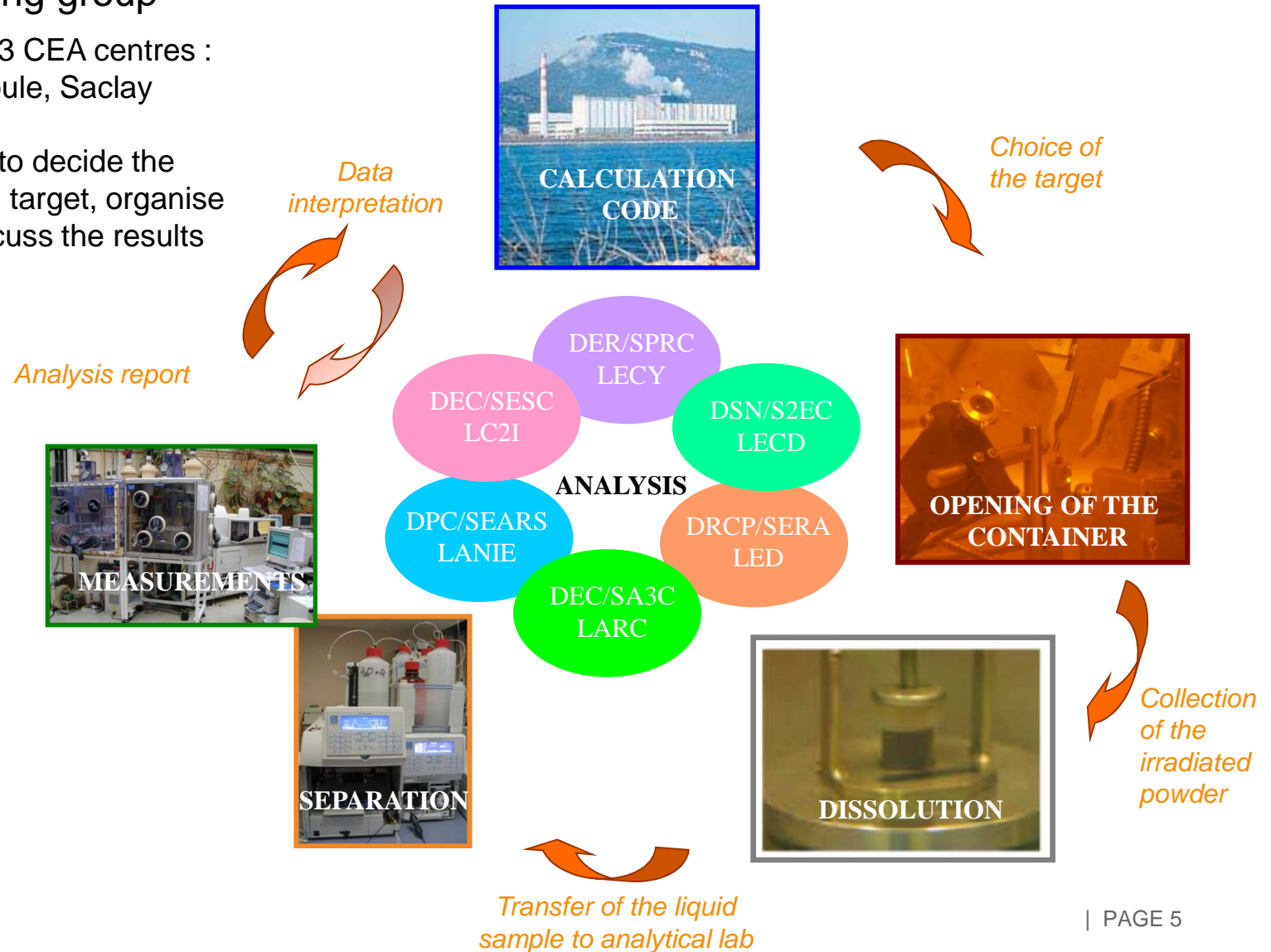
- Mass of the steel container
150 mg
- Mass of powder
2-5 mg
- Some elements studied (like Mo) are present in **impurities** in the steel container

