PERFORMANCE ON ACTINIDE TRANSMUTATION OF LEAD-THORIUM BASED ADS

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Abstract

The FACET group at CIEMAT is studying the properties and potentialities of several lead-cooled ADS designs for actinide and fission product transmutation. The main characteristics of these systems are the use of lead as primary coolant and moderator and fuels made by transuranics inside a thorium oxide matrix.

The aim of the study is to analyse the effect of some operation parameters (fuel transuranics composition, thermal power and transuranics recycling scheme) in the ADS performance, mainly in the achieved transmutation rates and in the accelerator requirements.

The model selected enhances the energy production by the ^{233}U breeding from thorium seed. This breeding can maintain the neutron multiplication during long burnups, improving the transmutation capacity. The fuel inventory isotopic evolution during burnup will be presented illustrating the general capabilities of this strategic option for transuranics transmutation.

Introduction

One of the major problems of nuclear power production is the undesired long-lived radioactive waste that comes from the spent fuel used for electricity production. Currently many countries plan to deal with these products storing them into geological repositories. However, in the recent years an increasing attention is paid to the transmutation option as a complementary activity to geological disposal in a near future [1]. Among the foreseen advantages of transmutation are a reduction in volume of the high level waste and a reduction in the long-term radiotoxicity inventory, with an impact in reducing the final costs and potential risks of the geological repository.

The transmutation of radioactive waste could be applied to two main groups of nuclides, each group with its own elimination methods and different impact in the waste management strategy:

- The transuranics (TRUs) mainly neptunium, plutonium, americium and curium isotopes coming from the LWR's discharge. They are responsible of the long-term radiotoxicity and their elimination should be done by fission since every neutron capture just increases the mass number of the considered actinide. Any optimised TRUs transmutation strategy has to reach the highest fission to capture ratios, something not easy to achieve in the case of the fissile under threshold TRUs, and take into account the energy production during transmutation process.
- The fission products (FP), some of them with a very large radiotoxical potential in the short or even in the long term. Their elimination means to transform them by neutron capture and subsequent radioactive decay into stable isotopes, something that has some extra energy cost.

The impact of the transmutation in the waste management policy of each country will depend on the size of the local nuclear power industry and the characteristics of the current and foreseen nuclear fuel cycle, among others. An optimal use of nuclear waste transmutation is unavoidable connected to the development of partitioning methods due to the necessity to separate waste streams: TRUs and FP from the spent fuel uranium matrix. Currently, nuclear fuel reprocessing is available only in a few countries in the world. In addition, some countries assume that plutonium is another nuclear fuel instead of as waste, and burn it with the MOX fuel technologies. To them, the TRUs elimination means neptunium, americium and curium elimination in specially design systems, with different properties compared to the TRU-with plutonium charged ADS.

Due to the diverse points of view about transmutation, there is a wide scope of R&D strategies that has produced a large amount of proposed policies and systems for nuclear waste transmutation that could be available in the near future. However, in the last decade the Accelerator Driven Systems (ADS) appear among the most promising transmutation systems. Basically the common components of every ADS are:

- A subcritical core ($k<1$), loaded with the unwanted TRUs and (maybe) FP to eliminate.
- A proton accelerator, producing a beam of some mA of intensity and an energy of a few hundreds of MeV. This beam produces the spallation neutron source that the subcritical core need in order to maintain its operation regime.

The subcritical core configuration of the ADS would allow burning up some atypical actinide mixtures for long irradiation periods; with this kind of systems it is not necessary to reach some reactivity excess in order to start the operation, because the external source maintain the steady state.

The Energy Amplifier Model of Lead-thorium Based ADS

The ADS model considered in the present study is the so-called Energy Amplifier (EA), a concept developed at CERN by Prof. C. Rubbia's research group [2].

Basically an EA is an ADS cooled by molten lead, material that also works as spallation target and neutron diffusing medium. Because of the special neutronics properties of lead, the neutron regime is fast (an energy spectrum centred on a few hundreds of keV). The fast EA design is, in principle, very adequate for elimination of those TRUs non-fissile under thermal spectra (see discussion in refs. [2]). The EA concept is very flexible permitting:

- A neutron multiplication constant (k_{eff}) that could vary between 0.93 and 0.98. The chose of k_{eff} value has an important impact on accelerator requirements.
- Both metal and oxide fuel has been proposed, depending the final selection on the power density limits and the working temperatures. Typically in the EA core, the TRUs charge to transmute is mixed into a thorium matrix. The presence of thorium allows the system to maintain the neutron multiplication: the fissile TRUs burning can be compensated by the ^{232}Th to ^{233}U breeding, followed by ^{233}U fission.
- A wide range of operating conditions, with different thermal power (a few MW up to 1 500 MW) or (equivalently) proton beam conditions (300 MeV to a few GeV of energy), depending on the model considered. The operating conditions will play an important role in the transmutation efficiency.
- Options under study have been presented by Rubbia's team [2] for FP incineration, distributing them in the lead diffusive media outside the EA core.

The simulations performed in this study have been focused primarily in the effects of the initial TRUs inventory composition, the thermal power and the proton beam energy on the performance of the system. The table 1 summarised the main parameters of the models under study. Two thermal energy outputs have been considered: 200 MW, which corresponds to a large-size demo EA facility, and a medium-size energy production unit of 800 MW. For simulation purposes, it has been supposed that the increase in thermal power is gained not directly by beam intensity increase but by proton kinetic energy growth, therefore going from 200 MW_{th} to 800 MW_{th} would imply a proton energy rise from 380 MeV to 1 GeV.

The study on the nuclear fuel composition effect on the EA performance has been done for three selected TRU mixtures. These mixtures have been calculated with ORIGEN2.1 [3] (table 2) modelling a typical PWR discharge. The differences are due to the cooling down time, considered as the delay time between the PWR discharge and the EA load (10, 25 and 40 years). The hypothesis is that the PWR discharge reprocessing produces a transuranics stream without any specific element separation inside it. This means that neptunium, plutonium, americium and curium are extracted in almost the same relative percentage they have in the spend fuel.

