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Americium, curium and neodymium analysis in ECRIX-H irradiated pellet: sample preparation for TIMS measurements

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Summary. This paper concerns quantitative isotopic analysis of Am, Cm and Nd contained in an irradiated AmO_{1.62}/MgO pellet. The complete analysis protocol is described, from dissolution of the pellets in a shielded line to the laboratory glove separation processes box for TIMS analysis. Emphasis is placed on the separation processes: by ion exchange resin in a hot cell and by HPLC in the laboratory. Intermediate measurements by X-ray fluorescence, alpha spectrometry, and ICP-AES are described.

1. Introduction

The primary objective of the ECRIX-H experiment was to assess the feasibility of transmuting americium microdispersed in an inert magnesia matrix (AmO_{1.62}/MgO) in heterogeneous mode in a partially moderated neutron flux in the Phenix reactor. Numerous characterizations were achieved on irradiated material by nondestructive measurements (visual inspection, metrology, gamma spectrometry, Eddy current testing [1–3]), and by destructive measurements (gas analyses, hydrostatic density measurements, scanning electron microscopy (SEM) [3], X-ray diffraction (XRD) [3], secondary ion mass spectroscopy (SIMS) [4], electron probe microanalysis (EPMA) [4]).

Among the destructive measurements, elemental and isotopic characterization of Am, Cm and Nd in an irradiated ECRIX-H pellet were carried out by the Analysis and Material Metrology Laboratory (LAMM) in the CEA Atalante complex at Marcoule. This work is the subject of this paper.

The LAMM laboratory performs analyses for R&D studies in Atalante, which frequently include high-activity analyses of spent fuel dissolution solutions.

Direct high-activity analyses requiring no sample preparation but which are subject to uncertainty of up to 30% can be distinguished from *indirect* analyses after sample treatment for much more precise analytical techniques that are not available in a high-activity environment.

Laboratory analysis of the intermediate-level solutions derived from high-level operations depends on the dilution

and/or decontamination factor to ensure that the sample activity and dose rate are compatible with safety standards for glove box work. When the analyte is present at low concentrations in the dissolution solution, the dilution factor is sometimes such that even the techniques with the lowest detection limits are ineffective. Separation techniques are then necessary for selective extraction of the analyte.

This article describes the steps required in the intermediate-level laboratory to obtain samples meeting the prerequisites for TIMS analysis. The results obtained are compared with the values calculated from neutronic simulation codes.

2. About neutronic simulations

ECRIX-H neutronic simulations were performed by using GEPHIX and CESAR calculation codes [5] devoted to the management of the PHENIX plant. Complementary simulations were also performed using the ERANOS and DARWIN codes [6] to obtain a more detailed description of fuel evolution during irradiation, including its composition in terms of helium and fission products.

Pellets on which chemical measurements were performed received about 95% of the overall fluence at the maximum flux plane. Based on the neutronic simulations, polynomial and linear interpolations of transmutation and fission rates were determined as a function of the received fluence. Using these interpolations, transmutation and fission rates can be extrapolated to account for the fluence at the maximum flux plan. Transmutation rate at the maximum flux plane is estimated to 94.3%.

3. Sample preparation for TIMS analysis

3.1 Experimental

The dissolution solutions were filtered by means of a vacuum filtration system with 0.5 µm PTFE filters.

Two types of resin were tested:

- Biorad AG1X8 with a mean particle size of 74–149 µm: about 2.5 mL packed in glass columns (4 mm diameter, 8 cm high) and equilibrated in 8 M nitric acid.

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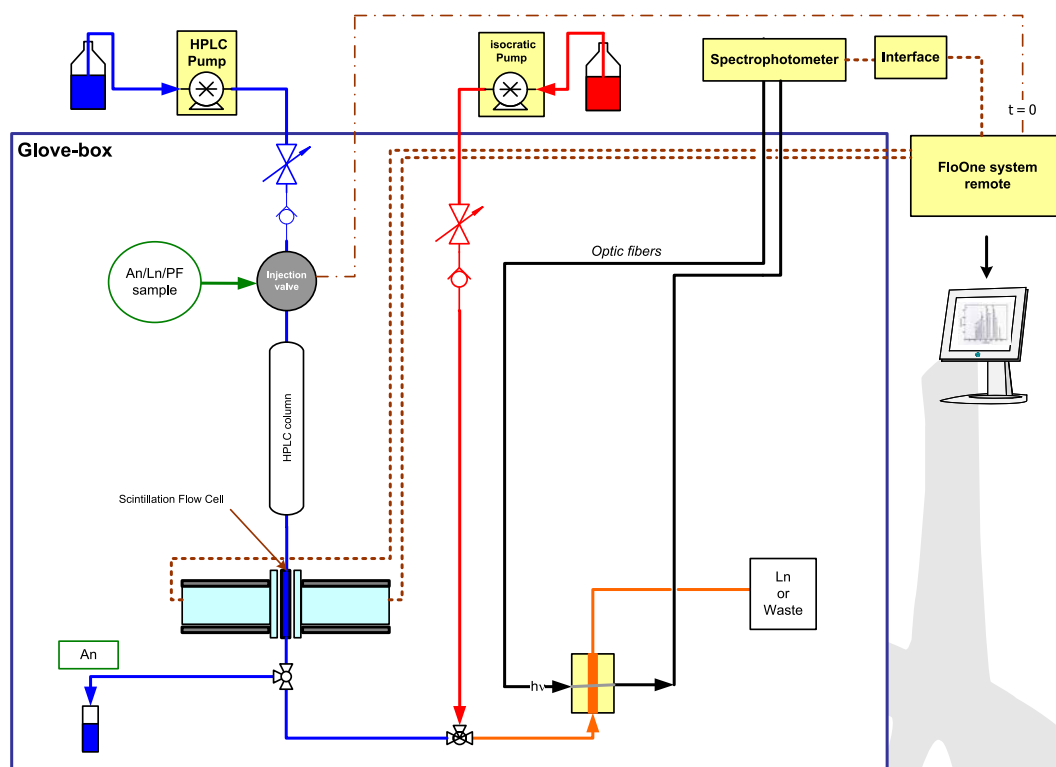