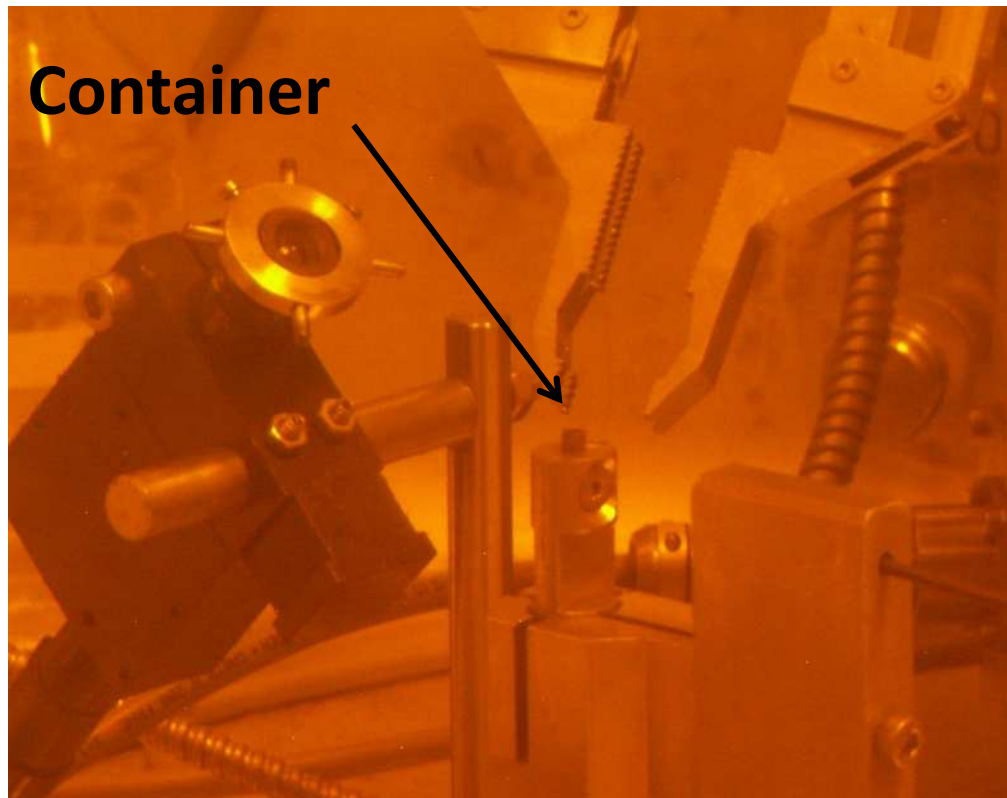
- **Developments of specific tools and protocols to collect the powder with limited contamination from elements of the containers**
- **Analysis of irradiated samples with the highest accuracy : dissolution and analysis by mass spectrometric methods**
- **Comparison calculation / experiment**

PROFIL Working group

Laboratories from 3 CEA centres :
Cadarache, Marcoule, Saclay

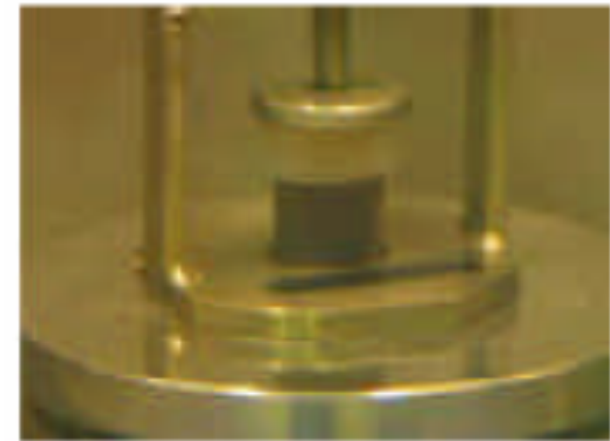
Regular meetings to decide the
strategies for each target, organise
the steps, and discuss the results



