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Feasibility study of the AOSTA experimental campaign

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Abstract. The reduction of the nuclear waste is one of the most important nuclear issues. The high radiotoxicity of the spent fuel is due to plutonium and some minor actinides (MAs) such as neptunium, americium and curium, above all. One way to reduce their hazard is to destroy by fission MAs in appropriate nuclear reactors. To allow the MAs destruction an important effort have been done on the nuclear data due to the poor knowledge in this field. In the framework of one of the NEA Expert Group on Integral Experiments for Minor Actinide Management an analysis of the feasibility of MAs irradiation campaign in the TAPIRO fast research reactor is carried out. This paper provides preliminary results obtained by calculations modelling the irradiation, in different TAPIRO irradiation channels, of some CEA samples coming from the French experimental campaign OSMOSE, loaded with different contents of MAs, in order to access, through particular peak spectrometry, to their capture cross section. On the basis of neutron transport calculation results, obtained by both deterministic and Monte Carlo methods, an estimate of the irradiated samples counting levels from the AOSTA (Activation of OSMOSE Samples in TAPIRO) experimental campaign is provided.

1 Introduction

Among the foundations of a sustainable use of nuclear fission energy certainly there are a reliable fuel cycle and a safe manage of radioactive waste. The spent fuel discharged from nuclear power plants constitutes the main contribution to nuclear waste. Most of the hazard from the spent fuel stems from only a few chemical elements such as plutonium and some Minor Actinides (MAs) such as neptunium, americium and curium, plus some long-lived fission products such as iodine and technetium. In this frame, one of the key aspects is the appropriate management of MAs. A technological route to reduce the risks associated with MAs is to transmute them in nuclear systems. However, due to a lack of experimental data, which has meant an absence of precision in MAs nuclear data, it remains difficult to

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establish a detailed design of transmutation systems with reliable accuracy and the capacity to precisely predict the composition of spent fuel. Several NEA and IAEA Working Groups addressed these issues, in particular recommending integral measurements, complementary to parallel efforts for differential measurements, for the following nuclides of MAs from viewpoints of design of transmutation systems and of fuel cycles: ^{237}Np, ^{241}Am, ^{242m}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm and ^{245}Cm. In the frame of the NEA Expert Group on Integral Experiments for Minor Actinide Management [1] a joint collaboration between ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development) and CEA (French Alternative Energies and Atomic Energy Commission) was established with the aim to study the feasibility of a MAs irradiation campaign in the TAPIRO fast neutron source research reactor located at the ENEA Casaccia center near Rome [2]. This paper provides preliminary results obtained by calculations modelling the irradiation, in different TAPIRO irradiation channels, of some CEA samples coming from the French experimental campaign OSMOSE [3] (Oscillation in MINERVE of isotope in 'Eupraxic' Spectra), loaded with different contents of MAs. On the basis of the neutron transport calculations results, obtained by both the deterministic ERANOS [4] and the Monte Carlo Serpent [5] codes, an estimate of the irradiated samples counting levels from the AOSTA (Activation of Osmose Samples in TApiro) experimental campaign, taking into account both geometry and efficiency of the counting system, is provided.

2 The TAPIRO fast neutron source research reactor

TAPIRO (**TA**ratura **PI**la **R**apida a potenza zer**O** - Fast Pile Calibration at 0 Power) is a fast neutrons source research reactor located at C.R ENEA-CASACCIA (Italy). The project, entirely developed by ENEA's staff, is based on the general concept of AFSR (Argonne Fast Source Reactor - Idaho Falls). It was built to support an experimental program on fast reactors and it is in operation since 1971.TAPIRO is currently used, in addition to education and training, for experimental programs in support of different research fields like nuclear data, nuclear fusion, aerospace industry.

