Experimental Assessment of Incineration Rates of Actinides in High Intensity Neutron Fluxes

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I Introduction

The Mini-Inca project [1] has been approved by the French Atomic Energy Center (*CEA*) to provide experimental evidences about the nuclear waste transmutation. Even if the reference solution in France is deep underground storage, the neutron-induced transmutation of actinides is still a promising way to reduce the long-term radiotoxicity of radioactive nuclear waste and improve the safety of geological disposal.
For this purpose, the scientific community is mainly considering advanced critical reactors or accelerator driven systems (*ADS*). A more efficient use of the nuclear fuel and innovative fuel cycles constitute a promising way to reduce the amount of nuclear waste and to increase the safety margin against transients of power [2].
The transmutation potential of any system is directly related to its neutron spectrum and to the corresponding nuclear parameters of the most important isotopes present (see for instance [3]). These two parameters are essential to assess both the economical viability of transmutation, and the comparative merits of each system. For some actinide isotopes, the most complete nuclear data libraries (*ENDF-B*, *JEF*, and *JENDL*) show discrepancies [4]. These nuclei usually are negligible in the conventional fuel cycle, but they are essential in a transmutation system where they will dominate the fuel composition. Due to the large number of rare isotopes playing a major role in the innovative fuel cycle, and to the difficulty to carry out comprehensive differential measurements, integral experiments are necessary to asses the quality of the existing nuclear data libraries and to validate the computing codes.

II Mini-Inca

II.a Experimental methods

The Mini-Inca project develops new experimental facilities and computational methods to carry out integral measurements of actinide transmutation in given irradiation conditions. The experimental part consists of the determination of nuclear parameters (mainly capture and fission cross-sections) and measurement of the transmutation potential in definite neutron spectra. Integral measurements will be performed after irradiations characterised by:

- low mass samples in order
  - to be non perturbative for the neutron flux;
  - to have low activities;
  - to have pure sample of rare isotopes;
- a set of methods to determine the composition of the sample:
  - from quasi on-line spectroscopy to off line mass spectrometry, on-line alpha spectroscopy and back-to-back fission micro-chambers [5];
- different high intensity neutron fluxes:
  - representative of future transmutation systems;

- to have access to short lived isotopes by multiple neutron capture;
- to have access to small formation cross section isotopes;

- activation monitors and new on-line detectors to determine the neutron flux.

II.b Experimental setup

Two types of irradiations are foreseen:

- short irradiations to have a precise determination of unknown nuclear parameters, as neutron capture and fission cross sections including branching ratios;
- long irradiations of mono-isotopic sample or known mixtures of isotopes to determine transmutation rates in given high intensity neutron spectra.

Irradiations will be carried out in the ILL 58MWth reactor in Grenoble. Its fuel element of 9 kg uranium enriched at 93%, is at the centre of a pool a 2.5m diameter of heavy water. The flux available for our experiment the highest in the world for experimental purpose [6].