Objective 1: quantitatively collect the powder and dissolve it

Container

- Operations conducted in the **CHICADE** facility (Cadarache) and **ATALANTE** facility (Marcoule) in hot cells
- Use of an **electro chemical discharge machining** first, and then opening by **mechanical abrasion**



- Protocol of dissolution regarding the chemical form of the elements: Acid digestion, and use of microwaves
- Specific developments for the dissolution of some « difficult » powders such as **Ag, Mo, Ru, Rh, Pd**

Objective 2: Analyse the isotope and element compositions with high accuracy

ACTINEO facility (Saclay, LANIE)

Analytical methodology

- **Separative methods:**
 - Ion exchange resins
 - Extracting resins
 - HPLC
- **Mass spectrometry:**
 - TIMS
 - Q ICP MS
 - MC ICP MS
 - ID-MS technique



Specific analytical developments

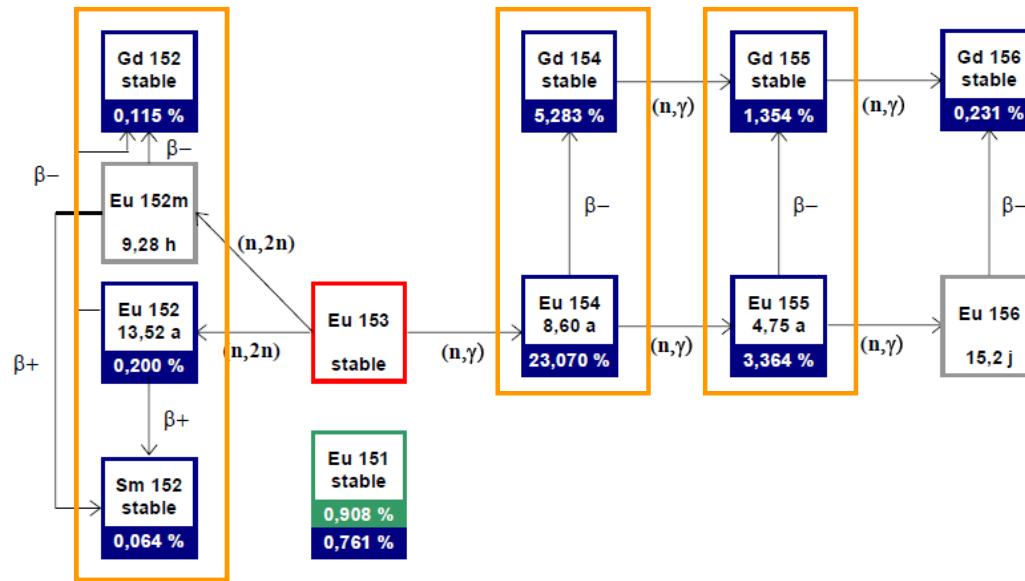
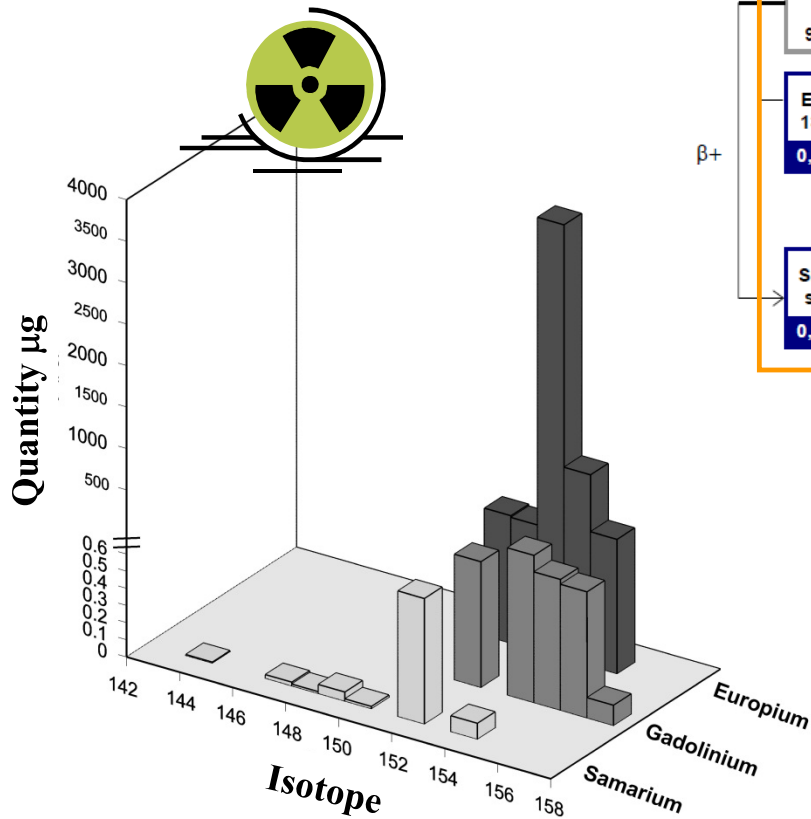