Fig. 1. Nuclearized HPLC system in a glove box.

- Eichrom Normal DGA used in pre-packed 2 mL columns with a mean particle size of 50–100 μm . Each column was preconditioned with 8 N nitric acid.

Gravity elution was performed with AG1X8 resin. An Ismatec peristaltic pump with a tube diameter suitable for controlling the elution of about 1 mL was used in a shielded cell for separation on Eichrom resin.

Measurements in the hot cell were performed by gamma spectrometry (Canberra) and L-line X-ray fluorescence (LXF). The LXF system is an X-ray fluorescence instrument with a graphite monochromator used as a bandpass filter centered on the energy of the 14.3 keV L-line of Pu.

Separations in the laboratory were carried out on a nuclearized glove-box HPLC system (Fig. 1) composed of an Agilent 1200 series quaternary pump, an Agilent 1200 series vacuum degasser, a Perkin-Elmer 610TR flow scintillation analyzer, an Ocean Optics HR4000 spectrophotometer, an Ocean Optics DH2000 light source, SEDI HCG400 optical fibers, and an Ocean optics 10 mm Z-flow cell. A GV Instruments VG54 thermal ionization mass spectrometer (TIMS) was used. All reagents were analytical grade.

3.2 Dissolution of irradiated pellets

Two irradiated pellets were dissolved by the Laboratory for Dissolution Studies (LED) in the C11/C12 shielded line of the Atalante complex (CEA Marcoule). The hot cell was cleaned prior to each dissolution by performing the dissolution protocol without any active material and analyzing the solution to evaluate the residual radioactivity level in the cell. The measured gamma activity was below $10^{-2}\%$ of the gamma activity of the dissolved pellet.

The mass of $\text{AmO}_{1.62}/\text{MgO}$ pellets for each dissolution was about 0.5 g. At each step in the dissolution protocol, the

exact masses were recorded (pellet mass, added nitric acid mass, *etc.*). The final dissolution solution volume was about 70 mL.

3.2.1 First dissolution

The first dissolution was carried out for 4 h in boiling 4 N nitric acid. The dissolution solution was filtered; many undissolved particles visible to the naked eye were recovered on the filter. Dissolution was not complete, and therefore the solution characterization is not representative of the actual Am, Cm and Nd concentrations in the pellet.

3.2.2 Second dissolution

The second dissolution was performed in a mixture of 4 N nitric acid and 0.1 N hydrofluoric acid, a chemically more aggressive solution, to ensure quantitative dissolution of the pellet. The insolubles recovered after filtration were washed with a solution of 11 N HNO_3 and 0.1 N HF to eliminate any silica remaining from chemical etching of the glassware by hydrofluoric acid. The undissolved mass after scrubbing represented about 2.5 wt. % of the irradiated pellet. Analysis of this solution yielded a quantitative assay of 97.5% of the dissolved pellet.

3.3 Initial high-activity analyses

Two filtered solution samples were sent by pneumatic transfer to the CBA shielded analysis line. The initial high-activity analyses were carried out without any prior sample treatment (Table 1) to obtain the approximate concentrations of the major elements of interest for comparison with the values estimated from neutronic simulation codes. This step was particularly important to finalize the development of the

Table 1. First analyses in shielded line.

