Detailed studies of Minor Actinide transmutation-incineration in high-intensity neutron fluxes

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Abstract

The Mini-INCA project is dedicated to the measurement of incineration-transmutation chains and potentials of minor actinides in high-intensity thermal neutron fluxes. In this context, new types of detectors and methods of analysis have been developed. The ^{241}Am and ^{232}Th transmutation-incineration chains have been studied and several capture and fission cross sections measured very precisely, showing some discrepancies with existing data or evaluated data. An impact study was made on different based-like GEN-IV reactors. It underlines the necessity to proceed to precise measurements for a large number of minor-actinides that contribute to these future incineration scenarios.

KEYWORDS: High-intensity thermal neutron fluxes, minor-actinides, incineration-transmutation, sensitivity, GEN-IV incineration scenario

1. Introduction

The feasibility assessment of innovative reactors and fuel cycle systems, proposed within the Generation IV initiative or advanced fuel cycle program (AFC) is still connected with nuclear data and their accuracies. It is most particularly true for systems considered as possible transmuters of transuranic elements or even simply for systems which have to respond to high-level requirements as waste minimization, sustainability, safety and non-proliferation. Most of the nuclear data are available in modern data files, but their accuracies and validation is still a major concern, especially for Minor Actinide (MA).

Indeed, a very recent study was performed about the impact of nuclear data uncertainties on the performance parameters (criticality, reactivity, irradiated fuel isotopic composition, neutron sources …) of future nuclear systems [1]. This report concludes that MA data uncertainties don't play a major role on essential nuclear parameters such as fuel burn-up or criticality. However, when regarding the target precisions, and more particularly the transmutation potentials, the accuracies on MA data should have to be reduced by at least a factor two for Am, Np and Cm isotopes.

This observation has motivated a very ambitious program, namely the Mini-INCA project [2], at the French Atomic Energy Commission (CEA), dedicated to detailed studies of MA

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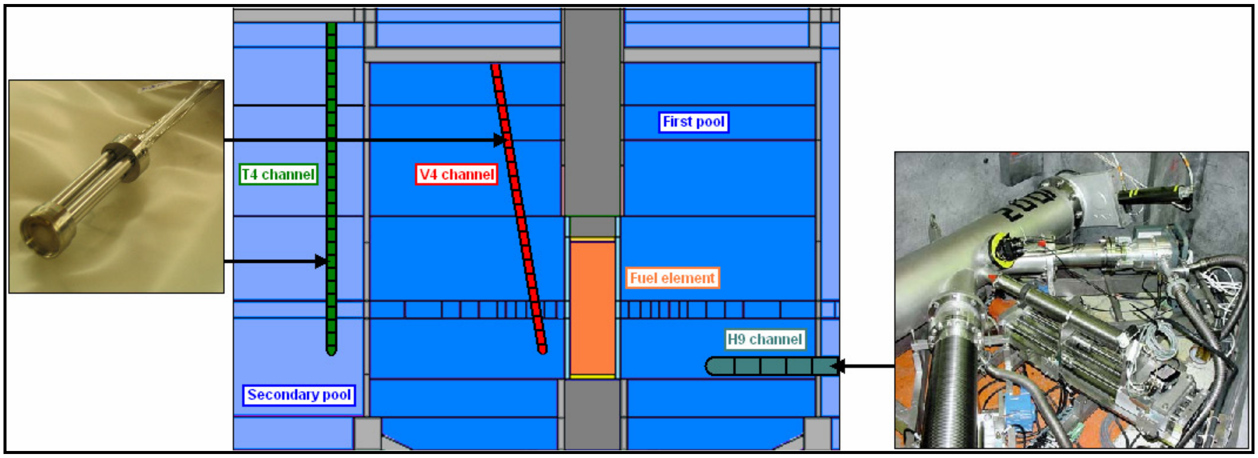
transmutation-incineration chains in high-intensity thermal neutron fluxes. The objectives of this program are to provide precise measurements of the MA nuclear parameters (i.e. capture and fission cross sections, decay half-lives), but also to give integral values such as the incineration potentials.