Chromatographic separation and hyphenated techniques

Direct separation using collision-reaction cell in ICP MS



■ Objective: Eu isotope ratios and ¹⁵²Sm/¹⁵³Eu ratio at uncertainty < 1%



■ Eu₂O₃ powder, enriched in ¹⁵³Eu (99.1%)

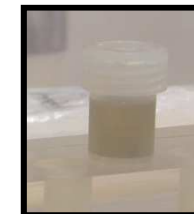
■ Dissolution HNO₃ 14N

■ Dilution required to keep the activity of the sample below 9900 MBq (= Max in gloves box)

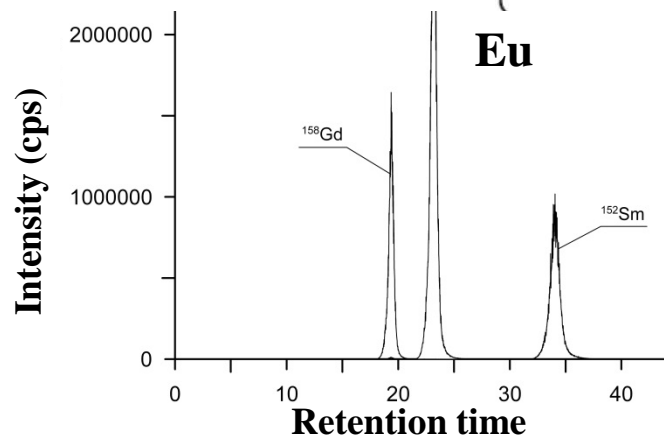
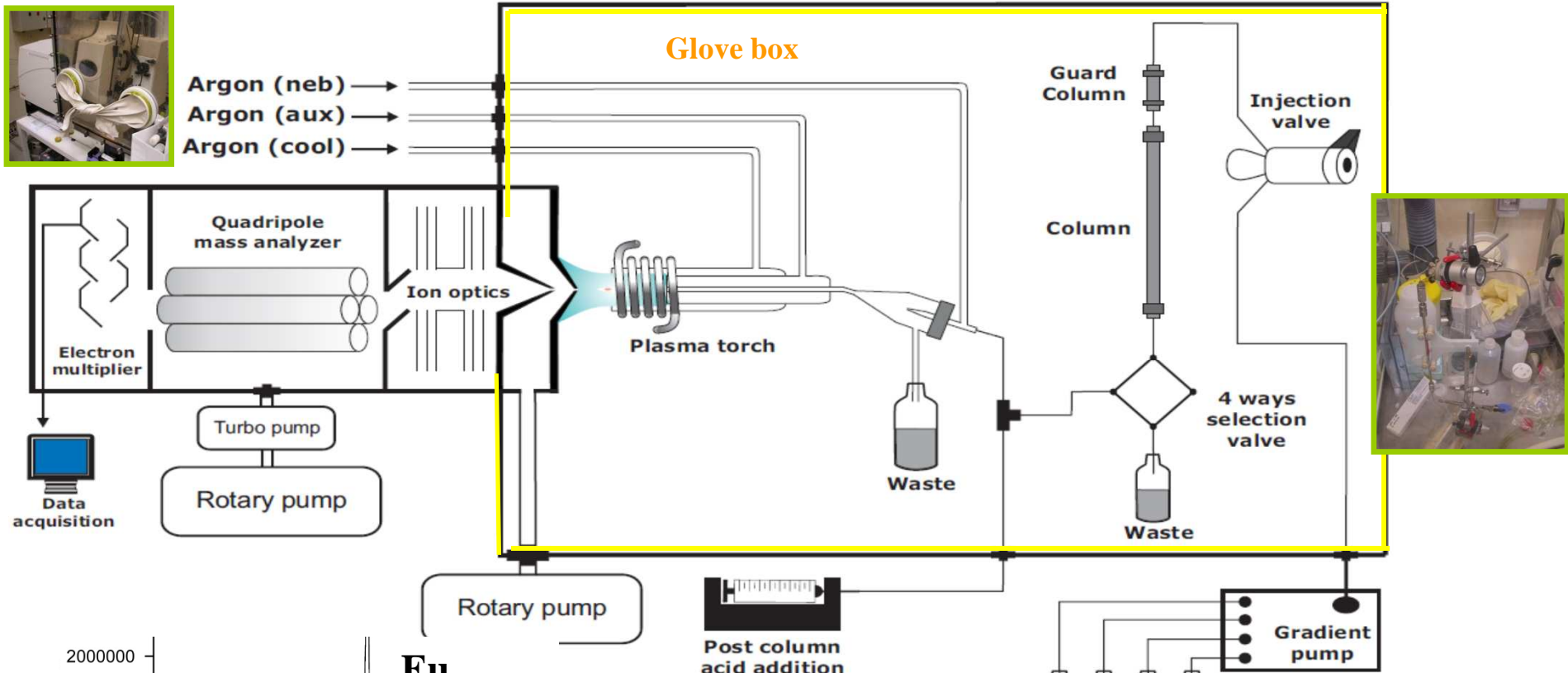
■ Eu = 32 µg