Basically, increasing the cooling down time produces an increase in the ^{241}Am concentration by ^{241}Pu decay. This conversion will have an important effect in neutronic properties evolution of the EA because while the ^{241}Pu is a fissile actinide, the ^{241}Am is fertile (by neutron capture it produces some amount of high fissile ^{242*}Am). There is also a slight decrease of the ^{238}Pu concentration while other plutonium isotopes (^{239}Pu, ^{240}Pu and ^{242}Pu) remain near to constant. Among the curium isotopes another rapid decayed isotope is the ^{244}Cm.

Table 1 **EA Models simulated**

Number	1	2	3	4
Thermal output (MW)	200	800	800	800
Proton energy (MeV)	380	1000	1000	1000
Fuel composition		(TRUs+Th)O_2		
TRU mixture cooling down time (years)	40	40	25	10
Fuel oxide mass (kg)		~10 ton		
TRUs/Th	~0.32	~0.32	~0.35	~0.38
(Th+TRUs)/Pb mass fraction (core)	~0.20	~0.20	~0.20	~0.20
Cladding material		HT9 steel		

All the simulated EA models share the geometry characteristics (table 3). The geometry models used during simulation are:

- A complete detailed description of the EA core, where each single fuel pin of every fuel bundles, including its own cladding and the bundle wall, can be distinguish.
- A homogenous model, where the internal structure of a fuel bundle is homogenised to a mixture of materials, preserving the total mass and every isotope mass fraction during the conversion (figure 1).

The differences in the Monte-Carlo estimators when using heterogeneous or homogeneous descriptions of the EA has been discussed elsewhere [4].

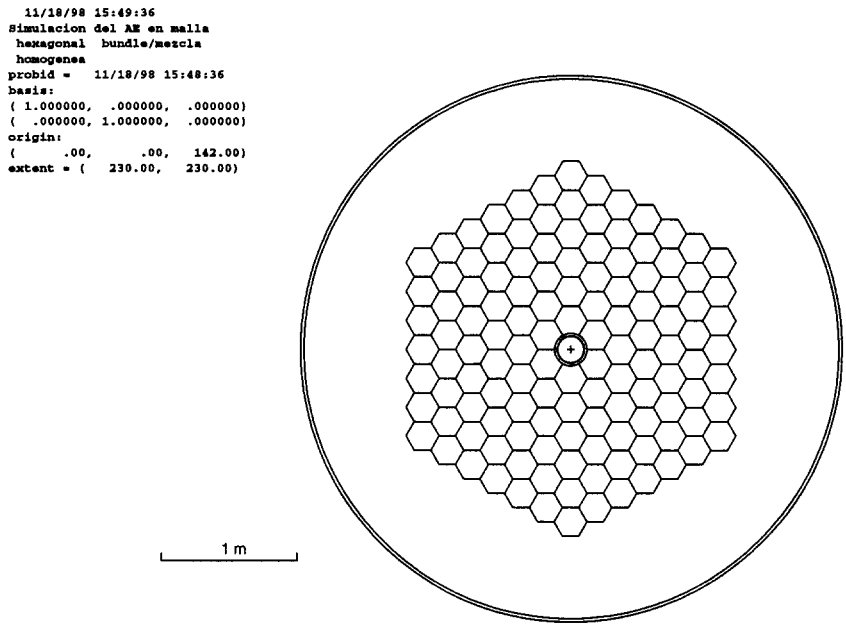
Table 2 **TRU mixtures mass fractions considered in the simulation**

Cooling down time	**10 years**	**25 years**	**40 years**
^{237}Np	5.31E-02	5.51E-02	5.78E-02
^{239}Np	0.00E+00	0.00E+00	0.00E+00
^{236}Pu	1.35E-08	3.57E-10	0.00E+00
^{238}Pu	1.71E-02	1.52E-02	1.35E-02
^{239}Pu	5.15E-01	5.16E-01	5.17E-01
^{240}Pu	2.13E-01	2.14E-01	2.15E-01
^{241}Pu	8.39E-02	4.08E-02	1.99E-02
^{242}Pu	4.96E-02	4.98E-02	4.99E-02
^{241}Am	5.65E-02	9.80E-02	1.17E-01
^{242*}Am	9.77E-05	9.10E-05	8.56E-05
^{243}Am	9.82E-03	9.83E-03	9.84E-03
^{242}Cm	2.55E-07	2.37E-07	2.07E-07
^{243}Cm	3.83E-05	2.68E-05	1.84E-05
^{244}Cm	2.04E-03	1.15E-03	6.52E-04
^{245}Cm	1.22E-04	1.22E-04	1.22E-04
^{246}Cm	1.38E-05	1.38E-05	1.38E-05
^{247}Cm	1.37E-07	1.37E-07	1.38E-07

Table 3 **Common geometry EA parameters**

EA core	
Configuration	Hexagonal
Number of fuel bundles	120
Fuel bundles	
Flat to flat	210.96 mm
Number of pin per bundle	169
Pitch between pins	15.8 mm
Total length	150 cm
Fuel pins external diameter	8.2 mm
Proton beam, spallation target	
Beam pipe material	HT9
Beam window material	W
Thickness	3 mm
External diameter	20 cm
Vessel	
Thickness	2.5 cm
Material	HT9
Lead column height (with regard to the core center)	6 m