It has a maximum power of 5 kW with a neutron flux around $4 \cdot 10^{12}$ n/(cm^2·s) in the centre of the core. The core is cylindrical with a diameter of about 12 cm and a similar height. It is made by metallic uranium (98.5 % uranium and 1.5 % molybdenum) with an enrichment of 93.5 % in ^{235}U. It consists of 2 parts: the upper part is fixed while the lower one is movable. The core is surrounded by a double layer of a copper reflector and by an external borate concrete biological shield. The core is cooled by helium. The reactor is equipped with 2 shim rods, 2 safety rods and a regulating rod. These rods are made of the same material of the reflector, i.e. copper, and the reactor is controlled increasing or reducing the neutron leakage. The system has different experimental channels with various diameters. A horizontal section of the reactor is shown in Fig. 1.

3 Energy averaged capture cross sections evaluation

Neutron transport calculations have been performed by two different methodologies, deterministic by the ERANOS code, and stochastic by the Monte Carlo Serpent code. In Fig. 2 the calculation models are shown.

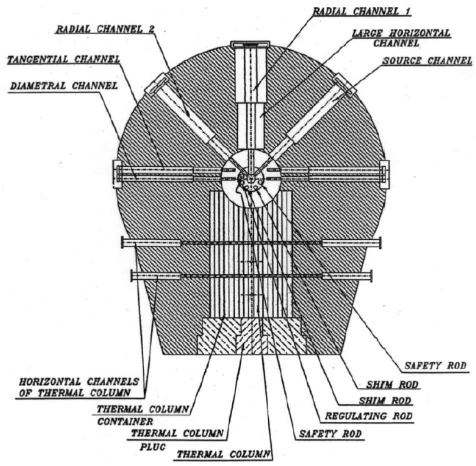
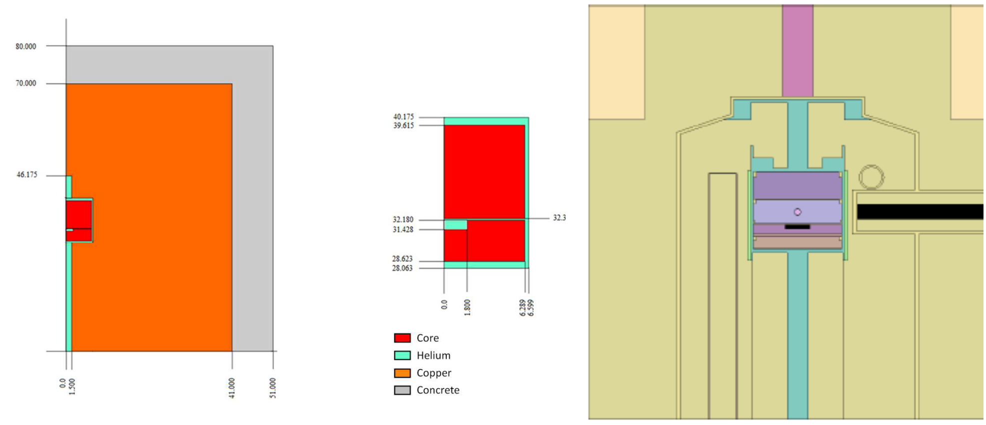


Figure 1. TAPIRO horizontal section.

Neutron fluxes, energy spectra and reaction rates have been evaluated for several minor actinides in correspondence of different radial positions in the diametral experimental channel (cf. Fig. 1).



(a) - RZ ERANOS model (b) - Serpent model

Figure 2. ERANOS and Serpent TAPIRO calculation models.

Subsequently position dependent average microscopic capture cross sections have been calculated to predict the impact on their values of the spectral variations across the system. Average capture microscopic cross sections $\bar{\sigma}_c(\mathbf{r})$ are defined as:

$$\bar{\sigma}_c(\mathbf{r}) = \frac{\int \sigma_c(\mathbf{r},E)\varphi(\mathbf{r},E)dE}{\int \varphi(\mathbf{r},E)dE} \tag{1}$$

with φ neutron flux. As an example, the behavior across the system of the average capture microscopic cross sections for the minor actinide ^{241}Am, calculated with both ERANOS and Serpent codes, is shown in Fig. 3.