2. Experimental approach

2.1 Irradiation possibilities

The experimental approach of the Mini-INCA project is mainly based on the use of high neutron fluxes provided by the High Flux Reactor of Laue Langevin Institut (Grenoble-France). It offers the advantages to use µg-mass samples, resulting in negligible local flux perturbation, and to study on-line the formation of short-live isotopes. Three irradiation channels could be used and have been equipped (see). The T4 channel provide pure thermal neutrons with an intensity of the order of 2 10^{13} n/cm^2/s whereas the V4 channel can provide different irradiation positions with different neutron flux energy spectra. In this channel, the thermal neutron proportion (E_n < 1 eV) varies from 100 % to 85 %, and intensities range from 6 10^{13} n.cm^{-2}.s^{-1} to 1.5 10^{15} n.cm^{-2}.s^{-1}. The H9 channel has a neutron flux closed to the one available in an intermediate position of theV4 one.

Figure 1 : The Mini-INCA installation at ILL. The core reactor is shown with the three irradiation channels: the two vertical ones V4 and T4, and the horizontal one H9. Also are shown the alpha and gamma-spectroscopy station on H9 (right photo) and the fission micro-chamber (left photo) that instruments the V4 channel.



In addition to these quasi-thermal neutron fluxes of ILL, the MEGAPIE target at Paul-Scherrer Institute (Villingen, Switzerland) will provide a moderated spallation neutron flux [3], which will permit to extend the MA transmutation-incineration study in epithermal region.

2.2 Experimental set-up

As shown on Figure 1, two complementary experimental set-ups have been developed for the ILL reactor. First, a dedicated spectroscopy bench [4] have been installed and connected to the source exchanger of the Lohengrin mass spectrometer (H9 channel). Equipped with a Si- and

Ge-detector, it allows a quasi-on-line characterization of irradiated samples by α and γ spectroscopy as well as repeated sequences of irradiation and measurement. The Si- and Ge-detector electronic system is able to record in high counting rates (up to 80 kHz for Ge-detector and up to 20 kHz for Si-detector). This α and γ spectroscopy bench allows a fast characterization of irradiated samples which gives a direct access to short-live isotopes.
In complement, specific fission micro-chambers were developed* to sustain high neutron fluxes and high temperature conditions. They are small gap ionization chambers filled with pure Argon gas and there is generally few micrograms of actinide deposited on their anode. Thanks to this thin gap between anode and cathode, the space charge effects due to high fission rates, are considerably reduced. Their functioning is more precisely described in ref [5].
Moreover, for MA transmutation studies, triple-barrel fission chambers were recently developed (see Figure 1). The principle of this new type of detector is based on three electrically independent fission chambers who are sharing the same gas. One of those chambers contains the actinide that will be studied in reference to ^{235}U which is deposited in another chamber. The last chamber does not have any deposit and is dedicated to γ and activation induced background measurements. Those both types of fission chambers allow to measure on-line the fission rate of the actinides as well as their incineration rates.

2.3 Experimental methodology

Since the many years of Mini-INCA projects, some MA transmutation-incineration chains were successfully characterized with the help of high fluxes [2,4,7]. However, these conditions impose some constraints such as the necessity of a strong coupling between the modeling of the experience, high precision nuclear techniques (α-γ spectroscopy, mass spectrometry, fission chambers) and dedicated analysis tools.
The MCNP code was used to modelized the ILL core reactor and to calculate the neutron flux distributions and the corresponding average cross sections [8] for the different irradiation positions. This calculation has also permitted to quantify the impact of resonances on the measured cross sections.
The different experimental techniques listed before give access to different nuclear parameters as it will be illustrated later on the case of ^{241}Am and ^{232}Th. Whereas capture cross sections could be determined by α-γ spectroscopy and/or mass spectrometry, fission cross sections are determined via fission chambers. Moreover, the latter gives an on-line measurement of the evolution of the irradiated actinide and its incineration.
Due to high values of neutron fluxes, the evolution of some MA could become very complex and difficult to interpret. In this context, there was a necessity to use sensitivity tools, in order to understand exactly the dependences of experimental observables (i.e. the current of a fission chamber) with all parameters that can contribute to their creations. That is the reason why we are developping, at the CEA, the ROOT MERCS module. This ROOT shared library is an one-dimension, one-group evolution code, which can compute the absolute and relative sensitivity tables for all nuclear parameters for different types of variables. The sensitivity coefficient $S_{i,j}$ define the impact of the parameter X_j for an experimental observable $Q_i(X_j)$, at a specific point of evolution:

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$$S_{i,j} = \frac{\partial Q_i}{\partial X_j} \cdot \frac{X_j}{Q_i} \tag{1}$$

The observable $Q_i(X_j)$ could be such as the concentration of a specific isotope and could be represented as a function of different parameters 'X_j' such as irradiation or cooling time, neutron flux, but also the cross sections and decay parameters of all isotopes which have contributed to its evolution.
Higher the sensitivity value is, greater is its impact on this variable. So that the best observables to determine a specific parameter X_k are the ones which are defined by a coefficient value $S_{i,k}$ far superior to other parameters:

$$S_{i,k}^{relative} = \frac{S_{i,k}}{\sum_j S_{i,j}} \rightarrow 100\% \tag{2}$$

In this case, the error of the variable Q_i will be dominated by the error of parameter X_k:

$$\Delta Q_i^{\ 2} = \sum_j S_{i,j}^{\ 2} \cdot \Delta X_j^{\ 2} + 2 \cdot \sum_{j \neq i} S_i \cdot S_j \cdot Cov(X_i, X_j) \xrightarrow[S_{i,k} >> S_{i,j}]{} S_{i,k}^{\ 2} \cdot \Delta X_k^{\ 2} \tag{3}$$

That is the reason why, when we are studying complex evolution chains, it is important to optimise the fit for wanted parameters by selecting an optimised set among all variables, with maximum sensitivity coefficients for those parameters. In order to gain a better understanding of such experimental methodology, next section will introduce a previous experiment on ^{241}Am neutronic chain, on which this sensitivity analysis was proceed.