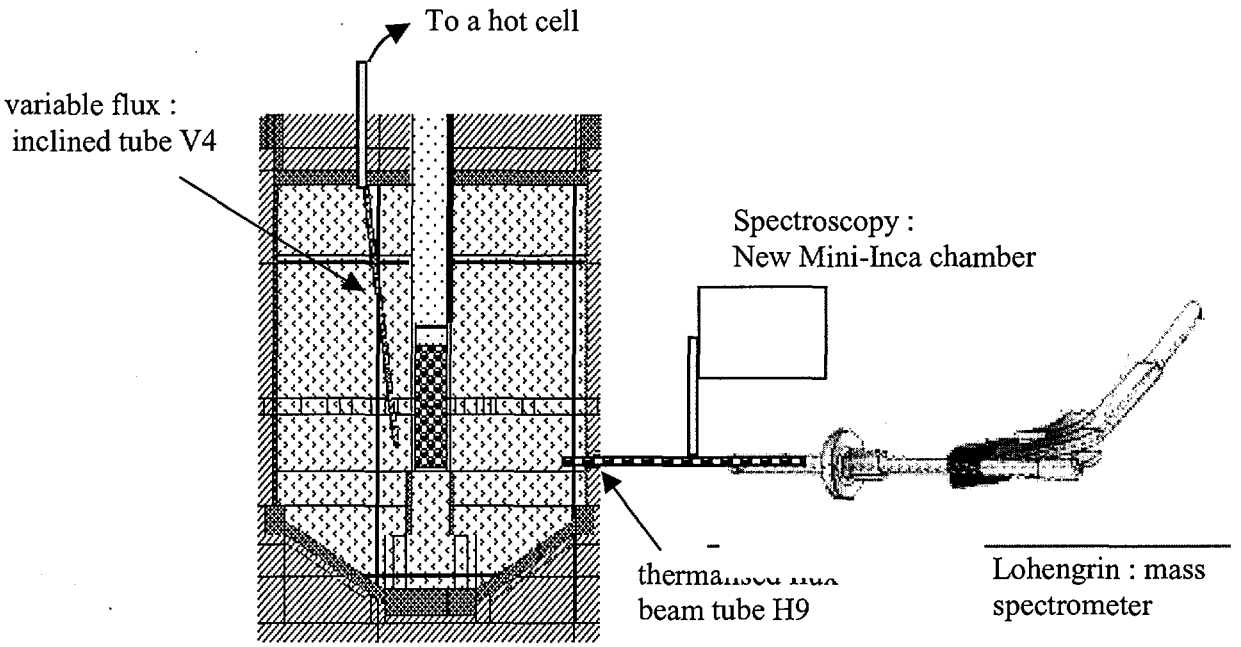


Figure 1: Schematic view of the ILL reactor and of the experimental setup.

For the Mini-Inca experiments, two beam tubes named H9 and V4, will be used. They are shown in the Figure 1. The inclined V4 channel gives access to thermal and epithermal neutron spectra of $2 \cdot 10^{15}$ n·cm^{-2}·s^{-1} at a distance of 12 cm from the core. The H9 tube has 98%-thermal spectrum of about $5 \cdot 10^{14}$ n·cm^{-2}·s^{-1}. The very high intensity of these two neutron fluxes is essential for the success of the experiments. While V4 will essentially give information about macroscopic transmutation potentials, the H9 tube will be used to determine nuclear parameters at the thermal neutron energy. Both of them will contribute to validate computer simulations of transmutation process.

II.c The sample

The actinides that will be studied are chosen for the poor knowledge of their nuclear parameters and the importance they could have in a real transmutation system.

The parameters that can be chosen and controlled in our experiment are the initial composition of the sample and the irradiation time. The observable is the final composition of the sample, determined by $\alpha - \gamma$ spectroscopy or mass spectrometry. During the irradiation, there will be fission fragments created inside the sample and their high activity can constitute a major problem for nuclear spectroscopy. Consequently, the sample has been designed to avoid this background and the resulting counting-rate problems. The detailed geometry of the sample is presented in the Figure 2.

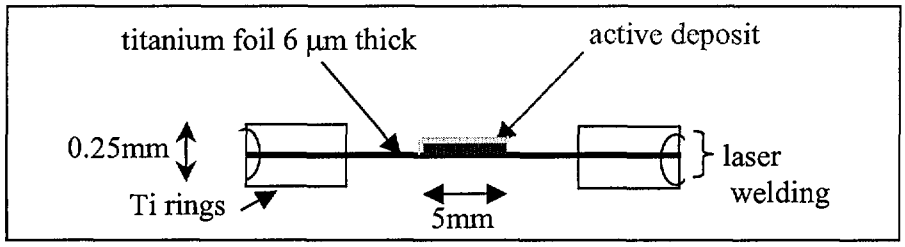


Figure 2: The sample backing in Ti.

The active material is electrodeposited on a 6μm thick titanium foil welded by a laser beam between two titanium rings of 0.25mm total thickness. Monte-Carlo simulations [7] show that fission fragments generated in the active deposit can pass through the backing and leave the target. A few tens of angstrom gold coating is made on the active deposit to reduce the sputtering of the target material by fission fragments. A number of samples such as ^{231}Pa, $^{229\text{-}232}$Th, $^{233\text{-}235}$U, ^{237}Np, $^{239\text{-}241\text{-}242}$Pu, $^{241\text{-}243}$Am, $^{243\text{-}244\text{-}245\text{-}248}$Cm and ^{249}Cf will be available.

II.d Neutron flux monitor for H9

In activation experiments the determination of the neutron spectrum is a key parameter for the quality of the result and the associated uncertainties. The shape of the thermal neutron flux is well known for H9 and consequently the spectrum is entirely determined by an integral measurement of its intensity by neutron activation analysis. A neutron monitor will be irradiated together with each transuranic sample and it will be analyzed by gamma spectroscopy inside the Mini-Inca chamber.
Considering the experimental conditions of H9, we will use an alloy Al-1%Co from the Institute for Reference Materials and Measurement in Geel (Belgium). A 6mm diameter disc, 0.1 mm thick, will be used, providing counting rates below 10000 cps.