Elements analyzed (\pm measurement uncertainty)	1 st dissolution	2 nd dissolution	Analysis technique
U ($\pm 30\%$)	6 ± 2 mg/L	25 ± 8 mg/L	X-ray fluorescence ^a
Pu ($\pm 5\%$)	138 ± 7 mg/L	807 ± 40 mg/L	Spectrophotometry
Am ($\pm 10\%$)	17 ± 2 mg/L	97 ± 10 mg/L	ICP-AES
Nd ($\pm 10\%$)	32 ± 3 mg/L	34 ± 3 mg/L	ICP-AES
H ⁺ ($\pm 5\%$)	3.8 ± 0.2 M	4.3 ± 0.2 M	Potentiometry
Density ($\pm 5\%$)	1.144 ± 0.057 g/mL	1.154 ± 0.058 g/mL	Weighing

a: Because spectral interference, a selective separation of U in a main Pu flow [7] was need.

Table 2. Comparative orders of magnitude of U, Pu, Am, Cm and Nd concentrations between two dissolution solutions: UOX 2 (cooled for 5 years) and ECRIX-H (cooled for 3 years).

Approximate concentration (mg/L)							Mass ratio	α activity ratio
Solution	U	Pu	Am	Cm	Nd	Mg	Am/Cm	Cm/Am
UOX 2	235×10^3	2800	150	23	1340	ε	6.5	~ 4
ECRIX-H	25	800	100	135	35	400	0.7	~ 60

analysis protocols and to guarantee that the quantities implemented are sufficient for the intended analytical techniques. Moreover, these values were indispensable for defining the protocol for tracing the elements of interest in the dissolution solution.

The volumes of both dissolutions were practically the same, allowing the concentrations of the two columns to be compared. The U, Pu and Am concentrations are visibly much lower in the first dissolution solution than in the second: there ratio between the concentrations for Pu and Am was about 6 : 1 (only about 4 : 1 for U). These initial results suggest that Pu and Am (and U?) could be colocalized in the insolubles. This hypothesis was supported by electron probe microanalysis (EPMA) and X-ray diffraction (XRD) observations [3, 4] indicating colocalization of Pu, Am, and Cm in a PuO_x phase.

3.3.1 Particularity of the matrix

Based on the initial results in Table 1 and the values calculated from neutronic simulation codes, it is interesting to compare the concentrations in the dissolution solutions for a spent UOX fuel pellet and for an ECRIX-H pellet (Table 2).

Typically for a sample from a spent fuel dissolution solution, a 1 mL fraction after dilution by a factor of 1000 is allowable in an intermediate-level laboratory. In the case of the ECRIX-H dissolution solution, such a dilution would result in concentrations of about 35 μ g/L for some elements such as Nd. This implies that only about 35 ng of matter would be transferred to the laboratory in a 1 mL solution sample. This quantity is too small for satisfactory quantification and repeatability, as TIMS measurements require at least 300 ng of matter per element for an analysis comprising five 10-cycle blocks. Moreover, the Mg-rich matrix of the ECRIX-H pellets can hinder HPLC column separation by cation exchange, which is performed before TIMS analysis.

The dose rate of the initial ECRIX-H dissolution solution is due to the presence of fission products, but also to

emitters that are generally found at ultralow concentrations in “typical” dissolution solutions: ²⁴²Am and ²⁴²Cm.

We therefore sought to identify a protocol capable of addressing these two criteria: diminish the sample dose rate (*i.e.* decontaminate the solution of the main gamma emitters); and eliminate the Mg from the matrix.

3.4 Isotopic analysis

3.4.1 Shielded cell separations

Two separation routes were selected:

- Separation on Biorad AG1X8 resin to retain U and Pu (the major elements in ECRIX-H); U and Pu were then eluted separately;
- Separation on Eichrom DGA resin to retain the trivalent elements; this resin retains the lanthanides (Ln) and Am/Cm, but not Mg and the main gamma emitters such as Cs.

The optimized protocol shown in Fig. 2 calls for the following remarks:

- The protocol is applied directly to a sample of the dissolution solution (without prior dilution).
- No valence adjustment for U and Pu is implemented. In HNO₃ 8 M media, Pu is mainly Pu(IV) and U is mainly U(VI).
- Neutralization of fluoride by a concentrated aluminum nitrate solution (as in dissolution No. 2 in the nitric + hydrofluoric acid medium) is not used. U and Pu are partially complexed by fluoride and thus not retained on the AG1X8 column. Given its high concentration, traces of U (and Pu in lesser extent) can be present in the Ln stream.
- However the valence adjustment by a ferrous sulfate solution and the fluoride neutralization by an aluminum nitrate solution would be possible without disturbing DGA performance. Fe(II)/Fe(III) and Al(III) have a low uptake by DGA [8, 9]. We chose to minimize the addition