Figure 1 **Homogenous model of the EA core fuel bundles array**



Simulation Procedure

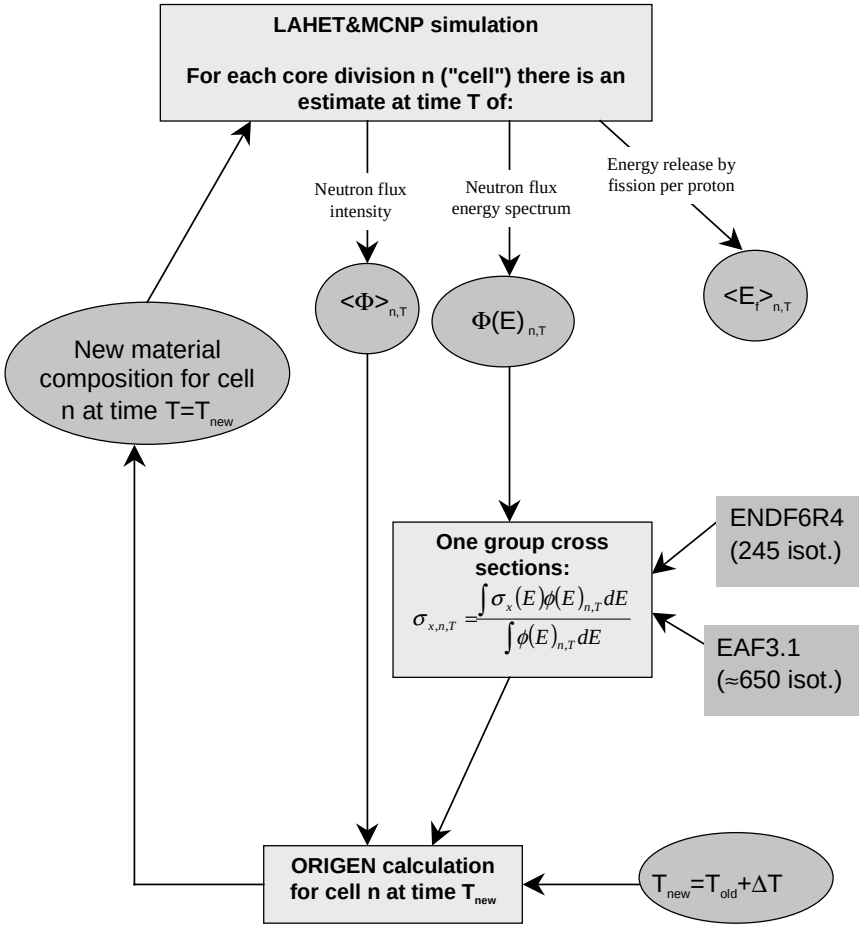
The simulations have been done using a combination of the following codes:

- NJOY94.61 for nuclear data processing [5]. The database used is the ENDF6R4 for neutron transport calculations and reaction rates calculation. During the simulations data of 245 isotopes have been used, being 196 fission fragments, and representing not less than 99% of the total mass inventory. The EAF3.1 database (about 650 isotopes) has been used for several reaction rates calculation whenever the isotopes considered are not available in the ENDF6R4 library. To handle with it, the EAF3.1 library has been converted to ENDF format.
- LAHET [6] for the simulation by Monte-Carlo of the proton beam interaction with lead. This code calculates the external neutron source.
- MCNP4B [7] for the complete neutron simulation of the spallation source produced by LAHET, including all the neutron progeny via any multiplication reaction below 20 MeV. It calculates the neutron multiplication, the energy release by fission that permits to know the beam intensity needed to work under a nominal power output, the neutron flux and specific power core distributions and the neutron flux energy spectra at every core positions.
- ORIGEN2.1 [3] for any burnup calculation.

The aim of the code combination is to perform a coupled neutronic and isotopic time evolution calculation, where the neutronics and fuel depletion simulation tools shared all the necessary data. The simulation procedure is as follows (figure 2):

- A mesh division of the EA core is performed, where each fuel element is divided into 10 axial zones. Each core division will have its own neutron flux estimate, energy release by fission and neutron flux energy spectrum estimators.
- Using LAHET and MCNP4B a complete simulation of the neutronics in steady state at considered conditions is done, obtaining the desired estimates.
- The following step is a set of burnup calculations, performing one for each core division with the previous resulted neutron fluxes. Special ad-hoc one-group integrated cross sections libraries are written in ORIGEN2.1 format and used for every burnup calculation. The one-group cross sections are obtained integrating the cross sections weighted by the spectrum obtained by MCNP4B for each core zone. The burn up time step considered is of the order of a few days.
- After all ORIGEN2.1 calculations have finished, an automatic procedure developed by FACET group translated the ORIGEN2.1 material descriptions formats to MCNP4B formats, creating a new MCNP input data file with an update of material composition for each core division.
- A new time step is performed beginning a new neutron transport simulation.

Figure 2 **Combined neutronic and isotopic combined time evolution calculation scheme**



During all the steps ORIGEN2.1 and MCNP4B shared the neutron flux data and material composition for all the core regions. Figure 2 shows a scheme of the procedure.

Definition of ADS parameters of interest for a performance evaluation

The main interest of the ADS in general and the EA in particular is their utilisation for TRUs transmutation. Therefore elimination efficiency parameters should be defined for system comparison purposes. In addition, from the neutronics point of view the most important aspects of the ADS performance are related with the accelerator requirements, once a fixed accelerator operation range is considered the maximum achievable burnup is fixed. Another EA relevant performance parameter is the energy gained by fission per unit of energy consumed in the accelerated beam.

The simplest way of defining the elimination efficiency is:

$$\eta = \frac{N_0 - N_f}{N_0} \quad \frac{1}{Time} \; ; \; \tau = \frac{1}{\eta}$$

Where N_0 is the initial load; N_f is the remaining amount after irradiation and Time is the burnup period; τ could be defined as the characteristic elimination time. The Time units can be also burnup units as GWdt. This formula is valid whenever the isotope disappearing could be adjusted to a linear fit. This is not the usual case, normally the isotopic transmutations follows at least an exponential fit, if not some other more complex laws. Therefore, another calculation procedure for the elimination efficiency considers that the isotope time evolution is an exponential function, giving:

$$N_f = N_0 \exp(-\eta \times Time) \; ; \; \eta = -\ln\left(\frac{N_f}{N_0}\right)\frac{1}{Time}; \; \tau = \frac{1}{\eta}$$

Again τ is the characteristic elimination time (burnup) as the time needed to reduce the isotope inventory in a factor e. The lower τ values mean the higher elimination capacity. Expressing τ in GWdt units different thermal power systems can be compared.