III Commissioning of the Mini-Inca Chamber

III.a Presentation of the chamber

A new detection system named Mini-Inca chamber have been developed and installed at the ILL reactor. It allows accurate alpha-gamma spectroscopy just after irradiation and even between successive irradiations of the same sample. The advantages of $\alpha - \gamma$ spectroscopy to determine the composition of the sample are that it is fast, it needs no chemistry and it is non-destructive. This sophisticated vacuum chamber allows to take the sample from the irradiation system of H9 and to position it in front of the detection system with an absolute precision of 0.2 mm. The detectors can be moved to choose the most suitable solid angles and consequently the counting rates. The chamber and the detectors are shielded by a 5 cm thick

lead wall and by a combination of borated polyethylene and B_4C to reduce the gamma and neutron background. In Figure 3 the chamber and the detectors are shown.

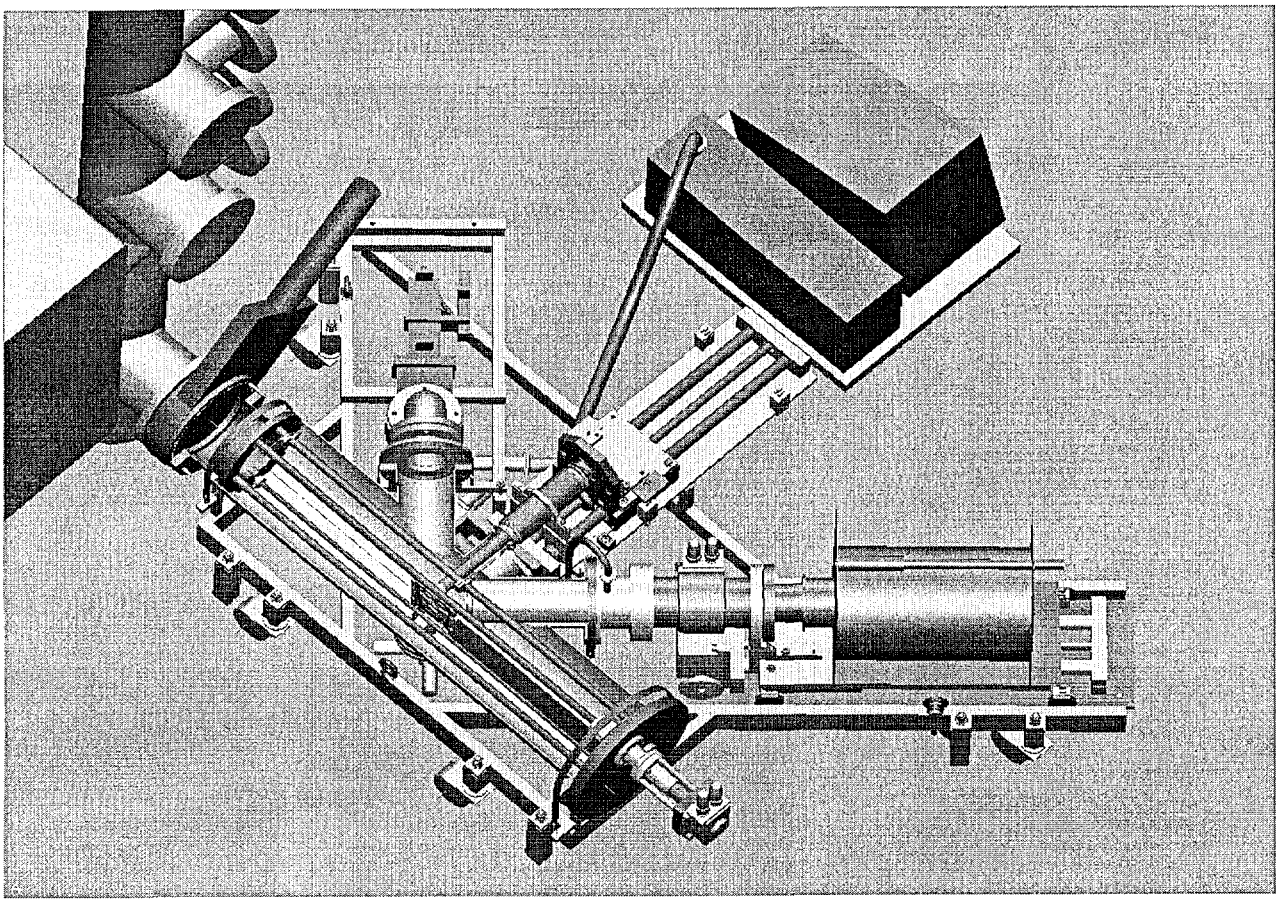


Figure 3 : inside view of the Mini-Inca chamber

The gamma detector is a high resolution coaxial germanium detector with a relative efficiency of 15%. A 0.5mm thick carbon-epoxy end-cup allows to have a wide energy range of gamma detection from about 20 keV to several MeV. It has a resolution of 0.825 keV (FWHM) at 122 keV and 1.8 keV at 1.33 MeV with a peak/Compton ration of 46:1. The detector (with its dewar) can move from 40 to 80 cm and in this way the fraction of solid angle, $(\Omega/4\pi)$, can be varied from 2.10^{-3} to 3.10^{-4}. The alpha detector is a high resolution PIPS (Passivated Implanted Planar Silicon) of active area of 100 mm^2 depleted on 300 μm. The resolution is 11.8 keV for 5.486 MeV alpha particles. It can move from 0.6 cm ($(\Omega/4\pi) = 0.1$) to 30 cm ($(\Omega/4\pi) = 10^{-3}$) from the irradiated sample.
Due to the high counting rate (>50 kHz) foreseen in a number of measurements, a dedicated high speed electronics is used. All signals are handled by a Digital Signal Processor after the pre-amplification stage. A transistor-reset preamplifier is used to suppress pile-up effects on the resolution of the Germanium detector. The preliminary test of the complete spectroscopic system have been carried out in autumn 2000. Several measurement with calibrated alpha and gamma sources will permit to obtain the relation between the target position, the experimental efficiency ($\Omega\varepsilon$) for each energy and the uncertainties associated.