■ Gd = 2.4 µg

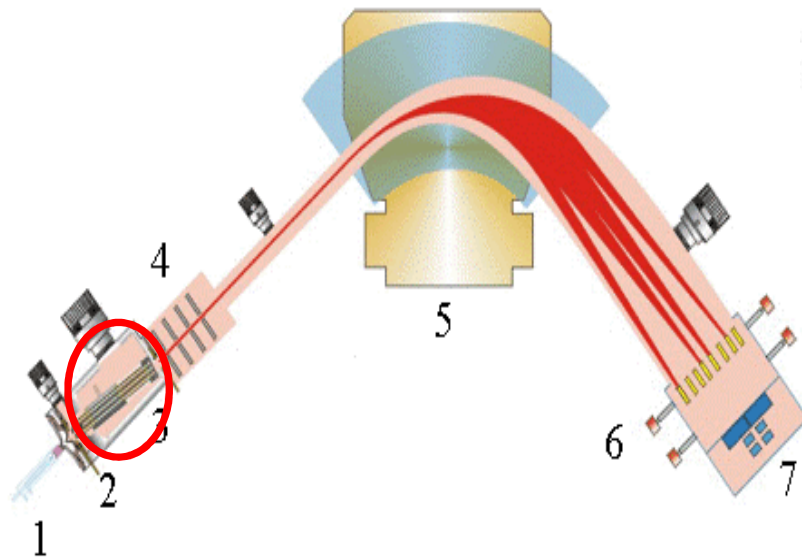
■ Sm = 0.018 µg



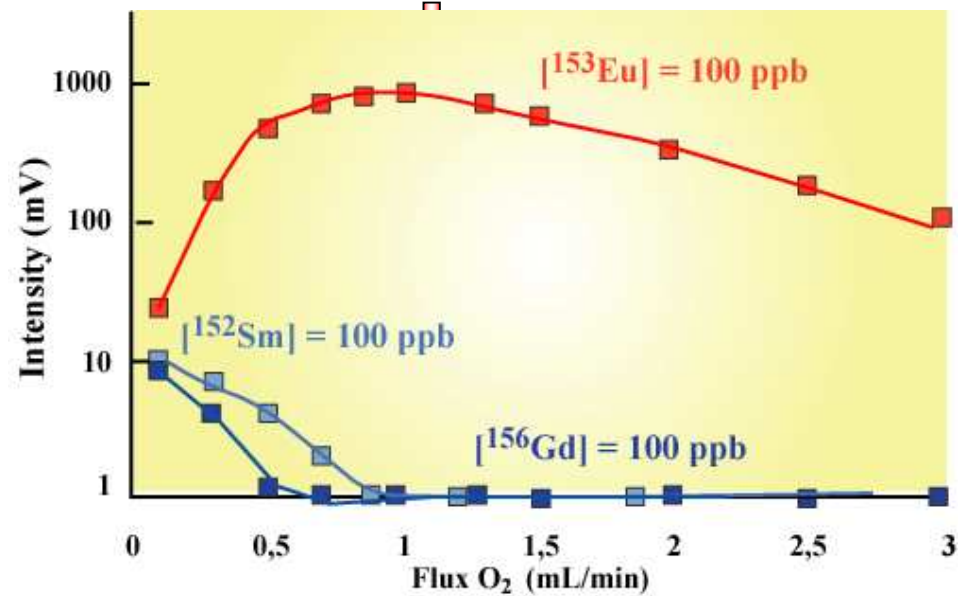
Sm ISOTOPE RATIOS BY COUPLING HPLC AND Q ICP MS



- Simplified separation procedure