For isotopes that follows non-simple exponential function laws the parameters η and τ are not constant during the entire burnup considered. But in any case, for comparison purposes at equal burnup intervals, these parameters could be considered as logarithmic effective elimination time constants.

The transmutation is not possible under non-appropriate neutronic conditions. Considering that the external source is responsible of maintaining the operating regime, assuming a desired burnup period with the EA working at constant power output means that the ADS accelerator should provide the current needed at any time. The accelerator performance would be defined as the maximum and minimum beam intensity, binding its operational range. The lower ratio implies the less demanded accelerator requirements, with its foreseen impact on cost and maintenance.

Another important parameter in the ADS neutronics study is the evolution of its neutron net multiplication (M) and its multiplication constant with external source (k_s), defined both as:

$$M = \frac{n_{n,f} + n_{n,xn} + n_s}{n_s} \; ; \; k_s = 1 - \frac{1}{M}$$

Where $n_{n,f}$ is neutron fission production per proton at steady state, $n_{n,xn}$ is the neutron production by reaction such as (n,2n) or (n,3n) per proton and n_s is the spallation neutron source per proton. During all the burnup the EA should be in subcritical state with a reactivity margin that is fixed by security limitations. In Montecarlo simulations of ADS, the numbers $n_{n,f}$ and $n_{n,xn}$ can be estimated either directly (by counting the number of particles transported) or indirectly using track-length estimators.

The concept of energy gain is related with the neutron net multiplication per proton. The energy gain is the energy produced per unit of energy transported by the proton beam E_p:

$$G = \frac{\sum_{i=1}^{N}\left(n_{n,fi}/\nu_i\right)\times\left(E_{fi}[MeV]\right)}{\left(E_p[MeV]\right)}\ ;$$

$$\sum_{i=1}^{N}\left(n_{n,fi}/\nu_i\right) = n_{n,f}/\nu\ ;\ \langle E_f\rangle = \frac{\sum_{i=1}^{N}\left(n_{n,fi}/\nu_i\right)\times\left(E_{fi}[MeV]\right)}{n_{n,f}/\nu};$$

$$n_{n,f} + n_{n,xn} + n_s \approx n_{n,f} + n_s \Rightarrow G \approx \frac{\langle E_f\rangle}{E_p\nu}\times(M-1)\times n_s$$

$$G = \frac{\langle E_f\rangle\times n_s}{E_p\nu}\frac{k_s}{1-k_s} = G_o\frac{k_s}{1-k_s}$$

Where G_0 is constant that depends on the nature of the fissile material loaded at EA core and the characteristics of the proton beam used; $n_{n,fi}$ are the neutrons per source proton that produce fission of isotope i; E_{fi} is the energy release by fission of isotope i and $<E_f>$ is the energy release by fission averaged over all fissile isotopes weighted by their fission probability in the considered system. With these performance parameter definitions, both transmutation efficiency and operational valid range comparisons could be performed between different EA configurations.

Effect of the thermal power output on the neutronics and transmutation parameters

The first presented comparison is the thermal power output effect on the EA performance. The 200 MW EA (case 1 of table 1) and 800 MW EA (case 2 of table 1) have been simulated and analysed for this purpose. The differences in their initial properties are the thermal power considered (200 MW and 800 MW respectively) and the proton beam energy (380 MeV and 1 GeV). The beam current is supposed to vary adequately for constant power output working regime. The initial fuel composition is the same in both cases (see tables 1 and 2) and also they shared the same geometry characteristics.

The time evolution of the main neutronics parameters (k_s, beam current intensity, energy gain and G_0) are shown in figures 3 and 4. The required accelerator operational limits are compared in table 4, where the G_0 has been assumed as constant with time for each model. It is clear than the lowest k_s evolution point has the lowest energy gain and the highest beam intensity demand, and reciprocally the highest k_s point has the lowest beam intensity and the highest energy gain.

As can be seen in figure 3, in the 200 MW EA, after 2000 days of burnup, the maximum beam intensity required was near the starting point (150 days). This means than the burnup could be still extended far from 2000 days from the point of view of accelerator performance. This is not the case of the results presented in figure 4 for the 800 MW EA, where after 1000 days of burnup the beam intensity begins to grow almost linearly. After 1400 days of burnup, time that is assumed as the reference time for transmutation performance analysis, the necessary current reaches a maximum of 18.30 mA. On the other hand, considering the different thermal power outputs, it should be noted that 2000 days of burnup in the 200 MW EA is equivalent to 500 days in 200 MW EA. The maximums over minimum ratios for the required accelerator beam intensity are 1.92 in the 200 MW EA model and 1.99 in the case of 800 MW EA.

Table 4. **EA neutronics parameters at accelerator intensity highest demand, lowest demand and initial configurations for the two models (200 MW and 800 MW)**

	Max. Intensity	**Min. Intensity**	**Initial**
		200 MWth	
K_s	0.9669 ± 0.0026	0.9827 ± 0.0017	0.9714 ± 0.0024
I(mA)	15.7 ± 1.2	8.2 ± 0.7	13.6 ± 1.0
Energy gain	33.5 ± 2.4	64.3 ± 5.9	38.7 ± 3.0
G_0		1.12	
Time (days)	150	1000	
		800 MWth	
K_s	0.9505 ± 0.0021	0.9753 ± 0.0018	0.9706 ± 0.0014
I(mA)	18.3 ± 0.5	9.2 ± 0.4	11.1 ± 0.4
Energy gain	43.7 ± 1.2	87.8 ± 3.2	71.9 ± 2.4
G_0		2.21	
Time (days)	1400	600	

Figure 3. **Time evolution of the k_s, beam intensity, energy gain and G_0 parameter for the case 1 of table 1**