III.b First experiments at ILL-Grenoble

The commissioning of the Mini-Inca installation on H9 required a number of preliminary measurements to characterise precisely the experimental environment. Different parameters as the contamination level of the beam tube, the thermalisation rate of the neutron spectrum, the count-rates and electronic linearity, as well as the sputtering of the target material have been measured.

Initially a dummy target without any active material has been irradiated to give a reference for the background estimation for different irradiation times. Then we have irradiated a ^{242}Pu sample, results are given in following paragraph. Two different flux monitors, Au and Co with and without a cadmium shielding, have still to be irradiated to determine the thermalisation rate of the neutron spectrum (see [8] for details). A sample of ^{239}Pu will also be studied to qualify the experimental methods by measuring its capture and fission cross sections. These values have been widely measured and this will provide a valuable benchmark for our experimental method.

Details on the possible measurements in H9 with the Mini-Inca installation can be found in reference [4] and two examples are described below.

III.c ^{242}Pu and ^{243}Am : presentation

The aim of the irradiation of ^{242}Pu is to determine the capture cross section of ^{242}Pu and ^{243}Am [4]. They are both very important for transmutation system based on molten salt and graphite moderated [9] . A schematic view of the evolution of ^{242}Pu and ^{243}Am in a thermal neutron spectrum is given in Figure 4.

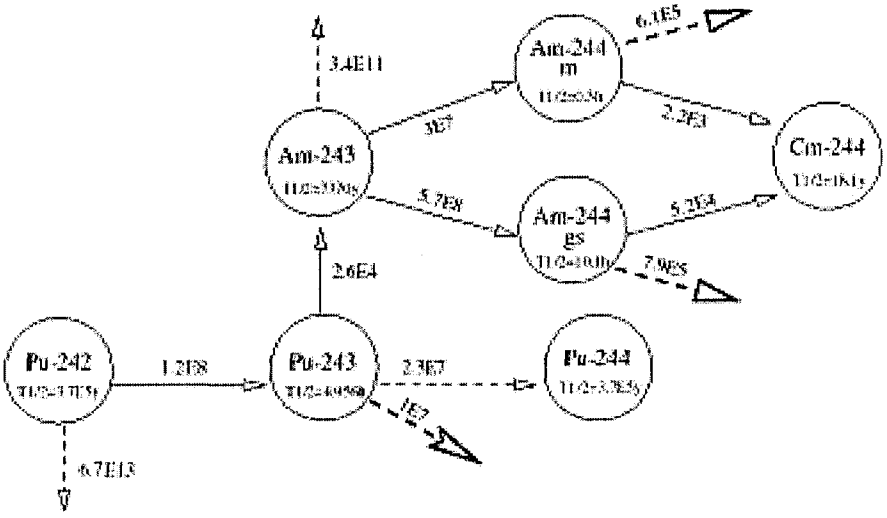


Figure 4: Evolution of ^{242}Pu and ^{243}Am in a thermal neutron flux

The thermal neutron capture cross section of ^{242}Pu can be measured by irradiation of a sample of 10 µg for two days in H9. After a cooling time of two days, the ^{243}Pu formed by neutron capture will be transformed by β-decay into the alpha emitter ^{243}Am. By measuring the alpha lines at 5275 and 5233 keV, the ^{242}Pu neutron capture cross section will be determined.

