MA AND LLFP TRANSMUTATION PERFORMANCE ASSESSMENT IN THE MYRRHA SMALL-SCALE ADS

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Abstract

A typical fast sub-critical core configuration dedicated to MA and LLFP transmutation studies has been defined containing loaded with 605.5 kg (heavy metal) of highly Pu-enriched MOX (U-Pu) fuel, 18.089 kg of MA and 24.70 kg of ^{99}Tc. The core was shown to achieve a primary source neutron multiplication factor, k_s, of 0.962 (k_{eff} = 0.956) yielding, for a 5 mA proton beam, a thermal power of 53.7 MW. The performances of that core has been assessed as to it potential to transmute the loaded MA and LFFFP considering a one year operation cycle consisting of 3 times 90 effective full power days of irradiation with 30 days shut-down in between

Introduction

In the framework of the long-term management of radioactive waste, partitioning and transmutation (P&T), is the only technology which is capable of accelerating the natural decay sequence and hence of reducing the radiotoxic inventory of some actinides and some fission products for which the natural decay reactions take hundred thousands of years to reach the initial uranium ore toxicity level. Among different nuclear reactors, Accelerator Driven Systems (ADS) are considered to have a very good potential for safely and efficiently burn MAs. They can indeed pave the way for a more environmentally safe and acceptable nuclear energy production through the transmutation of nuclear waste.

Fundamental and applied R&D are crucial in the development of ADS and demand the availability of appropriate experimental facilities. To address this need, the Belgian Nuclear Research Centre (SCK•CEN) is designing since 1998 a multipurpose ADS called MYRRHA. MYRRHA aims to serve as a basis for the European experimental ADS and to provide neutrons and protons for various R&D applications. It consists of a proton accelerator delivering a 350 MeV*5 mA proton beam to a liquid Pb-Bi spallation target that in turn couples to a Pb-Bi cooled sub-critical fast core [1].

In 2001, different European research centres, universities and nuclear industry companies decided to streamline and concentrate their efforts on ADS in order to be able to propose a common European experimental accelerator driven system. This lead to the PDS-XADS (Preliminary Design Studies of an eXperimental ADS) project performed within FP5 supported by the European Commission (2002-2004), with the objective to evaluate different candidates previously developed in Europe in order to select the most appropriate concept. MYRRHA was one of the three candidates and has the smallest scale in terms of total power (50 MW) [2, 3, and 4].

In 2004, preparing for FP6, all European design and R&D efforts in ADS are grouped within the Integrated Project EUROTRANS of which the final goal is to design a European Transmutation Demonstrator (ETD). Within this frame, SCK•CEN is willing to open the characteristics of MYRRHA to be updated and brought to finalization for better answering the objectives of the ETD/XT-ADS project (eXperimental demonstration of the technological feasibility of Transmutation in an ADS).

This paper reports the transmutation potential of a typical sub-critical core configuration of MYRRHA loaded with six U-free MA fuel assemblies. The performances as to the LLFP incineration in the resonance spectrum range are assessed considering six metallic ^{99}Tc pin bundles loaded in the peripheral channels of the fast core.

Typical sub-critical core configuration

Various core loading pattern have been investigated as candidate for the configuration of the MYRRHA core dedicated to MA transmutation studies. The major requirements for the BOL core consisted in achieving a power of 50 MWth while keeping the k_{eff} -value close to 0.95.