3. Detailed studies of ^{241}Am transmutation-incineration chain

3.1 Mass spectrometry analysis

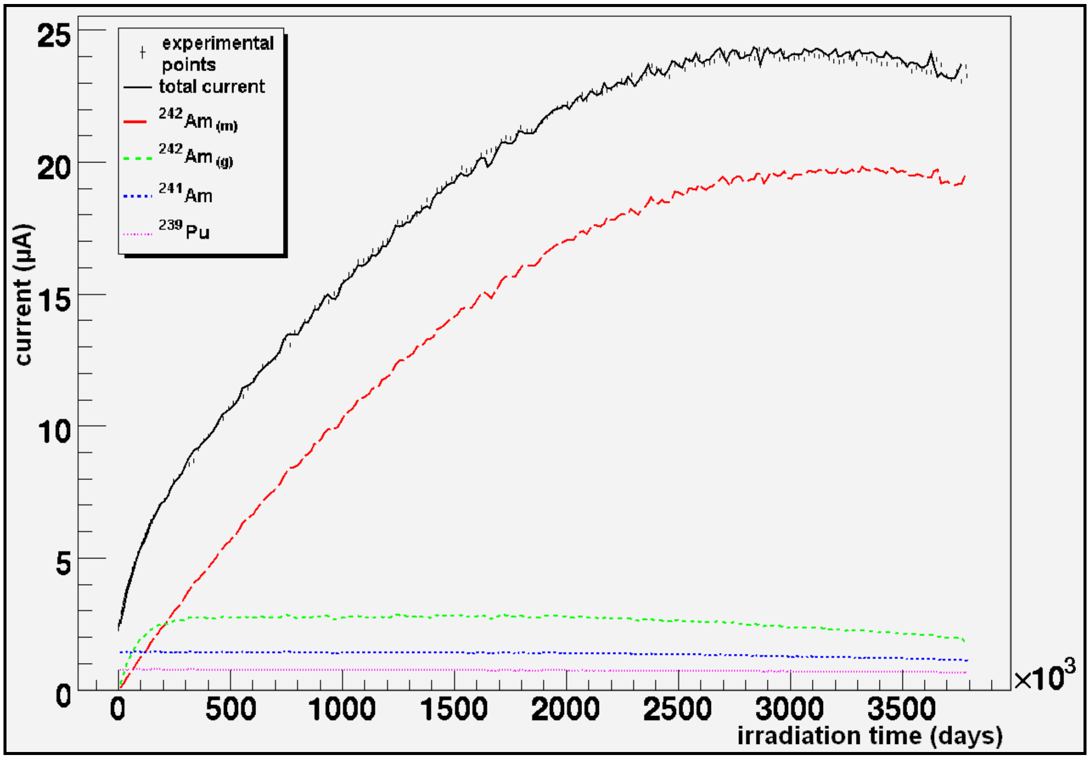
The mass spectrometry analysis was used on ^{241}Am samples irradiated in different neutron fluxes at ILL, to characterise one part of its transmutation chain. The samples were 97% in purity of ^{241}Am, with 2.5% of ^{237}Np and with some plutonium traces [6]. Two sets were irradiated for 11 and 24 days in the T4 channel, whereas four other sets were irradiated in the H9 channel for 1, 5, 11 and 19 days. After a cooling time of about 1 year, samples were analysed by thermal ionization mass spectrometry analysis (TIMS) [6].
The main difficulty of such experimental analysis is to find the best observables set to determine the maximum numbers of nuclear parameters (cross sections, decay parameters, …) with the best precisions. Indeed, in the T4 irradiation, for instance, there are 11 different isotopes, which offers the possibility to create 55 different possible isotopic ratios. However, only 10 can be selected and cross definitions have to be avoided in this final set, eg : [X/Y] = [X/Z]·[Z/Y]. The optimized set was determined by the used of the previously described ROOT MERCS module. By testing all possibilities, we have selected clusters of optimal observables and fitted, on the experimental data, the ROOT MERCS code with the following parameters as free parameters : ^{241}Am, ^{242}Am$_{(m)}$ and ^{242}Cm capture cross sections plus ^{241}Am isomeric branching ratio.

The covariance matrix, taking into account all the correlated errors, was solved numerically by varying the unfitted nuclear parameters in their gaussian error bars. The latter values were extracted from ref [9], when existing, otherwise calculated from CSISRS/EXFOR experimental datas [10] and from NUDAT [11]. The neutron fluxes were measured separately in ref [6] and their errors also included by the same way. The flux value for the H9 irradiation was corrected by 7.2% as compared to the given value in ref [6], in order to take into account the 2% of non-thermal neutrons and the temperature of the moderator (50°C). Due to the fact that T4 samples were irradiated in the same neutron flux spectra (only irradiation times are differents), the corresponding data were fitted simultaneously. The same procedure was done for the H9 samples. The results are presented in Table 1. The obtained values result from the average over T4 and H9 values after corrected from experimental values as explained in ref [8].

3.2 Fission chamber analysis

In order to determine with a better precision the both ^{242}Am fission cross sections, we have irradiated three fission chambers with ^{241}Am-, ^{235}U- and non-deposits, for 44 days at the upper position of the V4 channel ($\Phi \sim 6\ 10^{13}$ n.cm^{-2}.s^{-1}, pure thermal neutrons). The results are shown on Figure 2, together with the preliminary analysis using the ROOT MERCS code (lines). By using as input parameters, the previously measured ^{241}Am capture and branching ratio, and by adjusting the data with the code, we obtain the fission cross sections tabulated in Table 1.