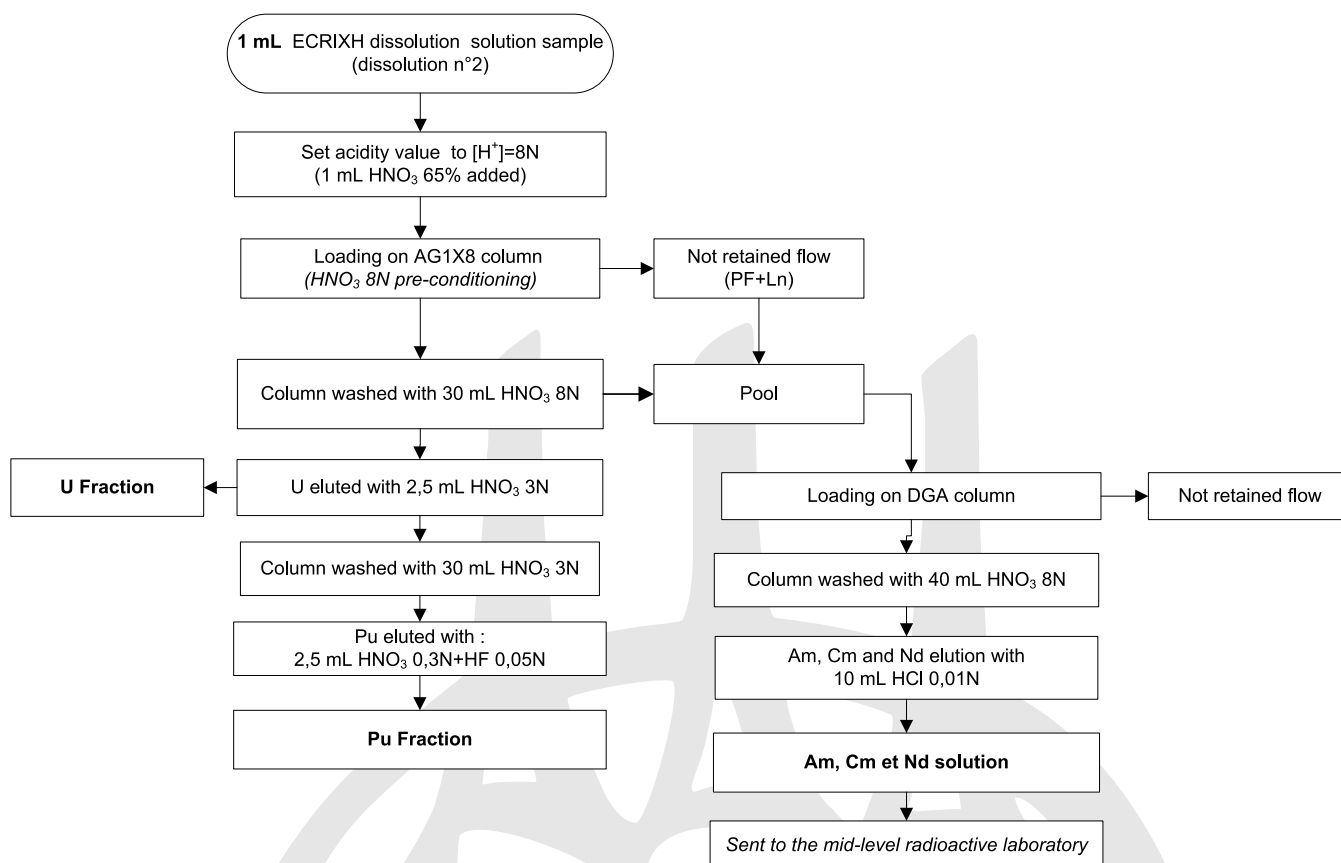


Fig. 2. Separation protocol implemented in a high-activity cell.

of reagents in hot cell in order to make easier the waste management.

- Analysis of the solution obtained by washing the AG1X8 column with 30 mL of 8 M HNO_3 showed the importance of this step: the solution contained lanthanides.
- Some tests were realized to evaluate distribution coefficient of Mg on DGA resin in our experimental conditions. Thus, 0.2 g of DGA resin and 2 mL of 100 mg/L Mg(II) in 8 N HNO_3 were mixed during 5 min. Mg was analyzed in the supernatant. These tests confirmed that Mg is not uptaken by DGA [8, 10].

For the ECRIX-H dissolution solutions the overall Am and Nd recovery efficiency of the protocol was estimated near 100% for both elements. The residual Mg in the Ln fraction was less than 1 wt. %.

Gamma spectrometry measurements were used to estimate a ^{137}Cs decontamination factor exceeding 10^4 .

The different fractions obtained by this protocol could be transferred to the laboratory without restriction.

3.4.2 Glove box separations

In the laboratory, the sample containing Am, Cm and Nd was again passed through AG1X8 resin to eliminate any residual Pu traces that could interfere with HPLC separation. The protocol is shown in Fig. 3.

A typical Am/Cm separation is illustrated in Fig. 4. The method used [11] was developed for solutions arising from the dissolution of spent UOX or MOX fuel after 5 to 10 years

of cooling, in which the Cm/Am activity ratios did not exceed 10 : 1 and the Am/Cm mass ratios ranged from about 4 : 1 to 8 : 1. In the case of the ECRIX-H pellet dissolution solution, the Cm/Am activity ratio was very high ($\sim 60 : 1$) whereas the Am/Cm mass ratio was near 0.7 : 1. The difference between the solution compositions explains why the method used was unable to satisfactorily resolve the Am and Cm peaks for the ECRIX-H dissolution solution.