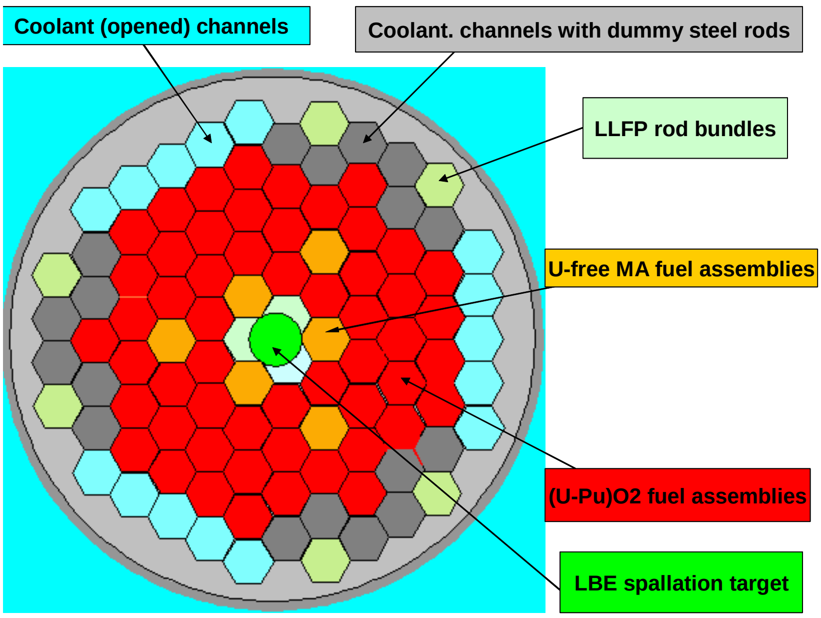
One typical sub-critical core configuration assessed hereafter is displayed in Figure1. This core configuration consists of a lattice of hexagonal channels loaded with 53 fuel assemblies housing 30wt% Pu/(Pu+U) MOX fuel pins arranged in a triangular pitch of 8.55 cm and having an active length of 60 cm. The (U-Pu)O_2 fuel pellets have a density of 10.55 g/cm^3 (95%TD) and each assembly contains 91 fuel pins yielding 605.5 kg of heavy metal as a total load. The initial isotopic vectors of the MOX and of the MA pellets are gathered in Table 1 [4].

The minor actinide load consists of six assemblies each one housing a lattice of 91 pins consisting of $(Pu_{0.4}\text{-}Am_{0.5}\text{-}Cm_{0.1})O_{2-x}$ fuel within a MgO inert matrix. Besides the active length (40 cm), the dimensions of the MA assemblies are the same as those of the MOX fuel ones. The density of the MA pellet is 6.077 g/cm^3 resulting in a total amount of 18.089 kg of MA, half of which (3 assemblies) being loaded in channels "A" lying closest to the spallation target and the other 3 assemblies in channels "D" (see Figure1).

Table 1. **Composition of the fuel and MA pellets**

Vector	wt% fraction
MOX fuel pellet	
U/Pu/O	61.77/26.47/1176
U-234/U-235/-U-236/U-238	0.003/0.404/0.10/99.583
Pu-238/Pu-239/Pu-240/Pu-241/Pu-242	1.27/61.88/23.50/8.95/4.40
MA pellet	
Pu/Am/Cm/Mg/O	23.25/30.32/6.06/19.18/20.19
Pu-238/Pu-239/Pu-240/Pu-241/Pu-242	5.06/37.91/30.31/13.21/13.51
Am-241/Am-243	66.67/33.33
Cm-244/Cm-245	90/10

Figure 1. **Typical core configuration at BOL**



The ^{99}Tc sample is assumed to be a simple metallic rod covered by steel clad. Each assembly houses 37 LLFP rods surrounded by two rings of steel dummy rods. Adopting a density of 11.5 g/cm^3, this results in a total amount of 24.70 kg loaded in the fast core peripheral channels. To soften the neutron spectrum thereby enhancing resonance captures in ^{99}Tc, assembly boxes containing a lattice of steel dummy rods are loaded in transition zones between the fuel assemblies and the LLFP ones.

Computational tools and method

The prompt radiation transport calculations were performed with the MCNPX Monte Carlo multi-particle transport code [5] running in a parallel computer environment with MPI-multiprocessing. Core calculations were carried out in keff-mode in order to set-up the sub-critical core configuration and in source-mode in order to derive the neutronics characteristics of the latter.