Figure 2 : ^{241}Am fission chamber recorded current as a function of the irradiation time (s). The neutron flux intensity and variations were measured by the ^{235}U chamber. Both of them were compensated by the non-deposit chamber to be corrected from background current. The full line is the result of the adjustment with the MERCS code. Also are indicated the partial contribution of each isotopes.



3.3 Results

As shown in Table 1, there are quite some differences between the results and the common data libraries and for some, greater than the experimental errors.

Table 1 : Comparison between experimental results at 25 meV and nuclear data libraries.

reaction	^{241}Am i.b.r	^{241}Am (n,γ)	^{242}Am$_{(m)}$ (n,γ)	^{242}Cm (n,γ)	^{242}Am$_{(m)}$ (n,f)	^{242}Am$_{(g)}$ (n,f)
This work	0.895030 ± 0.001632	714.179 b ± 23.34 b (preliminary)	1147.49 b ± 114.103 b	20.9680 b ± 1.82 b	6838.71 b ± 341.94 b (preliminary)	2892.69 b ± 173.56 b (preliminary)
ENDF/B-VI.8	0.9000	618.79 b	1344 b	16.87 b	6629 b	2269 b
JENDL-3.3	0.9100	639.50 b	1231 b	15.90 b	6400 b	---
JEFF-3.1	0.9100	650.41 b	1234 b	15.95 b	6416 b	---
Min. deviation	0.56 %	8.93 %	7.29 %	19.53 %	3.07 %	21.55 %
Max. deviation	1.67 %	13.36 %	17.2 %	24.16 %	6.42 %	21.55 %

Except for ^{241}Am capture cross section, these deviations could mainly be explained by the lack of experimental data. Indeed, there are today three measurements for the ^{241}Am isomeric branching ratio [11], only one for the ^{242}Am$_{(m)}$ capture cross section [15] and there are currently no measurement for ^{242}Cm cross section. However, according to ref [9], the ^{241}Am capture cross section is known at almost 10 % (in thermal region) , which is in good agreement with previous table deviations. Our new value for ^{241}Am(n,γ) is compatible with ref [6], whereas the isomeric branching ratio is 2% less than their value. The ^{242}Am$_{(m)}$ fissions cross section is close to data library value, whereas the ^{242}Am$_{(g)}$ show differences of 22%. These new data complete a previous measurement on the ^{243}Am(n,γ)^{244}Am cross section [4] to get a total characterization of the ^{241}Am transmutation chain.

4. Detailed study of the ^{232}Th transmutation chain

The ^{232}Th transmutation chain could be important for the development of inovative fuels. Its chain was studying by mass spectrometry analysis after the irradiation of a pure 100 µg ^{232}Th sample in the V4 channel (at the middle position, $\Phi_{50cm} \sim 8.10^{14}$ n.cm^{-2}.s^{-1}). The analysis was performed with the ROOT MERCS code in the same way that previously describe. Neutron fluency was monitor with ^{59}Co(n,γ)^{60}Co standard reaction irradiated together with the sample and analysed with the γ-spectroscopy chamber. A resume of obtained results is given in Table 2.

Table 2 : Experimental results values and errors at 25 meV and compared with data libraries.

reaction	^{232}Th (n,γ)	^{233}Pa (n,γ)	^{234}U (n,γ)	^{235}U (n,γ)
This work	7.40 b ± 0.21 b	39.41 b ± 0.14 b	98.37 b ± 3.10 b	96.65 b ± 11.07 b
ENDF/B-VI.8	7.40	40.03	99.78	98.69
JENDL-3.3	7.40	41.46	103.05	98.71
JEFF-3.1	7.40	41.60	100.10	98.81
Min. deviation	0.04 %	1.57 %	1.44 %	2.12 %
Max. deviation	0.06 %	5.57 %	4.76 %	2.24 %