The decontamination of Cs was particularly effective: no radiometric signal was detected except for Am and Cm.

Although Cm fractionation was performed without difficulty, the fraction containing Am (and Cm) was reinjected in the HPLC column for a second purification.

The separation obtained is shown in Fig. 5. This time the resolution of the Am and Cm peaks was excellent and the fraction containing Am was pure. Resolution was calculated with

$$R_s = \frac{1.18 (t_2 - t_1)}{w_{\frac{1}{2},1} + w_{\frac{1}{2},2}} \quad (1)$$

where $w_{1/2}$ was estimated by Peak Analyser module of Origin[®] software. The values are reported in Table 3. Supposing that efficiency of radiometric detector was very close for Am and Cm counting, then their peak areas can be directly compared. Am and Cm specific activities were estimated to 2.6×10^{12} Bq/g Cm and 4.9×10^{10} Bq/g Am (isotopic results were used). From Fig. 5, peak areas ratio was about $A_{\text{Cm}}/A_{\text{Am}} \sim 2.3$. Thus mass ratio in the reinjected fraction was estimated to $m_{\text{Cm}}/m_{\text{Am}} \sim 4.3 \times 10^{-2}$. Cm mass represented about 4% of Am mass in reinjected fraction, and

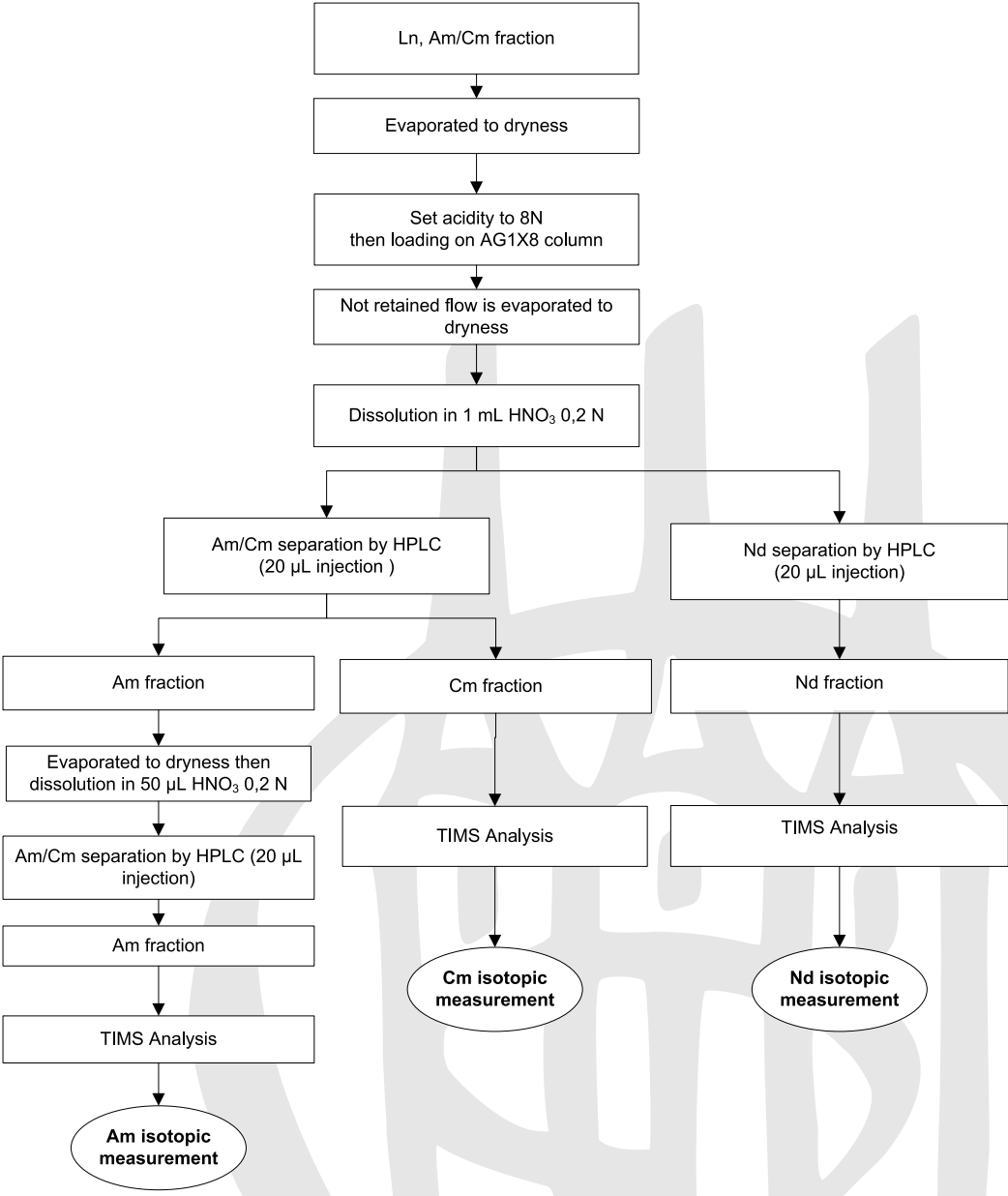


Fig. 3. Separation protocol implemented in a laboratory glove box.