The MCNPX 2.5.e version has been adopted, taking advantage of many new capabilities and enhancements offered by this code version. The most relevant is perhaps the so called "*mix-and-match*" capability which enables mixing nuclear data tables and physics models for different nuclides and using the data tables to their upper energy with physics models above the upper data table energy, even when the upper energy boundary of data tables for different nuclides differs.

Thanks to the mix-and-match capability, we have been able to use pointwise continuous-energy cross section data from any available MCNP low-energy libraries over their stated ranges (0 -20 MeV) along with the LA150N neutron library in tabular range (0 - 150 MeV) for 42 isotopes including lead, bismuth and most of steel components (Cr, Fe, Ni,).

As to nuclear data for the fuel components, we had to choose mainly between ENDF/B6.8 and JEF2.2 for which cross-sections at various temperatures were available. In our preliminary calculations we have chosen cross-sections from the ENDF/B6.8 in order to remain consistent with the LA150N library since the latter is typically extends ENDF/B6 data from 20MeV to 150 MeV. However sensitivity studies as to the nuclear data for MOX fuel had shown that ENDF/B6 yields significantly higher values for keff and ks compared to JEF2.2. Moreover, from the FP5-MUSE benchmark we observed that JEF database yields value closer to the experimental ones. Therefore cross-sections from JEF2.2 were used for the fuel (1500°K assumed for all fuel assemblies) and all other material not available in the LA150N library.

For protons transport, the LA150H library was used in tabular range (1.0 to 150.0 MeV) for 41 available isotopes. Beyond this range and for non available isotopes, physics models were used. Pions, muons, and kaons are treated only by physics models whereas photons from 1 keV - 100 MeV were treated using the standard MCNP libraries.

The transmutation calculations have been performed using the ALEPH code [6], a MC burn-up code currently under development at SCK•CEN. It is designed to use any version of MCNP or MCNPX (we currently use MCNPX 2.5.e), NJOY 99.90, ORIGEN 2.2 and JEF 2.2 data files (in their original ENDF-6 format) to perform burn up calculations. For every burn up step, MCNPX is used to calculate a spectrum in the cells containing material to be burned. These spectra are then used to calculate burn-up-dependent one-group cross section libraries for ORIGEN along with the right power and/or flux history required for depletion calculation. The microscopic point-wise continuous cross sections used by ALEPH to calculate the one-group cross sections are generated by NJOY. The same nuclear data are used in MCNPX to perform the required core calculations. After the depletion calculation, the material compositions of the burned cells are updated including the most relevant activation and fission products that accounts for a user-fixed fractional level of the total absorption in a cell (99% in the present case). For the transmutation calculations described in this paper, we have calculated 3 cycles of 90 effective full power days (EFPDs) of irradiation (subdivided into 3 burn-up steps of 30 days) followed by a 30 days shut-down period. As a major simplification, only cells containing the MA and LLFP were burned, leaving the fuel assemblies in fresh state throughout the assumed operational period.

MYRRHA ADS geometrical model

The modeling capability of the MCNPX code package enables a rather complete and detailed model of MYRRHA XADS. All relevant features of the fast core components have been taken into account. The geometry modeling of the fast core was carried out in great detail, the various material zones (fuel, gap and cladding) of each fuel and sample pin and those of the fuel assembly being explicitly represented. The upper and the lower structures of the assemblies, viz. the grid, inlet and outlet tube, were modeled in a realistic way with only minor geometrical distortions with respect to the actual design. On Figures 2 through 4, the close views of the MYRRHA core as modelled reveals details of fuel pin, MA pins, steel dummy rods and LLFP sample pins assemblies.