The irradiation of a 10 µg sample of ^{243}Am during 4 hours will provide missing nuclear parameters on its transmutation chain. A gamma spectroscopy measurement of the 985 keV line from ^{244m}Am for 3 hours just after irradiation will allow us to determine the half-life of the ^{244m}Am and the cross section of ^{243}Am(n,γ)^{244m}Am. After a cooling time of 3 days, the ^{244m}Am will be completely disappeared. A gamma spectroscopic measurement of the 154, 744 and 898 keV lines from ^{244gs}Am will allow us to determine the cross section of ^{243}Am(n,γ)^{244gs}Am.

IV The ^{242}Pu experiment

The first experiment, with ^{242}Pu was carried out in November 2000. A sample containing 9.77μg of ^{242}Pu and a flux monitor of 7.46mg of alloy were together irradiated two days Here we present preliminary results which have still to be completed and verified.

IV.a Background measurement

To validate the experimental installation we have irradiated two dummy targets (i.e. empty target frame) for two hours and also for two days. This test-experiment was performed in order to make sure that all our measurement system was working in the ILL environnement. These are the spectra γ-spectrum (on the left) and α-spectrum (on the right) obtained.

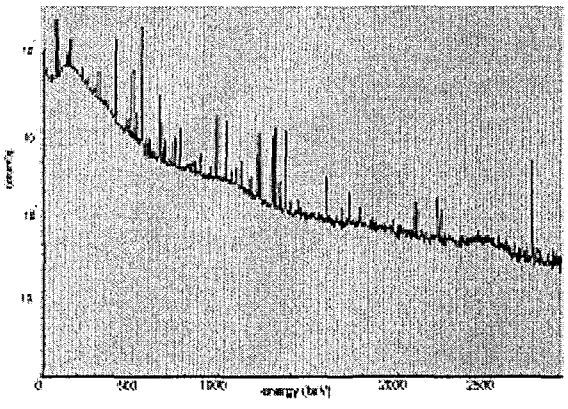


Figure 5 : γ-spectrum

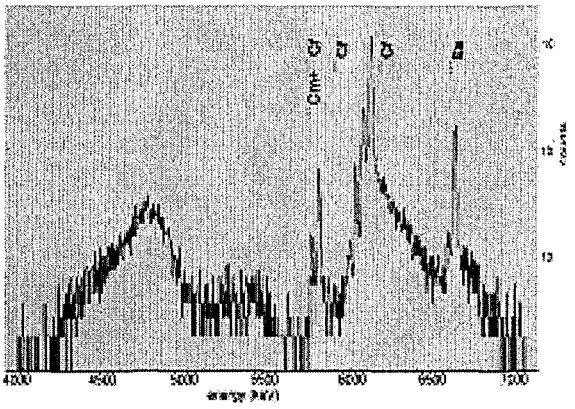


Figure 6 : α-spectrum

In the γ-spectrum we can identify usual activation products of structure elements (^{76}As, ^{122}Sb, ^{99}Mo, $^{46\text{-}47\text{-}48}$Sc, ^{24}Na) and few fission products ($^{131\text{-}132}$I...). We note that these results are consistent with the preliminary tests we made at ILL before and with the expected behaviour of our sample.

In the α-spectrum we can see an important background due to γ, β and a fews ray which are attributed to ^{252}Cf , ^{250}Cf, ^{244}Cm, ^{249}Cf and ^{253}Es, all for high α-energies. These α emitters are present due to contamination from the Lohengrin beam tube. This irradiation tube is widely contaminated by actinides exposed, to a very high neutron flux, so their presence could be understood and we see them because of their short half-live.

In conclusion, this irradiation gives a few positive answers. First on the technical solution taken for the target and chamber. Indeed our new target succeed the irradiation test without deformation, and the radioactivity did not prevent to have it in the chamber immediately after irradiation. Then on the shielding, inside and outside the chamber : the dose rates weren't too high near the instrument and we obtain quite high resolution α-γ spectra.