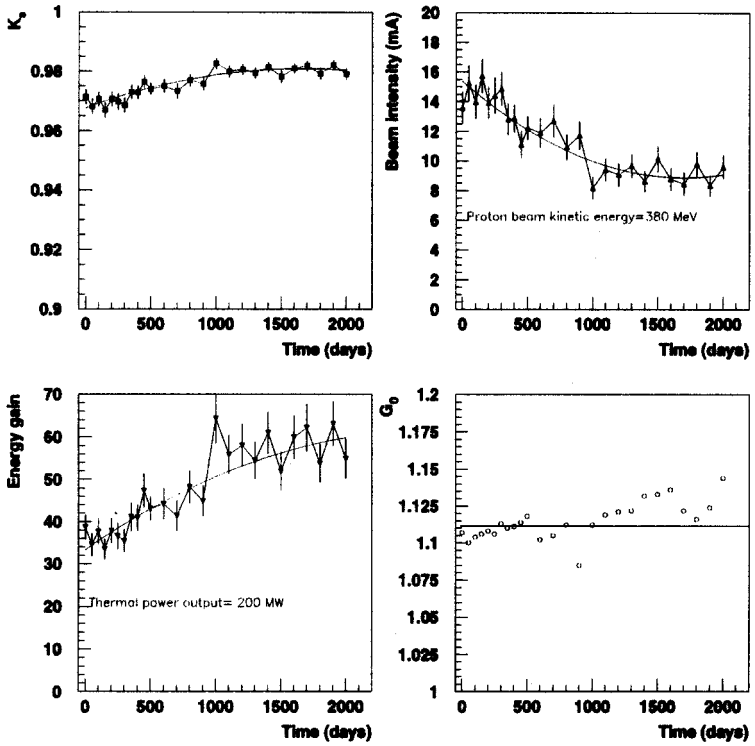


Figure 4 **Time evolution of the k_s, beam intensity, energy gain and G_0 parameter for the case 2 of table 1**

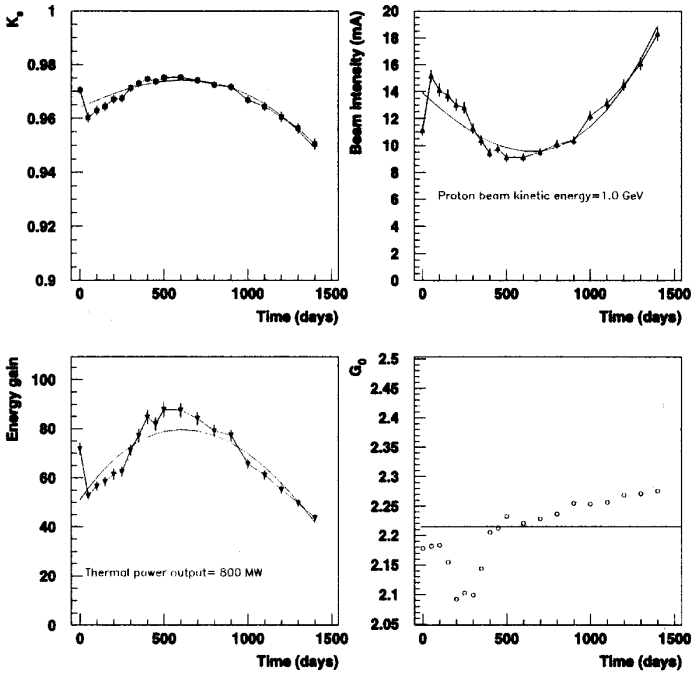


Figure 5. **Main plutonium isotopes time evolution for the EA 200 MW of case 1**

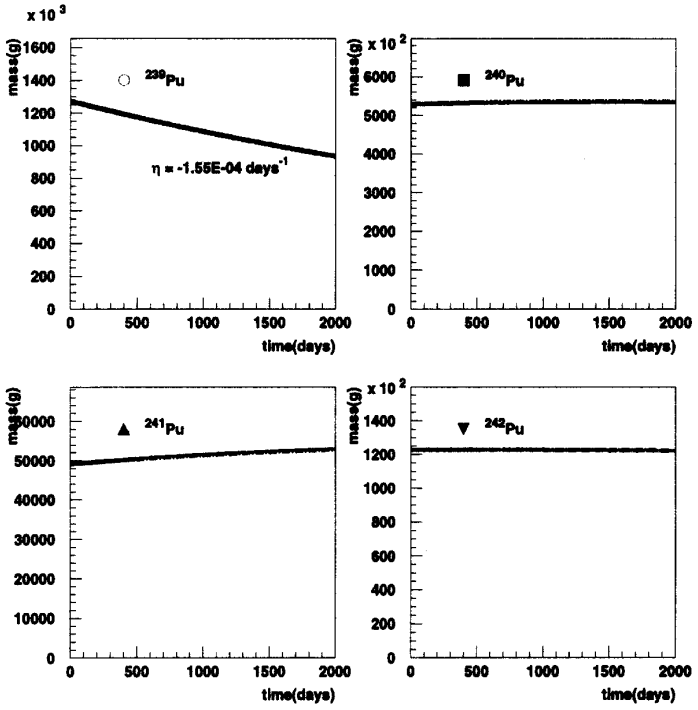
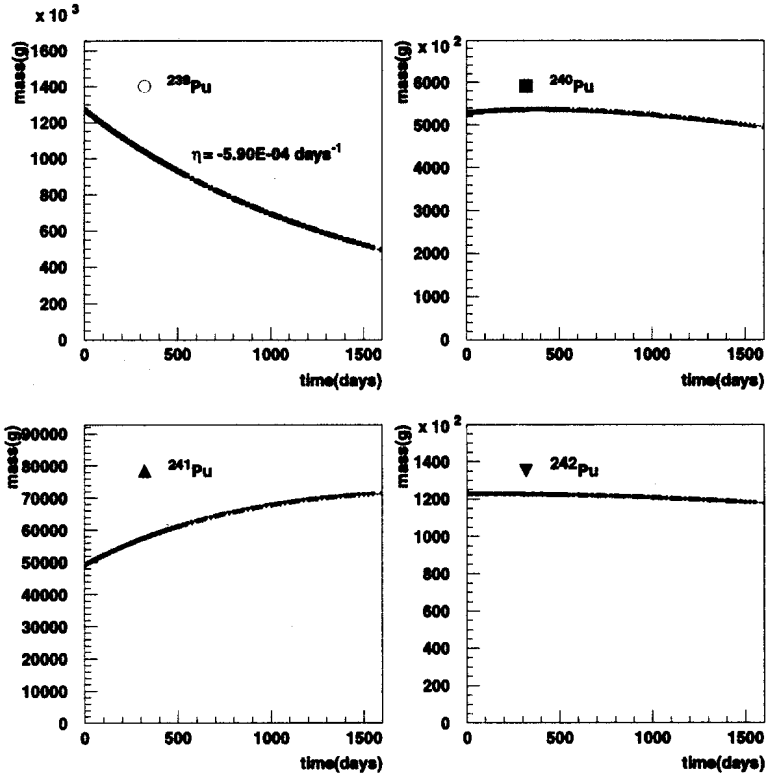