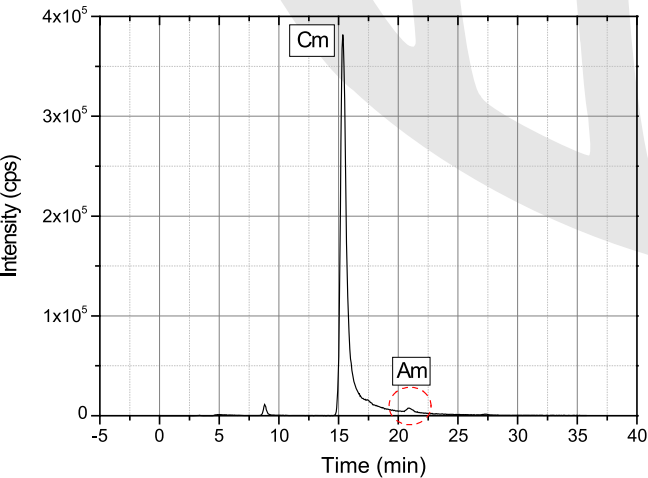


Fig. 4. HPLC chromatogram of the first injection of the (Am, Cm, Nd) fraction. Column: Nucléosil 100-5SA 250 × 4; Eluent: 0.1 M HMB adjusted to pH 3.6 with NH₃; Elution rate: 0.8 mL/min; Injection loop: 20 µL; Detector: FloOne 610TR (Perkin-Elmer); Detection cell: heterogeneous glass 82.5 µL; Energy range: 0–2048 keV.

Table 3. Calculated resolution values between Am and Cm peaks.

Separation	Peak name	Retention time <i>t</i> (min)	Height <i>H</i> (graph unit)	Area	Width at half-height <i>w</i> _{1/2} (min)	Resolution <i>R</i> _S
Fig. 5	Cm	15.8	2412	2027	0.677	4.46
	Am	20.9	1087	891	0.673	
Fig. 6	Cm	13.2	1.44	—	0.520	1.17
	Am	14.3	1.11	—	0.590	

relative mass ratio ($m_{\text{Cm isotope}}/m_{\text{Am isotope}}$) were about 6.5 ‰ to 242 mass and 7.6 ‰ to 243 mass. This second purification is debatable because benefit on measure is just in precision limit of TIMS.

Nd was separated by reversed-phase chromatography (Fig. 6) on the basis of the protocols described in [12–14]. The column used C18 as the stationary phase and the eluent

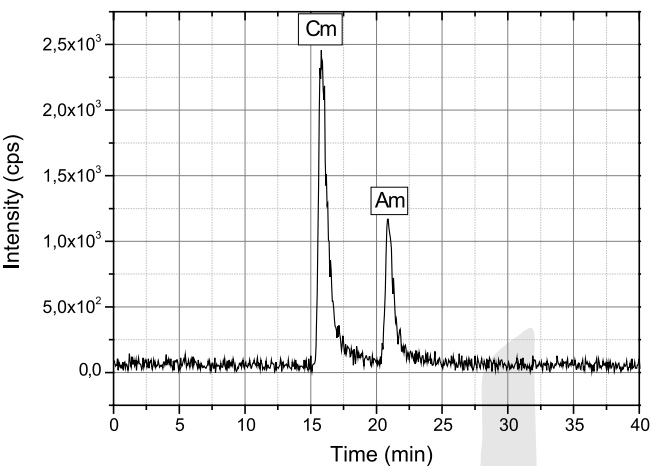


Fig. 5. HPLC chromatogram of the second injection of the (Am/Cm) fraction. Same operating conditions as in Fig. 4.