IV.b α–analysis of the non-irradiated target

We had some uncertainty in determining the area of the α-peaks, because of the difficulty to make good separation of neighbouring peaks. Since ^{242}Pu rays are far from others we made a simple sum of ROI and subtracted the linear background in order to obtain area of majors peaks of ^{242}Pu. The relation between the activity, A, and the area, S, is given by the classical expression:

$$S = A \cdot I_\alpha \varepsilon \cdot \left(\Omega/4\pi\right) \cdot t_c$$

With Ω : the solid angle ; $I_\alpha\varepsilon$: product intensity of the α-ray and efficiency of the detector.

Due to the complexity of the exact formula for the solid angle and the insufficiency of classical approximations, we correct it for our typical distances and use the following formula

$$\Omega_{app}(d) = 2\pi \cdot \left(1 + \frac{1}{\sqrt{1 + R^2/d^2}}\right) \cdot \left(1 - 0.024402 \cdot d^{-1.7573}\right)$$

with R: radius of our PIPS, d the distance. Our target is a 5mm diameter disc.

TheFigure 7 show the actual difference between Ω and Ω_{app} which never excess 0.7% in our range of use, between 1 and 30cm.

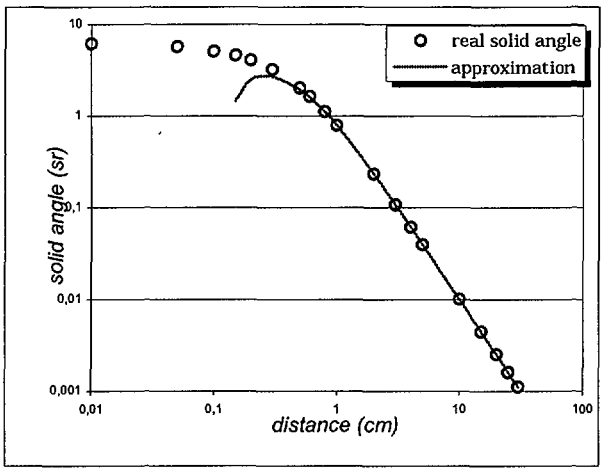
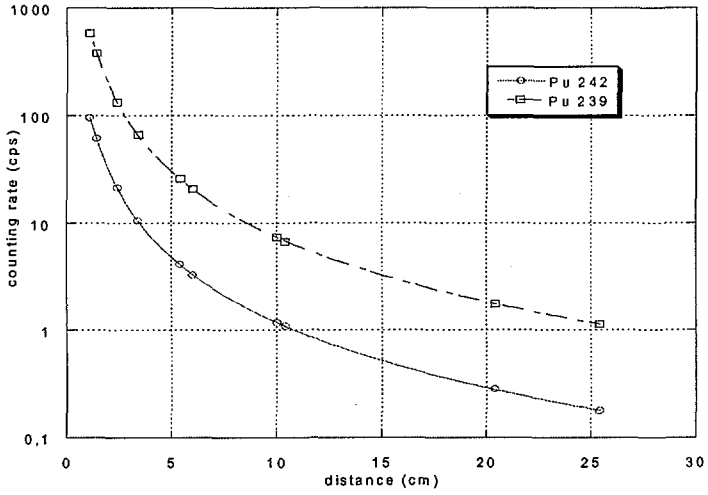


Figure 7 : Solid angle as a function of distance

The distance is obviously a very important parameter in the solid angle determination and we measured it geometrically. However we can also determine it independently from our physical measurements. By moving the detectors (changing only Ω) we have a set of points following a law proportional to Ω(d). Then a simple fit gives a new measurement of the distance providing the "activity" ($AI\varepsilon$). The difference between these distances, the "offset", gives important information on the validity of the fit. This method also reduces the uncertainty of one single measurement. It is quite similar of performing a few measurements in the same conditions and make the mean of the results. As an example, we provide detailed results for non-irradiated ^{239}Pu and ^{242}Pu target on figure 8.



target	^{242}Pu	^{239}Pu
Offset (cm)	-0.0182	0,00748
Activity (bq)	1430	9076
χ^2	11,16	12,69

Figure 8 : experimental points and fit

IV.c Target evolution during irradiation

In the classical Bateman equation we can neglect the radioactive decay of ^{242}Pu and ^{243}Am (due to their long decay time). Therefore we consider ^{243}Pu immediately decaying into ^{243}Am, so we have $N_{242Pu}(t)=N^0_{242Pu}\cdot e^{-\sigma\cdot\phi\cdot t}$ and $N_{243Am}=N^0_{242Pu}-N_{242Pu}(t)$.