Figure 2. **Close-view of fast core model features**

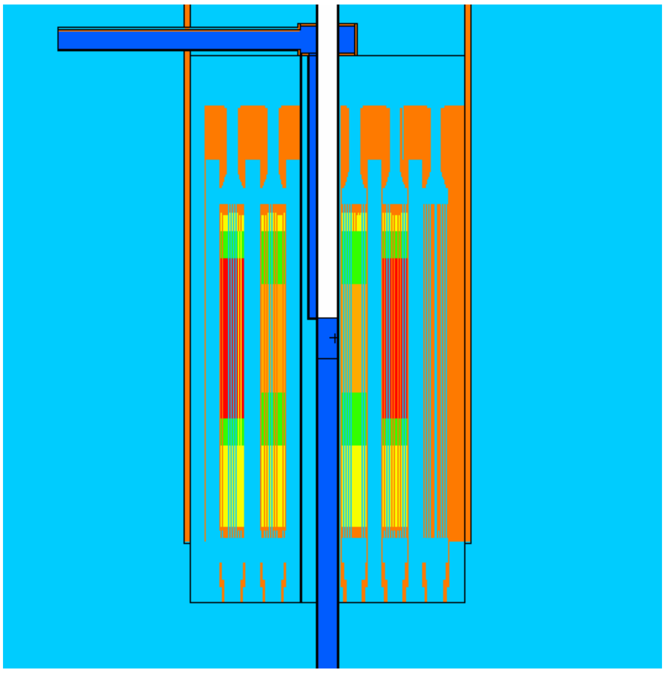


Figure 3. **Radial cut-view of fast core model (mid-plane)**

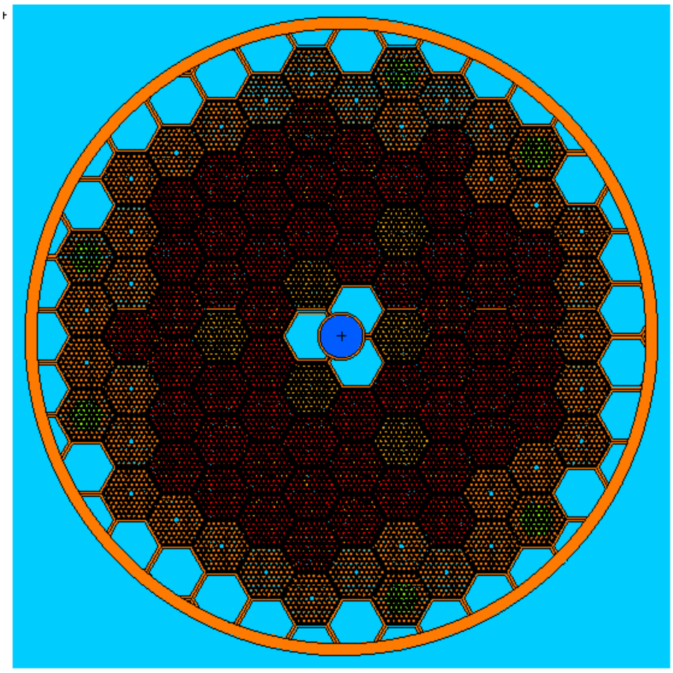
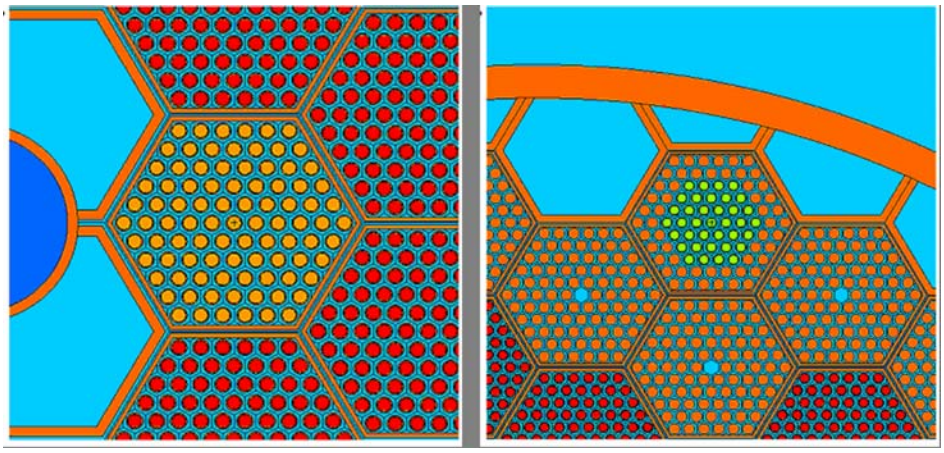


Figure 4. **Close-view of fuel, MA, steel dummy and LLFP pin lattices**



Sub-critical core static characteristics and performances

The MCNPX source multiplication calculation with 160000 protons histories tracked has yielded a net neutron production of 5.94 neutrons per incident proton (n/p). Normalized to a 5 mA-proton beam intensity, this results in a volume-and-energy-integrated primary spallation (non-fission) source intensity of 1.87 10^{17} n/s.