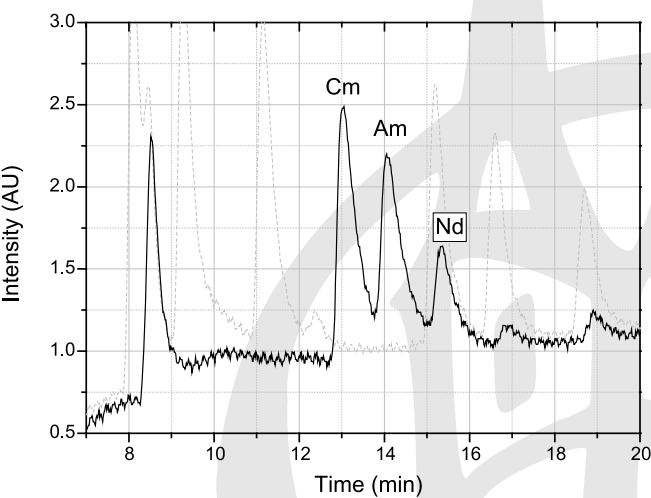


Fig. 6. Chromatogram of Ln separation from an ECRIX-H pellet dissolution solution after treatment (refer to text). The dotted line represents the injection of a surrogate Ln solution. Elution: 0.09–0.23 M HIBA in 20 min in the presence of 0.01 M octanesulfonic acid adjusted to pH 3.7 with NH₃; Column: Vydac 201TP5415 150 × 4.6; Elution rate: 1 mL/min; Injection loop: 20 µL; Post-column derivation: PAR 0.05 g/L in 1 M acetic acid + 2 M NH₃. PAR flow rate: 0.3 mL/min. Detector: HR4000 (Ocean Optics) at 520 nm; Detection cell: Z-flow with 1 cm optical path.

was α -hydroxyisobutyric acid (HIBA) with *n*-octanesulfonic acid as a modifier. Detection was performed by spectrophotometry after adding a metallochromic complexing agent, PAR (4-(2-pyridylazo)resorcinol). Some parameters (elution rate, pH of eluent, gradient separation) were adjusted to optimize the resolution of Nd with Ln adjacent bands (potential isobaric interferences). Indeed Nd injected mass was more important (from 10 to 50 times) than one used in [12–14]. Direct fractionation of the flow detected by spectrophotometry would result in the presence of PAR in the sample and hinder the process of deposition on the filaments for TIMS analysis. Fractionation of part of the flow between the column outlet and the point of PAR addition was investigated. Under these conditions it is very difficult to sample exactly what is detected by spectrophotometry (different transit times for the two channels, allowance for the residence time and dead volume of the spectrophotometry cell, etc.). Given the difficulties involved in performing simultaneous

fractionation with a detection channel, but considering the good repeatability of the separations, we decided to implement “blind” fractionation. After identifying the retention time of the peaks of interest in the first separation (Fig. 6), we conducted a second separation in which PAR was replaced by 10^{−3} M nitric acid at the same flow rate.

The Nd and Am peaks were poorly resolved, but as Am does not create isobaric interference with Nd, its presence does not hinder the isotopic determination of Nd. Mg was eliminated from solution sufficiently that it did not interfere with the chromatogram and column performance. We can notice that Am and Cm are also separated by this method. Resolution was calculated for Am and Cm adjacent bands (Table 3). These results highlight that the quality of the separation between Am and Cm is better with the first method. However, an optimization study should allow to improve the resolution between Am and Cm of the second method. This work is already scheduled in our laboratory.

The preparation of injected sample was imposed by required mass quantity for TIMS measurement. Then, the calculus is in function of the least concentrated element to analyze in the original sample. The global irradiation of the injected sample can become important in some case and we have noticed that the used columns performances deteriorate with time. It can be necessary to carry out a little adjustment of some parameters to get a good separation back.

The isotopic compositions were determined for Am, Cm and Nd (Tables 4 to 6). A scan of surrounding mass values confirmed absence of isobaric interference. The results of each dissolution are the average of two independent measurements.

The estimated uncertainties on the isotopic results were based on the known performance of the method, and on the experience acquired in the laboratory and on prior validation data. Isotopic reference standards are routinely measured to check the instrument response.

Table 4. Am isotopic assay (at. %).

	1 st dissolution	2 nd dissolution	Neutronic simulation after 3.5 years of cooling	4 years of cooling
²⁴¹ Am	35.5 ± 0.04	34.17 ± 0.03	39.46	39.72
²⁴² Am	1.60 ± 0.02	1.55 ± 0.02	0.97	0.97
²⁴³ Am	62.90 ± 0.03	64.28 ± 0.06	59.57	59.32

Table 5. Cm isotopic assay (at. %).

	1 st dissolution	2 nd dissolution	Neutronic simulation after 3.5 years of cooling	4 years of cooling
²⁴² Cm	0.24 ± 0.02	0.25 ± 0.01	0.30	0.14
²⁴³ Cm	11.45 ± 0.06	11.34 ± 0.06	6.65	6.64
²⁴⁴ Cm	69.94 ± 0.07	70.02 ± 0.07	69.17	68.86
²⁴⁵ Cm	15.11 ± 0.08	15.06 ± 0.08	18.35	18.70
²⁴⁶ Cm	3.13 ± 0.03	3.17 ± 0.02	5.26	5.38
²⁴⁷ Cm	0.14 ± 0.01	0.14 ± 0.01	0.22	0.23
²⁴⁸ Cm	< 0.05 (DL)	< 0.05 (DL)	0.04	0.04

Table 6. Nd isotopic assay (at. %).