As a matter of fact we observe experimentally much stronger decrease of ^{242}Pu. We can interpret this as an effect of sputtering or evaporation of our irradiated target. A disappearance term in the differential equations has to included. We have considered different hypothesis on the loss of ^{242}Pu and ^{243}Am. For instance, the loss factor for ^{242}Pu can be taken either proportional to the mass or simply constant (we can easily imagine a model with a surface evaporation, and a constant surface during irradiation). For ^{243}Am we can assume loss being either very small, or proportional to the mass of ^{243}Am or proportional to the ratio N^{243}Am /N^{242}Pu.

We have solved the differential equations corresponding of these hypothesis. Finally, it is probable that the loss of matter is either a mass phenomenon (H1 : disappearance term proportional to the number of atoms) or a surface one (H2 : disappearance term constant for ^{242}Pu and proportional for ^{243}Am). these hypothesis give the following formulas:

$$\text{H1}: \sigma\phi t = \ln\left(1+\frac{N_{243Am}}{N_{242Pu}}\right)$$

$$\text{H2}: \sigma\phi t = \frac{N_{243Am}}{N^0_{242Pu}}\cdot\frac{X}{2\cdot\left(1-e^{-X}\right)-X} \quad \text{where } X=1-\frac{N_{242Pu}}{N^0_{242Pu}}$$

IV.d Analysis of the irradiated target

In Figure 9 is shown the α-spectrum obtained after the irradiation. The cooling time was negligible and the following table shows results of peak-area for a measurement time (corrected with dead time) of 4000 minutes.

	Net Area (counts)
^{242}Pu	461387
^{243}Am	23319
$N^{242}Pu/N^{243}Am$	1006
$N^{242}Pu/N_0{}^{242}Pu$	0.696

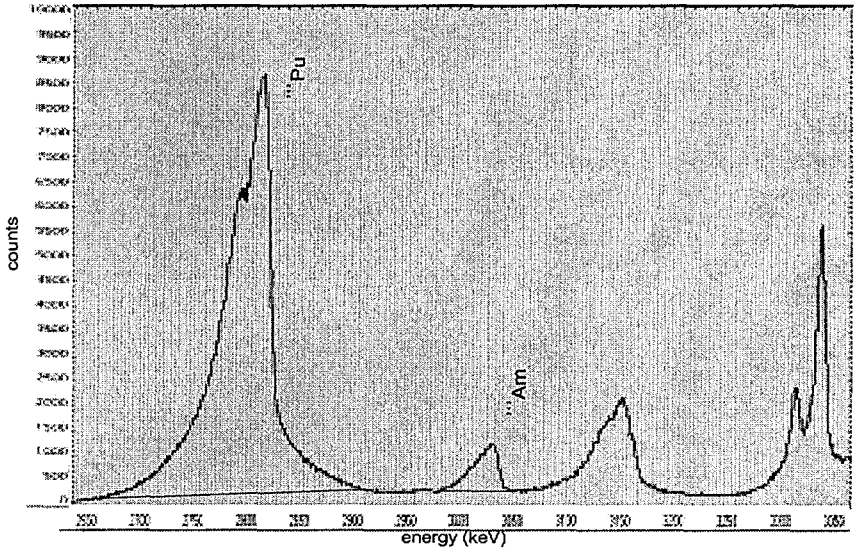


Figure 9 : α-spectrum of the irradiated target of ^{242}Pu

For a preliminary analysis we have taken a simple linear background shown on the figure and the area measured on the spectra are given in the table above.

With the formula of precedent paragraph we obtain these value for the product σϕt :

H1 : $\sigma\phi t = 9.928 \cdot 10^{-4}$

H2 : $\sigma\phi t = 9.543 \cdot 10^{-4}$

The integrated neutron flux was determined by NAA as explained in § II.d. For γ-spectra we have the same source of uncertainties plus one to correct counting-rates. the correction was needed due to high counting-rates and important dead time ; we use a pulser to re-normalise areas.