- Reduction of the quantity of sample used by a factor 10



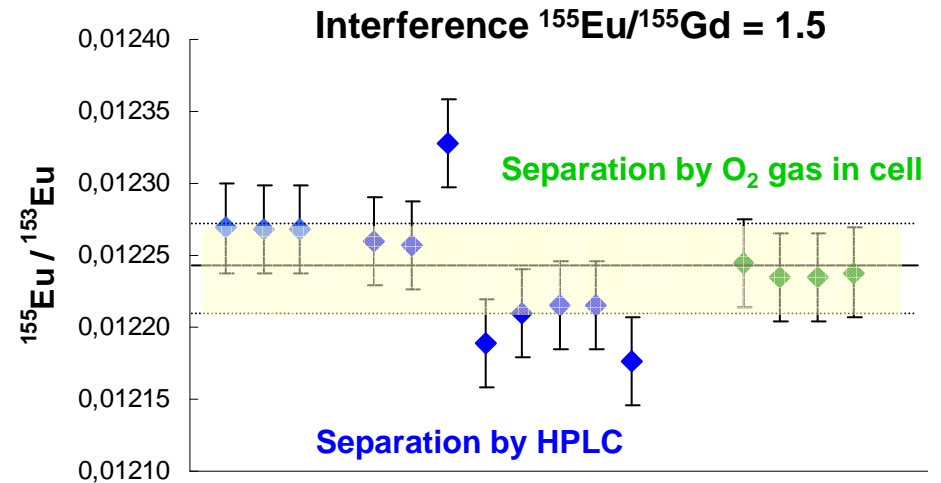
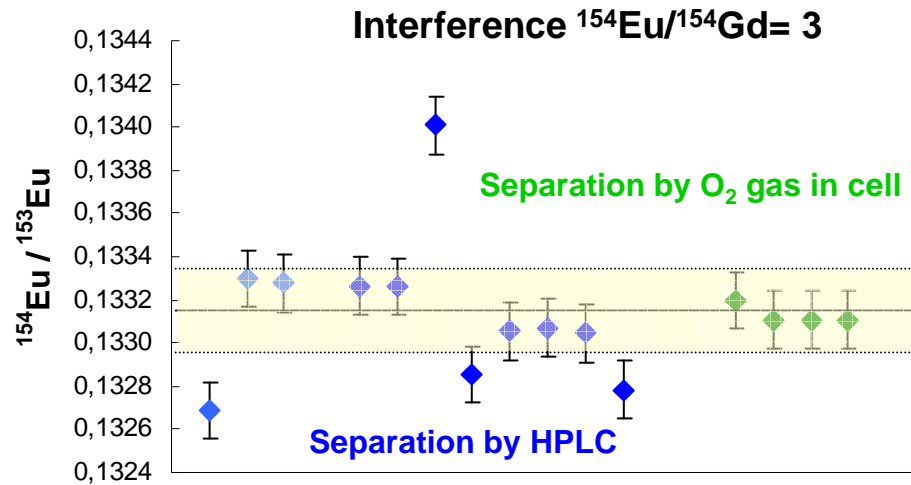
- Introduction of a reaction gas to perform *in situ* chemical separation