	1 st dissolution	2 nd dissolution	Neutronic simulation after 3.5 years of cooling	4 years of cooling
¹⁴² Nd	0.74 ± 0.04	0.95 ± 0.10	0.85	0.84
¹⁴³ Nd	19.97 ± 0.02	19.69 ± 0.10	18.69	18.65
¹⁴⁴ Nd	27.53 ± 0.03	27.58 ± 0.14	28.94	29.07
¹⁴⁵ Nd	15.19 ± 0.02	15.09 ± 0.08	14.89	14.86
¹⁴⁶ Nd	18.97 ± 0.02	19.08 ± 0.10	19.22	19.19
¹⁴⁷ Nd	< 0.1 (DL)	< 0.1 (DL)	0.00	0.00
¹⁴⁸ Nd	10.81 ± 0.01	10.86 ± 0.05	11.00	10.99
¹⁵⁰ Nd	6.78 ± 0.03	6.76 ± 0.07	6.41	6.40

3.5 Quantitative analysis

Analysis by isotopic dilution was chosen for quantitative determination to ensure highest possible accuracy. Isotopic dilutions were carried out on samples of the high-activity dissolution solution. Source tracing takes into account possible variations in the dissolution solution (concentration by evaporation, for example), as well as the efficiency of the different separation steps implemented prior to TIMS analysis.

Isotope dilution is subject to certain conditions, however:

- Mixing between the tracer and the sample must result in ideal chemical and isotopic homogeneity (same valence state and same physical state).
- No chemical treatment of the sample/tracer mixture must affect isotopic fractionation.

The tracers were selected and adjusted so that after addition the major isotope in the tracer was present in the same proportion as the major isotope in the sample (Table 7). Prior sample dilution by a factor of 250 was necessary to trace Cm because its concentration was too high with respect to the available tracer concentration.

All dilutions were carried out by weighing. Solution tracing was performed twice. The separation protocols were identical to those used for isotopic analysis.

The results calculated from the general equation for isotope dilution are expressed as a mass concentration of the element in the irradiated pellet dissolution solution before or after dilution. Based on the mass of the final filtered dissolution solution (from which the analysis samples were taken), the dissolved pellet mass, and the dilution factor, the results are expressed in milligrams per gram of initial heavy metal (mg/g_{ihm}) in Table 8.

The uncertainties were calculated by applying the law of propagation of uncertainty. Only the uncertainty on the

Table 7. Tracers and isotopic ratios obtained in the sample-tracer mixture.

Major isotope in tracer	Estimated isotopic ratio in sample + tracer solution
²⁴¹ Am	241/243 = 0.90
¹⁴⁵ Nd	145/144 = 1.22
²⁴⁸ Cm	248/244 = 1.12

Table 8. Concentration of heavy nuclides in the pellets at the end of irradiation: measured and calculated values.

Element	Concentration (mg/g _{ihm})		
	Measurement after 3.5–4 years of cooling	Neutronic simulation after 3.5 years of cooling	Neutronic simulation after 4 years of cooling
Am	63.4 ± 0.81	79.1	79.4
Cm	99.2 ± 5.3	96.7	95.2
Nd	29.3 ± 0.29	34.7	34.7

initial Am content of the pellet (16.65 wt. %) was disregarded.

The Cm content was also determined by alpha spectrometry. Based on the measured isotopic composition, the Cm concentration was estimated at 98.7 ± 11.7 mg/g_{ihm}. This result is in very good agreement with the value indicated in Table 8.

On the basis of the results in Table 8, the dissolved pellet mass, and the initial Am mass ratio of 16.65%, the Am transmutation efficiency is estimated at 93.7%.

4. Conclusion

Two irradiated ECRIX-H pellets were dissolved in view of two distinct objectives.

Concerning the reprocessability of pellets containing americium, a large quantity of insolubles can be observed (about 10 wt. % of the initial fuel).

The second objective was addressed by the dissolution of a pellet in nitric + hydrofluoric acid to characterize some elements of interest (Am, Cm and Nd) in order to consolidate the evolution codes and determine the fission and transmutation rates.

Separation protocols were implemented in a high-activity cell to diminish the sample dose rate while transferring sufficient quantities of the elements to be measured. Additional separations were then performed in the laboratory.

Isotopic and quantitative analyses were carried out for Am, Cm and Nd. The values obtained were in very good agreement with the estimates given by the evolution code.

The Am transmutation rate was estimated experimentally at 93.7 at. %, which is slightly higher than the value obtained by neutronic simulation *i.e.* 92.6 at. %. The same trend was also observed by means of radial EPMA analysis on pellets [3].

Dissolution tests and analysis on two UO₂ pellets irradiated in the same pin will be performed later to quantify precisely the neutron fluence accumulated by the Am_{1.62}/MgO pellets and rejustify the simulation code calculation.

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