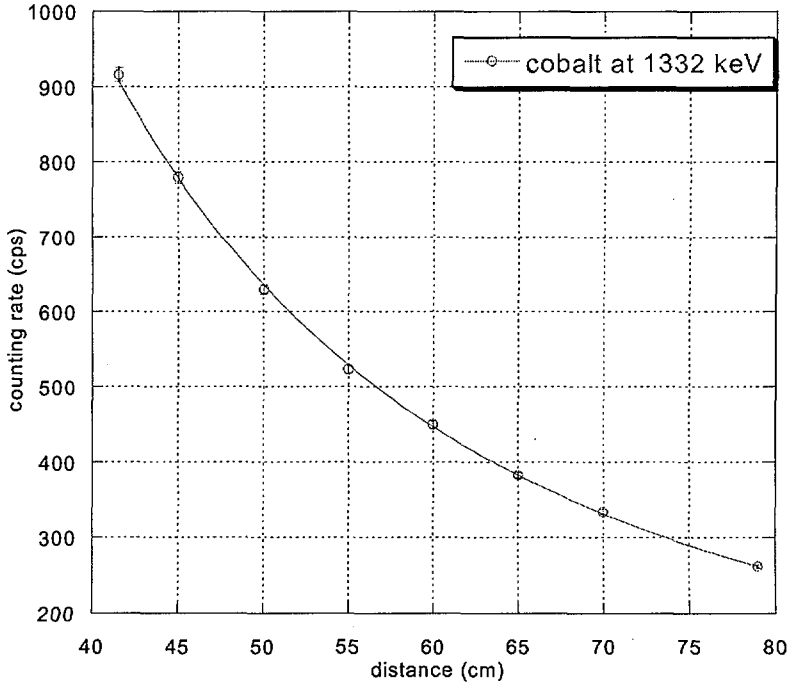


Figure 10 : determination of monitor activity

We make different measurement at various distance, and by using same methods as described before we obtain the previous fit.
Finally, we have an offset of 1.9 cm and an "pseudo-activity" (in fact it is the product $AI\varepsilon$ of IV.b) of $1.03 \cdot 10^{6}$ bq. This gives a flux of $3.36 \cdot 10^{14}$ ±6% $n \cdot cm^{-2} \cdot s^{-1}$ (see § II.d for more details)
This a quite low but the irradiation took place at the end of the cycle of the ILL reactor and just after a shut down, so this value is possible.

In conclusion depending on the different hypothesis made we find the following results for the capture cross-section of ^{242}Pu to ^{243}Pu :

H1 : $\sigma_c = 17.2b$ H2 : $\sigma_c = 16.5b$

This is very preliminary study and results that should have a uncertainty associated of about ~10%. This is in reasonable agreement with JEF2.2 [10] evaluation in a thermal spectrum of 50°C giving 16.23 barns.

Conclusion

A new portable, modular and precise tool for α-γ spectroscopy studies is now available in the frame of the Mini-Inca project. This instrument is under commissioning at the ILL-Grenoble and validation experiments have begun. Some new methods of data handling are also developed and have been presented.
In the future the same experimental methods will be applied to others isotopes in thermal (with H9), epithermal (with V4 in particular), and perhaps even fast neutron spectra.
A full set of experimental data will then be available for the transmutation potential of different spectra. This will constitute an essential step toward correct simulations of realistic transmutation systems in order to determine the conditions in which an efficient transmutation of nuclear waste is possible.

References

[1] G. Fioni et al., Proc. of the 5th Int. Information Exchange Meeting on Actinide and Fission-Product Partitioning and Transmutation, Mol (Belgium), OECD/NEA Paris (1999) 231-238.

[2] G. Fioni et al., Proc. of the Seminar on Fission, ", C. Wagemans, O. Serot and O. D'Hondt Editors, ISBN 981-02-4241-7, World Scientific, (2000) 203-214

[3] F. Lelièvre, *PhD Thesis*, Université d'Orsay-Paris XI (in French)

[4] F. Marie et al., *Integral Measurement of Capture and Fission Cross Section of Minor Actinide of Interest*, CEA/DSM/SPhN-00-50 internal report (in French)

[5] M. Fadil et al.,Proceedings of the Int. Conf. on Neutron Field Spectrometry in Science, Technology and Radiation Protection, Pisa, June 2000, Nucl. Inst.. Meth. A, in press

[6] D. Ridikas et al., *On the fuel cycle and neutron fluxes of the high flux reactor at ILL Grenoble* Proc. of the 5th Int. Specialists' meeting SATIF-5, OECD/NEA Paris (2000) in press

[7] Ziegler, SRIM code, *Copyright: International Business Machines Corporation, 1984 , 1986, 1989, 1991, 1992, 1994, 1995, 1998. All Rights Reserved*

[8] C.H. Westcott et al., *2nd Int. Conf. on the Peaceful Use of Atomic Energy*, Vol. 16 p70, IAEA, Geneva (1958)

[9] P. Goberis, Modelling of innovative critical reactors with thermalized neutron fluxes in the frame of the Mini-Inca project, rapport de stage (2000), DAPNIA/SPhN, CEA Saclay

[10] http://necs01.dne.bnl.gov/CoN/index.html