The k_{eff}-multiplication factor value has been calculated by running the MCNPX calculation in the KCODE-mode. For the adopted configuration the value obtained is $k_{eff} = 0.95625 \pm 0.00014$. The k_s-value was obtained by analyzing the neutron balance in the MCNPX code output of a sub-critical fixed source problem run in (nps-mode). We have derived a k_s-value of 0.96215 ± 0.00034 using the well known formula which gives the number of fission in ADS, N_0 being the number of *primary* source neutrons:

$$N_{fiss} = \frac{N_0}{\nu} \frac{k_s}{1 - k_s}$$

The thermal power due to fission was calculated using a fission-rate-weighted average value of 210 MeV per fission taking into accounts energy release by delayed processes mainly the delayed gammas and the fission product decay. The total thermal power amounts to 53.68 MW. Beside this, one has to consider the 1.43 MW deposited inside the spallation target by the 5 mA proton beam.

Over the one-year operational cycle, the average total fluxes are $3.17\ 10^{15}$ n/cm^{2}s, $2.73\ 10^{15}$ n/cm^{2}s and $1.08\ 10^{15}$ n/cm^{2}s, respectively in MA assemblies A, MA assemblies D and LLFP assemblies.

The average neutron spectra within the MOX fuel pellet and within the MA pellet are shown in Figure 5. The four first spectra overlap quite well in the energy rangy 10^{-4} MeV - 10 MeV and are typical of ADS fast core. In Figure 6, the spectra computed inside the ^{99}Tc rod is plotted along with the pointwise microscopic ^{99}Tc (n,γ)-cross-section. One observes the flux dips matching with the resonance peaks as a result of the resonance self–shielding. Moreover there are more neutrons in epithermal energy range compared to the case of fuel channel, indicating the actual potential of such a core configuration to incinerate LLFPs.

Figure 5. **Typical neutron spectra inside MYRRHA core**

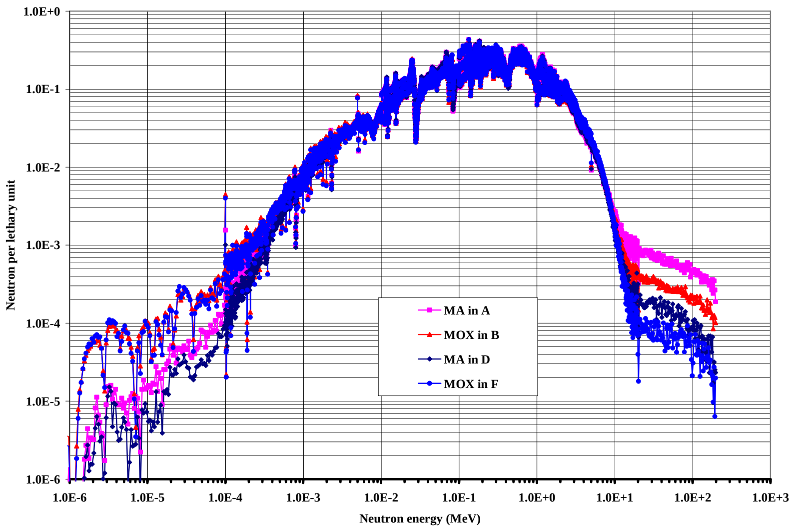
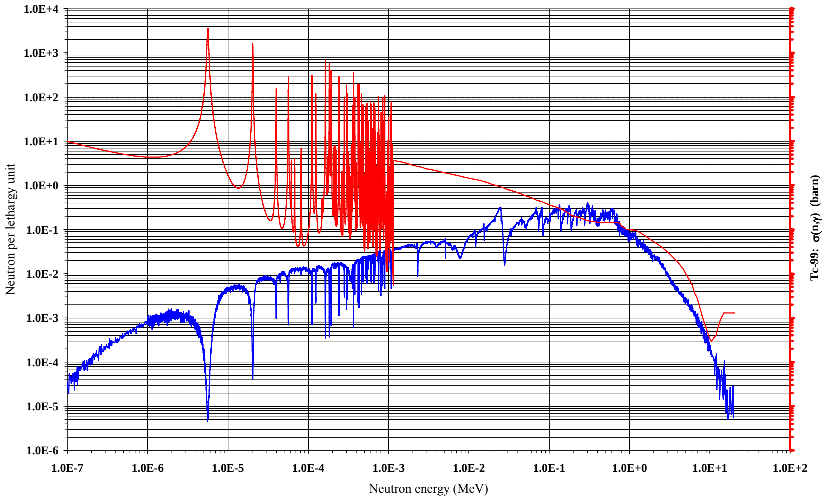