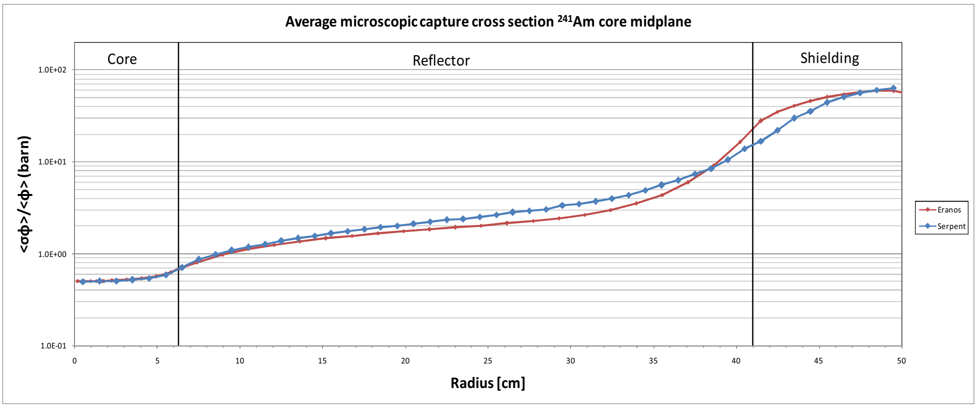


Figure 3. ^{241}Am microscopic capture cross section.

A satisfactory agreement can be observed, also for the other samples, among ERANOS and Serpent codes results.

4 Evaluation of the counting rates after irradiation of the OSMOSE samples

The selected OSMOSE samples contain ^{237}Np, ^{242}Pu, ^{241}Am, ^{243}Am in a double Zircaloy sheath (Fig. 4). The matrix for all the samples is composed by natural uranium.

Internal sheath [mm]	9.56
External sheath [mm]	10.6
Length [mm]	103.5

Figure 4. OSMOSE samples.

It was assumed to irradiate these samples following a weekly scheme characterized by 5 hours of irradiation and 19 hours of cooling repeated for 4 times and then 5 hours of irradiation and 2 hours of cooling. Activity values have been evaluated after these last 2 hours of cooling (Fig. 5). Three sample positions have been considered along the diametral channel: r=12.07 cm (near the core), 24.58 cm (about reflector center) and 45.4 cm (entrance of the thermal column), see Fig. 2 (a). In correspondence of such positions have been calculated by the FISPACT code [6] the activity values, for each OSMOSE sample, associated to the irradiation scheme shown in Fig. 5. The obtained results, by using the ERANOS neutron flux results for this preliminary analysis, are shown in Table 1.

The counting rate [counts/s] for each detector have been evaluated by the relationship:

$$C = A \cdot I_\gamma \cdot \varepsilon$$

with A activity level [Bq], I_γ intensity of the γ or X emission [%], ε geometric efficiency of the detector [%]. The geometric efficiency ε depends by the scintillator type used for detection and by the overall experimental geometrical arrangement. In our case it was considered as scintillator n-type coaxial HPGe detector made of high purity germanium, showing high precision and efficiency for both γ and X rays in the energy range 3 keV÷10

MeV. The geometric efficiency has been evaluated by Monte Carlo MCNP code [7] modeling the arrangement of the counting system (Fig. 6).

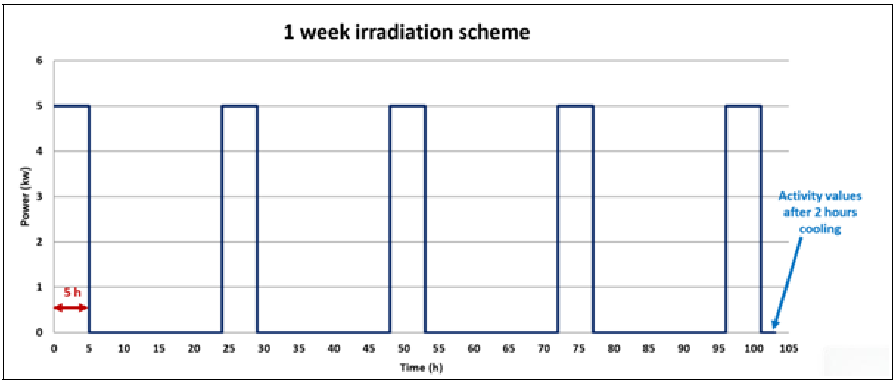


Figure 5. Irradiation scheme.