Figure 6. **Main plutonium isotopes time evolution for the EA 800 MW of model 2**



Considering the transmutation performance, the main plutonium isotopes evolutions with time appear in figure 5 for the 200 MW case and figure 6 for the 800 MW model. In both models the ^{239}Pu is eliminated at a rate that depends on achieved average fuel burnup. An exponential function adjustment can be done in both ^{239}Pu-evolution curves, giving characteristic transmutation time constants τ of 147.18 GWdt and 153.1 GWdt for 200 MW and 800 MW models respectively. These values are the average fuel burnup necessary for a ^{239}Pu depletion factor of e. In the case of the 800 MW EA, the average burnup after 1400 days has been of 127.4 GWdt, and the remaining amount of ^{239}Pu is near 0.43 times the initial load. Considering the 200 MW model, the average burnup after 1400 days is 32 GWdt, and the remaining ^{239}Pu is about 0.78 times the initial load.

In the 200 MW case, the ^{240}Pu, ^{241}Pu and ^{242}Pu isotopic concentrations grow slightly or are near by constant. On the other hand, in the 800 MW the ^{241}Pu mass grows up to 1.44 times the initial value, while both ^{240}Pu and ^{242}Pu isotopes slightly decrease.

In addition, both in the 200 MW_{th} and 800 MW_{th} EA cores, the ^{237}Np and ^{241}Am isotopes are eliminated, as figures 7 and 8 show. There is a rapid increase in the ^{242}Cm concentration, due to the ^{242}Am decay. The ^{242}Am are produced by ^{241}Am (n,γ) reactions that with some branching ratio produces ground state ^{242}Am. Another curium isotope that grows in some substantial way is the ^{244}Cm. In the model 2, after 1400 days of operation, the ^{244}Cm concentration grows more than twice and in the model 1 the increase is of 15%. In any case the total curium inventory increase is quite modest (in the 800 MW EA the final curium inventory is 1% of the remaining TRUs).

As has been said, the EA is a breeder ADS. Basically, the main breeding reaction is the ^{232}Th conversion to ^{233}U by neutron capture followed by two β decays (^{233}U and ^{233}Pa). Figures 9 and 10 summarised the ^{232}Th to ^{233}U breeding process rates for cases 1 and 2 respectively. In the 200 MW_{th} EA the ^{232}Th characteristic disappearing constant is 693 GWdt, being the ^{233}U conversion rate of 1.13×10^{-3} kg per kg of ^{232}Th and GWdt. The ^{233}Pa to ^{232}Th ratio is almost constant after near 200 days, being this equilibrium ratio of about 9×10^{-4}. The results for the 800 MW_{th} EA are shown in figure 10, where the ^{232}Th characteristic disappearing constant is 903.6 GWdt, and the ^{233}U conversion rate is 6.26×10^{-4} kg per kg of ^{232}Th and GWdt. The differences are because the ^{233}U production is less efficient with increasing irradiation time. The ^{233}U inventory growth is limited to a maximum given by the ^{233}U/^{232}Th asymptotic equilibrium. The ^{233}Pa to ^{232}Th ratio is almost constant after about 200 days, being this equilibrium ratio of near 3.9×10^{-3}. As expected this ^{233}Pa over ^{232}Th ratio is neutron flux intensity dependent. In addition, the ^{242}Cm over total curium inventory ratio are shown in figures 9 and 10. For case 1 this level reaches a maximum of near 0.68 after about 600 days, and in the model 2 it grows up to 0.85 after 250 days, time when the ^{242}Cm mass begins to decrease.

Table 5 summarised the transuranics evolution for the two considered cases. The elimination constant has been calculated using the equation of 3 instead of exponential adjustment to the evolution data.

Figure 7. **Time evolution of ^{237}Np, ^{241}Am, ^{242}Cm and ^{244}Cm for the EA 200 MW of case 1**