Figure 6. **Average neutron spectrum within ^{99}Tc rod Vs σ(n,γ) of ^{99}Tc**



MA Transmutation performances

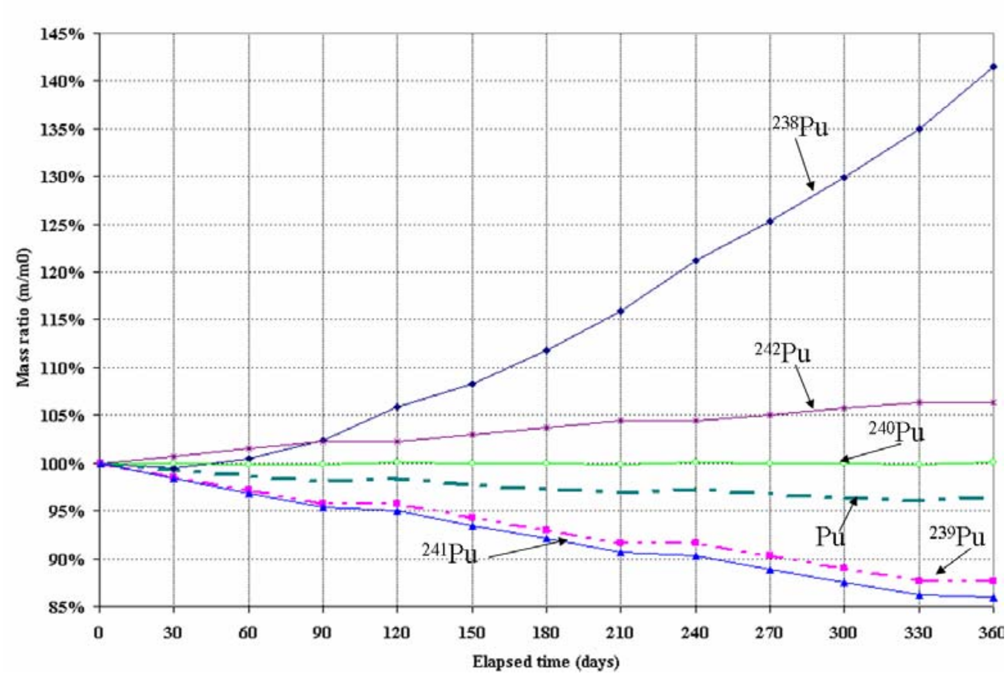
In Table 2, the net mass variation is given for the various components of the MA sample pellet. The incinerated amount of MA is 876 g over the initial load of 18.089 kg. There is a net removal of Pu and Am and a net build-up of Cm.