5. Impact on MA incineration in some GEN-4-like reactors

5.1 Presentation of simulated scenarios

To understand the relevance of such measurements, we have studied the impact of the uncertainties associated to some MA cross sections on the transmutation of ^{241}Am and ^{237}Np, in two based-like GEN-4 reactors: the European Fast Reactor (EFR), a Gas-Turbine Modular Helium Reactor (GTMHR) and a moderated target in a high-intensity neutron flux (HITF) that could be part of an EFR. These scenarios were chosen in order to be representative of fast, partially moderated and thermal neutron fluxes. The incineration cycle was the same for all scenarios : three irradiations of 1 year each, separated by two cooling times of 1 month to simulate the core-refuelling period. The isotopic evolutions were computed with MERCS module by using neutron fluxes calculated with ECCO/ERANOS for the EFR, and Tripoli for GTMHR [12]. For each scenario, two MA targets : ^{237}Np and ^{241}Am, were simulated. The focus was done on the incineration and transmutation potential and also on the ^{244}Cm isotope formation, which represents the greatest neutron source problematic for the manipulation after irradiation. The results are given in Table 3.

Table 3 : Calculated irradiation-transmutation potentials and ^{244}Cm formation for the three scenarios at the end of 3 years cycle. The errors indicated in brackets are the associated errors due to the uncertainties on capture and fission cross sections of involved MA. The uncertainties were taken from [9], except for the fission of ^{238}Np (10%) and the fission of ^{242gs}Am (40%).

		Incineration	transmutation rate	^{244}Cm formation
EFR	^{241}Am target	6.5 (1.3) %	23 (2.1)%	~ 0 %
	^{237}Np target	6.5 (1)%	22 (2)%	~ 0 %
GTMHR	^{241}Am target	49 (8.7)%	98 (0.1)%	5 (2.6)%
	^{237}Np target	35 (3.8)%	72 (2.2)%	0.026 (0.015)%
HITF	^{241}Am target	82 (6.9)%	96 (1.2)%	3 (2.6)%
	^{237}Np target	80 (2.6)%	~ 100 (0.005)%	0.55 (0.37)%

This table shows the impact of data uncertainties on the incineration and transmutation potential uncertainties is of the order of 10-20% for an EFR whereas it does not exceed 10% for a HITF. But, when considering the ^{244}Cm formation, the impact is much more important for moderated reactors than for rapid one and is of the order of 50%.

5.2 Sensitivity tables

Sensitivity tables have been computed for all scenario cases in order to underlight nuclear parameters which have a great impact on the three defined observables. Only the most unknown contributors to the ^{241}Am and ^{237}Np transmutation chains are listed in 4 & 5. When regarding the incineration potential and ^{244}Cm formation of ^{241}Am target, the most sensitive parameters are the ^{241}Am neutron capture and its isomeric branching ratio (i.b.r.) for all scenarios. On the other hand, when ^{237}Np incineration is concerned, the most important one is its own neutron capture. It has to be stressed than ^{238}Pu appears to be a key isotope for both ^{241}Am and ^{237}Np incineration chains in a GTMHR. In the case of HITF concept the formation of ^{244}Cm is mainly driven by its own neutron capture, whereas for GTMHR, the overall Pu chain and ^{243}Am have significant contributions.

Tables 4 & 5: Sensitivity tables for the most unknown cross sections involved into the ^{241}Am and ^{237}Np transmutation chains.

sensitivity (%/%)		^{241}Am target							
		^{241}Am i.b.r.	^{241}Am $\sigma_{capture}$	^{242}Am$_{(g)}$ $\sigma_{fission}$	^{242}Am$_{(m)}$ $\sigma_{capture}$	^{242}Am$_{(m)}$ $\sigma_{fission}$	^{243}Am $\sigma_{capture}$	^{242}Cm $\sigma_{capture}$	^{244}Cm $\sigma_{capture}$
EFR	incineration	0.55	0.28	0	0	0.19	0	0	0
	transmutation	0	0.88	0	0	0	0	0	0
	^{244}Cm	1.05	0.93	0	0.50	0.07	0.79	0.16	0.03
GT-MHR	incineration	0.81	0.24	0	0.05	0.05	0	0.01	0
	transmutation	0	0.08	0	0	0	0	0	0
	^{244}Cm	1.23	0.28	0	0.34	0.34	0.54	0.04	0.13
HITF	incineration	0.08	0.07	0.03	0	0.01	0	0	0.05
	transmutation	0	0.11	0	0	0	0	0	0
	^{244}Cm	0.52	0.06	0.15	0.05	0.04	0.18	0.01	1.44