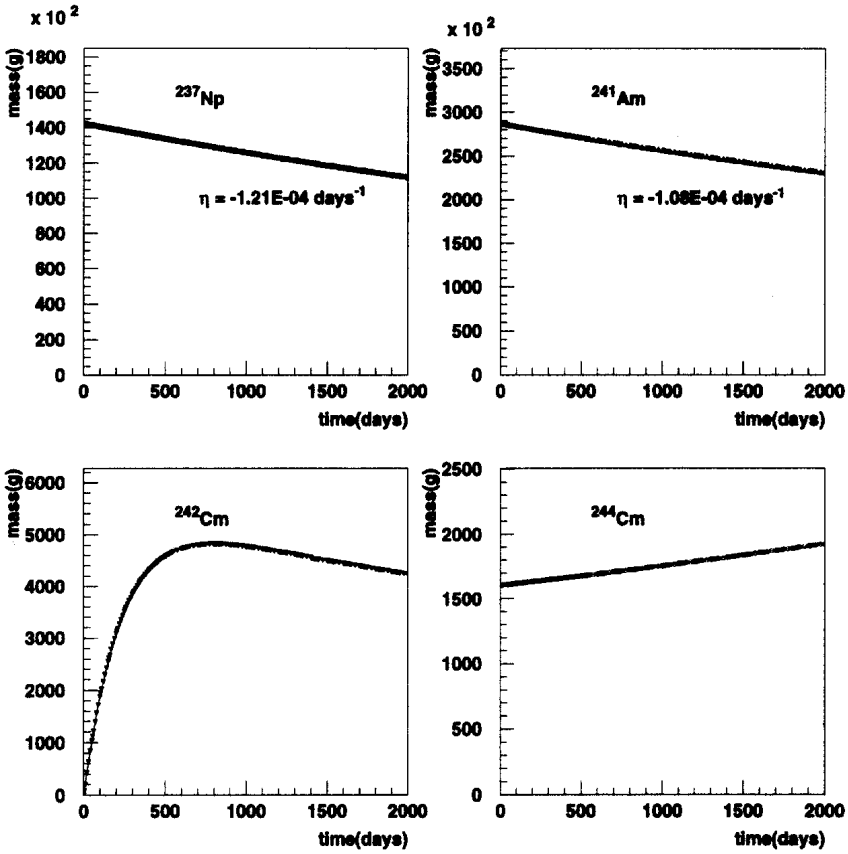


Figure 8 **Time evolution of ^{237}Np, ^{241}Am, ^{242}Cm and ^{244}Cm for the EA 800 MW of case 2**

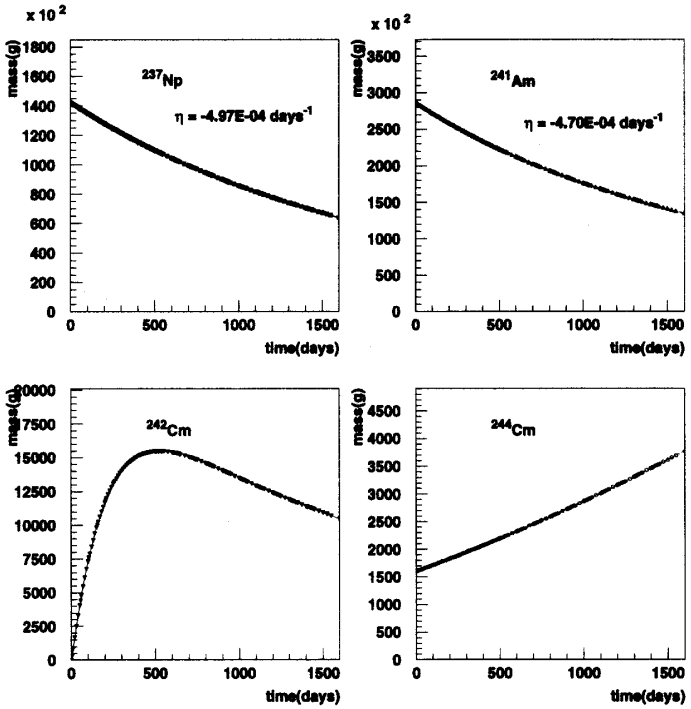


Figure 9 **Time evolution of ^{233}U, ^{233}Pa to ^{232}Th ratio, ^{233}U to ^{232}Th ratio and ^{242}Cm to total curium inventory ratio for the EA 200 MW of case 1**

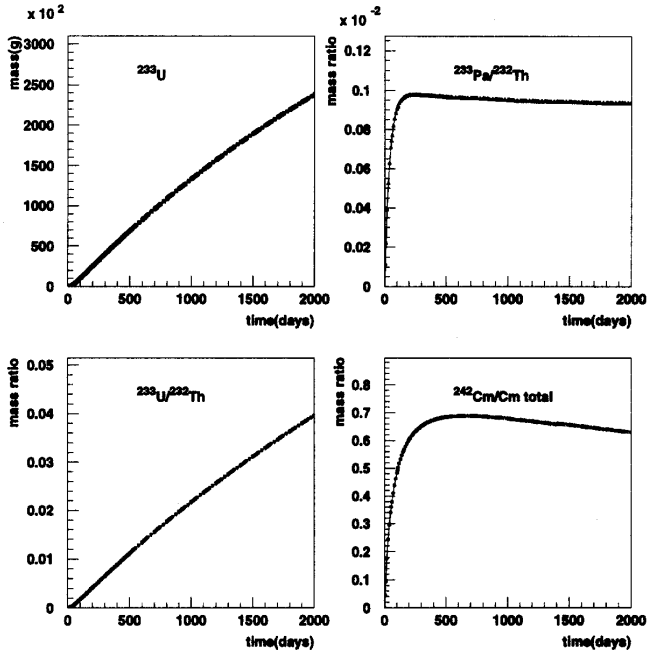


Figure 10 **Time evolution of ^{233}U, ^{233}Pa to ^{232}Th ratio, ^{233}U to ^{232}Th ratio and ^{242}Cm to total curium inventory ratio for the EA 800 MW of case 2**

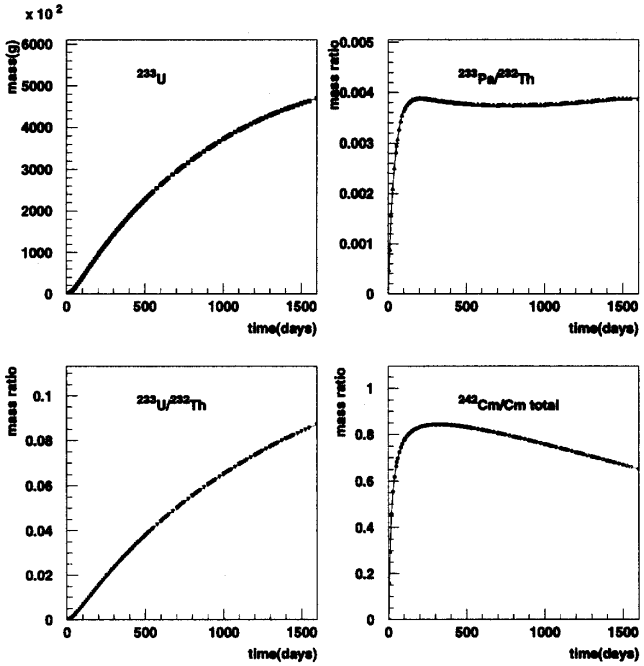


Table 5 **Time evolution parameters of the main transuranics in the 200 MWth EA model**