↪ Separation of Eu⁺ and SmO⁺-GdO⁺



No more isobaric interference at O₂ flow rate > 1,2 ml.min⁻¹



Uncertainty of a few ‰

- Reduction of the time consuming step
- Reduction of radioactive wastes
- Reduction of the time contact with radioactive samples
- Less sensitive to contamination of the sample by using an ICP MS collision - reaction cell

- ^{235}U target: direct separation in collision reaction cell for **U-Pu separation** using CO_2

Gourgiotis et al. (2010) JAAS, 25, p. 1939

- ^{92}Zr target: direct separation in collision reaction cell for **Mo-Zr separation** using N_2O

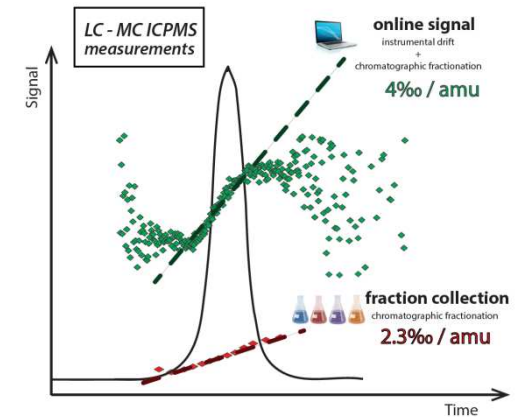
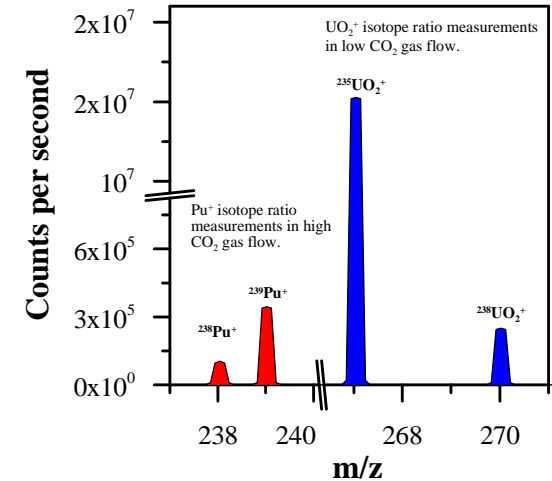
Moureau et al. (2008) JAAS, 23, p. 1538

- ^{109}Ag target: coupling between HPLC and Q ICP MS

Gautier et al. (2011) J. Chromat., 1218, p. 5241

- ^{235}U target: coupling between HPLC and MC ICP MS for **on line Nd isotope ratios** measurements

Gueguen et al. (2015) JAAS, 30, p. 443



- **Analytical developments** are essential in order to obtain **accurate isotope and elemental ratios** after irradiation
 - Very low quantities of elements present
 - High activities of the samples
 - Precise and accurate data are required

- These experiments constitute a unique database for **neutronic calculation codes** for **fast neutron reactor** and for **transmutation studies**

- At this stage of the projet **22 PROFIL-R targets** and **8 PROFIL-M targets** have been analyzed

- Analytical protocols are under development for Ru, Pd, Rh, Tc targets

Acknowledgements

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