Figures 7 through 9 display the time-evolution of mass ratios for the three MA components pin over the considered one year operation cycle, viz. 3 times 90 EFPDs irradiation period followed by 30 days of shut-down. One observes a rather low decrease of only 4% in the Pu element mass. The mass decrease occurs for fissile isotopes (-12% for ^{239}Pu and 14% for ^{241}Pu but one observes a build-up of fertile isotopes, mainly the ^{238}Pu, produced during the irradiation by the (n, 2n) reaction in ^{239}Pu and, during the shut-down, the through the α-decay of ^{242}Cm, the latter resulting from neutron capture on ^{241}Am during the irradiation.

Table 2. **Masse balance for the transmutation of a (Am, Pu, Cm) MA sample**

MA components	component amount (g) BOC		Δmass = mass(EOC)-mass(BOL)		
	MA in A	MA in D	MA in A	MA in D	sum MA
^{238}Pu	183.05	183.05	73.68	64.04	137.72
^{239}Pu	1371.43	1371.43	-195.87	-168.75	-364.62
^{240}Pu	1096.49	1096.49	-6.91	-1.65	-8.56
^{241}Pu	477.88	477.88	-72.56	-65.80	-138.36
^{242}Pu	488.74	488.74	35.46	31.09	66.55
Pu	**3617.60**	**3617.60**	**-166.21**	**-141.06**	**-307.27**
^{241}Am	3015.55	3015.55	-392.50	-335.65	-728.15
^{242m}Am			60.13	52.77	112.90
^{243}Am	1509.40	1509.40	-159.44	-136.91	-296.35
Am	**4524.95**	**4524.95**	**-491.82**	**-419.78**	**-911.60**
^{242}Cm			116.49	101.16	217.65
^{243}Cm			2.01	1.47	3.48
^{244}Cm	813.62	813.62	57.85	46.52	104.36
^{245}Cm	90.40	90.40	9.21	7.85	17.06
^{246}Cm			1.84	1.55	3.38
Cm	**904.03**	**904.03**	**185.56**	**156.99**	**342.55**
	1808.05				**-876.32**

Figure 7. **Time-evolution of Pu's mass ratios (m/m_0)**



$$^{241}Am + n \rightarrow {}^{242}Am(90\%) \xrightarrow{\beta^-(16\ heures)} {}^{242}Cm \xrightarrow{\alpha(162\ jours)} {}^{238}Pu$$

$$\searrow {}^{242m}Am(10\%) \overset{\beta^-(141\ ans)}{\nearrow}$$

The net mass decrease of the Am element is about 11%. As to the Cm element, Figure 9 shows a steady increase for the isotopes present at start-up, namely ^{244}Cm and 2^{45}Cm.

$$^{243}Am + n \rightarrow {}^{244}Am \xrightarrow{\beta^-(10.1\ heures)} {}^{244}Cm \xrightarrow{\alpha(18.1\ ans)} {}^{240}Pu$$

One also notes the build-up of ^{242}Cm produced by the decay ^{241}Am. The net increase of the Curium is about 15%. One may conclude that the transmutation of curium is not achievable as far as Am is present in the MA sample.

Concerning the ^{99}Tc, the mass incinerated is 431.1 grams that is 1.75% of the initial load. A better design of the ^{99}Tc rod and by use of better material than steel rod to soften the spectrum would likely improve that performance.