Table 1. Energy average cross sections and activity levels for each sample at different position in TAPIRO.

	Position	r = 12.07 cm	r = 24 .58 cm	r = 45.5 cm
OSMOSE Samples	ϕ ($n \cdot cm^{-2} \cdot s^{-1}$)	6.94E+11	1.74E+11	8.79E+09
Np237	$\sigma_{c,Np237}$ (barn)	1.04	1.73	19.57
	A (Bq)	2.04E+08	8.49E+07	4.85E+07
Pu242	$\sigma_{c,Pu242}$ (barn)	0.34	0.62	22.48
	A (Bq)	1.15E+08	5.30E+07	9.68E+07
Am241	$\sigma_{c,Am241}$ (barn)	1.32	2.06	32.61
	A (Bq)	1.15E+08	4.50E+07	3.61E+07
Am243	$\sigma_{c,Am243}$ (barn)	1.13	1.83	33.40
	A (Bq)	2.10E+07	8.10E+06	7.14E+06

Table 2 summarizes the results obtained. In particular, in Table 1 are shown, for each sample, the γ or X rays characteristics (energy and intensity), the geometric efficiency and the counting level obtained by the relationship (1). The counting rate levels shown in Tab. 2 seem to predict the feasibility of the AOSTA experimental campaign although, as evident, a more detailed analysis is needed in the next future to confirm these promising preliminary results.

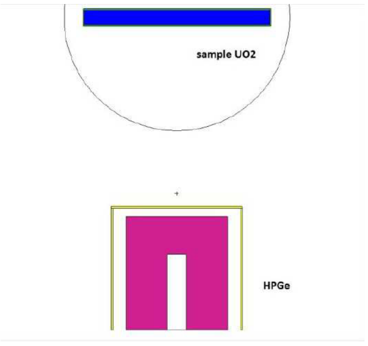


Figure 6. MCNP modeling of the counting system.

Table 2. Counting rates for the selected OSMOSE samples in TAPIRO (ERANOS neutron fluxes).

	Position	r = 12.07 cm	r = 24 .58 cm	r = 45.5 cm
OSMOSE Samples	ϕ (n·cm^{-2}·s^{-1})	6.94E+11	1.74E+11	8.79E+09
Np237	Np238 E γ (keV)	984.45	984.45	984.45
	γ Intensity (%)	25.19	25.19	25.19
	ε Detection (%)	0.186	0.186	0.186
	C (cps)	95487	39779	22738
Pu242	Pu243 E γ (keV)	84	84	84
	γ Intensity (%)	23.10	23.10	23.10
	ε Detection (%)	0.021	0.021	0.021
	C (cps)	5559	2572	4698
Am241	Am 242 E $X_{K\alpha1}$ (keV)	103.374	103.374	103.374
	$X_{K\alpha1}$ Intensity (%)	5.70	5.70	5.70
	ε Detection (%)	0.107	0.107	0.107
	C (cps)	7014	2745	2204
Am243	Am244 E γ (keV)	743.971	743.971	743.971
	γ Intensity (%)	66.00	66.00	66.00
	ε Detection (%)	0.213	0.213	0.213
	C (cps)	29466	11391	10032

Conclusions

In the frame of the NEA Expert Group on Integral Experiments for Minor Actinide Management, a joint collaboration between ENEA and CEA was established with the aim to study the feasibility of a MAs irradiation campaign, named AOSTA (Activation of Osmose Samples in TApiro), in the TAPIRO fast neutron source research reactor located at the ENEA Casaccia center near Rome. Preliminary neutronic analyses of the AOSTA experimental campaign have been performed by both deterministic and stochastic methods, and a good agreement has been observed among the results provided by the two calculation approaches.The counting rate levels obtained by these simulations seem to predict the feasibility of the AOSTA experimental campaign although, as evident, a more detailed analysis is needed in the next future to confirm these promising preliminary results.

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