sensitivity (%/%)		^{241}Am target		^{237}Np target					
		^{238}Pu $\sigma_{capture}$	^{242}Pu $\sigma_{capture}$	^{237}Np $\sigma_{capture}$	^{238}Np $\sigma_{fission}$	^{238}Pu $\sigma_{capture}$	^{242}Pu $\sigma_{capture}$	^{243}Am $\sigma_{capture}$	^{244}Cm $\sigma_{capture}$
EFR	incineration	0	0	0.18	0	0	0	0	0
	transmutation	0	0	0.89	0	0	0	0	0
	^{244}Cm	0	0.32	0.97	0	0.98	0.97	0.97	0
GT-MHR	incineration	0.31	0	0.65	0.03	0.46	0	0	0
	transmutation	0	0	0.51	0	0	0	0	0
	^{244}Cm	0	0.44	0.89	0	0.73	0.89	0.78	0.06
HITF	incineration	0.03	0.02	0.11	0.09	0.09	0	0	0
	transmutation	0	0	0	0	0	0	0	0
	^{244}Cm	0	0.32	0.40	0.34	0.17	0.39	0.19	1.27

5.3 Discussion

From the previous analysis, it is clear that the parameters, which have a significant sensitivity and/or a great uncertainty, could have big impact on the incineration of MA. For example, ^{241}Am isomeric branching ratio which has a strong sensitivity factor, is only defined by 3 measurements [13], [14], [15], at thermal energy, with a global associated error of 10%. The new experimental value proposed in this work represents an improvement of the error bar (0.19%) by a factor 50 as compared to the last measurement of Shinohara [13]. Moreover in ref [9] the ^{241}Am neutron capture is evaluated with 10% uncertainty whereas the value measured in this work has a precision of 3%. The consequence is a better accuracy on the estimate of the ^{241}Am incineration, with another consequence that is a better optimisation of futur transmuters.

This small sensitivity study places an emphasis on the necessity to improve MA nuclear parameters by making precise measurements as it is done in this work. Even the most measured parameters on this list (i.e. ^{241}Am $\sigma_{capture}$, ^{242}Am$_{(m)}$ $\sigma_{capture}$, ^{238}Pu $\sigma_{capture}$, ^{237}Np $\sigma_{capture}$, ^{242}Pu $\sigma_{capture}$), which have estimated errors [9] ranging between 3% and 20%, could consequently generate errors of several percents. However, the measured data presented in this work are mainly based on thermal neutron fluxes and then in principle have to be used only for moderated reactors. But, for isotopes which are difficult to produce (e.g. ^{242}Am$_{(m)}$) and then to measure in the resonance and fast regions, these data are the only ones able to constraint the evaluations.

6. Conclusion

The use of high-intensity thermal neutron fluxes in the Mini-INCA project has permitted the development of dedicated detectors and analysis tools. In this framework many transmutation chains have been studied such as ^{241}Am, ^{232}Th and ^{237}Np. A detailed analysis has been presented on only two of them, showing the methodology and the precision on the determination of the fission and capture cross sections at the thermal point. Finally, the conclusions of the GEN-IV incineration-transmutation studies have underlined that a better accuracy on some MA nuclear parameters is needed. There is a necessity to proceed to precise measurements in different neutron energy ranges, as it is done within the Mini-INCA project in the thermal region. Future MEGAPIE measurements in PSI will complete those measurements with the use of more epithermal neutron fluxes.

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