Figure 8. **Time-evolution of Am's mass ratios (m/m_0)**

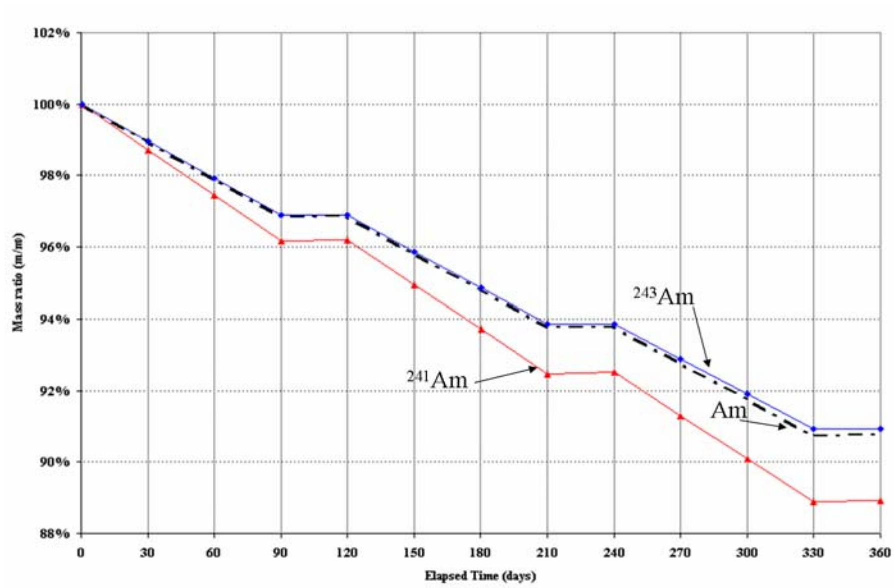
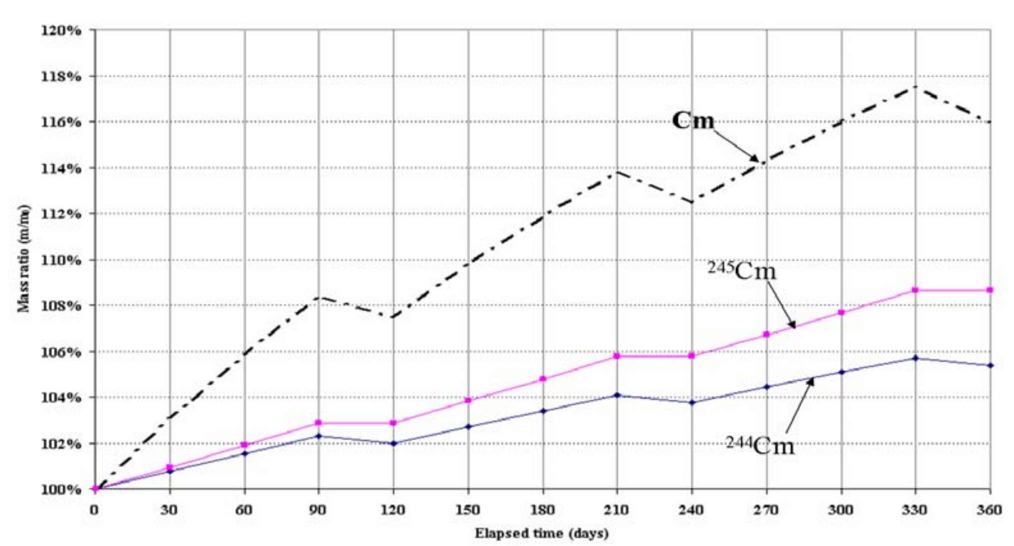


Figure 9. **Time-evolution of Cm's mass ratios (m/m$_0$)**



Conclusions

A typical fast sub-critical core configuration dedicated to MA and LLFP transmutation studies has been defined containing loaded with 605.5 kg (heavy metal) of highly Pu-enriched MOX (U-Pu) fuel, 18.089 kg of MA and 24.70 kg of ^{99}Tc. The core was shown to achieve a primary source neutron multiplication factor, k_s, of 0.962 (k_{eff} = 0.956) yielding, for a 5 mA proton beam, a thermal power of 53.7 MW.

Moreover a more recent MCNPX code version offering many new capabilities and enhancements beyond former versions has been used along with more appropriate nuclear data libraries in order to assess the capability of the defined core to transmute a realistic load of MA and LLFP.

The net MA destruction amounts close to 900 grams over an initial amount of about 18.1 kg. The net mass decrease is observed for the Pu and Am components due to removal of their fissile isotopes whereas a mass increase is observed for all Cm isotopes.

It is shown that the peripheral channels of the fast core may be used to incinerate LLFP. The achieved performance may be improved by using appropriate moderating material to soften the neutron spectrum.

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