Isotope	Initial (g)	Final (g)	kg /kg initial/ GWdt	Elimination constant (y)	Elimination constant (GWdt)
^{237}Np	1.42E+05	1.20E+05	-4.95E-03	2.24E+01	1.86E+02
^{238}Pu	3.33E+04	6.70E+04	3.18E-02	—	—
^{239}Pu	1.27E+06	1.02E+06	-6.20E-03	1.74E+01	1.45E+02
^{240}Pu	5.29E+05	5.37E+05	4.50E-04	—	—
^{241}Pu	4.91E+04	5.20E+04	1.86E-03	—	—
^{242}Pu	1.23E+05	1.23E+05	-6.54E-05	1.84E+03	1.53E+04
^{241}Am	2.87E+05	2.46E+05	-4.52E-03	2.47E+01	2.05E+02
^{242}Am	0.00E+00	2.20E+01	—	—	—
^{242*}Am	2.11E+02	5.65E+03	8.10E-01	—	—
^{243}Am	2.42E+04	2.88E+04	5.98E-03	—	—
^{242}Cm	5.10E-01	4.54E+03	2.79E+02	—	—
^{243}Cm	4.52E+01	1.47E+02	7.09E-02	—	—
^{244}Cm	1.61E+03	1.82E+03	4.28E-03	—	—
^{245}Cm	3.02E+02	3.59E+02	6.03E-03	—	—
^{246}Cm	3.41E+01	4.03E-02	-3.14E-02	5.69E-01	4.73E+00
^{247}Cm	3.39E-01	1.77E-06	-3.14E-02	3.15E-01	2.62E+00

Table 5 (cont.) **Time evolution parameters of the main transuranics in the 800 MW_{th} EA model**

Isotope	Initial (g)	Final (g)	kg /kg initial/ GWdt	Elimination constant (y)	Elimination constant (GWdt)
^{237}Np	1.42E+05	7.06E+04	-1.80E-02	5.48E+00	1.82E+02
^{238}Pu	3.33E+04	1.09E+05	8.15E-02	—	—
^{239}Pu	1.27E+06	5.53E+05	-2.02E-02	4.61E+00	1.53E+02
^{240}Pu	5.29E+05	5.06E+05	-1.53E-03	8.78E+01	2.92E+03
^{241}Pu	4.91E+04	7.06E+04	1.56E-02	—	—
^{242}Pu	1.23E+05	1.19E+05	-1.13E-03	1.20E+02	3.95E+03
^{241}Am	2.87E+05	1.48E+05	-1.73E-02	5.77E+00	1.92E+02
^{242}Am	0.00E+00	5.26E+01	—	—	—
^{242*}Am	2.11E+02	9.02E+03	1.49E+00	—	—
^{243}Am	2.42E+04	4.17E+04	2.57E-02	—	—
^{242}Cm	5.10E-01	1.14E+04	7.99E+02	—	—
^{243}Cm	4.52E+01	9.58E+02	7.20E-01	—	—
^{244}Cm	1.61E+03	3.46E+03	4.13E-02	—	—
^{245}Cm	3.02E+02	6.65E+02	4.31E-02	—	—
^{246}Cm	3.41E+01	8.35E+01	5.18E-02	—	—
^{247}Cm	3.39E-01	3.79E+00	3.63E-01	—	—

Effect of the Delay Time between the PWR Spent Fuel Discharge and the EA Fuel Load on the Neutronics and Transmutation Parameters

As has been explained in section 2, another parameter of interest is the influence of the transuranics composition in the EA performance. For this purpose three different EA models have been considered (cases 2, 3 and 4 of table 1). The differences between the three are the TRUs load compositions that appear in table 2. All the three cases have a thermal power output of 800 MW.

From the point of view of time evolution of some neutronics parameters, figure 11 and 12 show the simulation results for cases 3 and 4 respectively. Case 2 results are presented in figure 4. As can be seen in figure 12, for the case 4 there is a progressive fall of the k_s and an increase of beam intensity demand. This means that the new fissile material breeding is not able to compensate reactivity fall because of fissile TRU burning. The beam intensity maximum to minimum ratio is as high as 3.86, considering that the irradiation period is of 1400 days.

Figure 11 indicates that for case 3 there is a reactivity recovery similar to that of case 2 (figure 4). The beam intensity maximum to minimum ratio is 1.78, less than in case 2 (1.99), with a maximum k_s of 0.9643 taking place at 600 days of burnup (also 600 days in the model 2) and a minimum at the end of the cycle (1400 days) of 0.9384.

Table 6 summarised the transmutation performance parameters for the main TRUs loaded in the EA model 3 and 4. A comparison of the characteristic elimination constants indicates that for ^{237}Np (value around 178 GWdt), ^{239}Pu (150 GWdt) and ^{240}Pu (2950 GWdt) are near the same for the three models (with differences smaller than 5%). For ^{241}Am this elimination is clearly a function of its initial concentration: while for the cases 2 and 3 the value is almost constant of 195 GWdt, for case 4 it rises to a value of 255 GWdt.

Evolution of TRU Transmutation Efficiency with the EA Burnup Cycle

Among the transmutation strategies there are two options for closing the fuel cycle:

- The one-through irradiation step: In this option the TRUs loaded fuel is burned up only once. The discharge will be stored almost directly in the secular repository.
- The closed cycle: Every spent fuel discharge is reprocessed and the remaining TRUs are reloaded in the transmutation system. This strategy allows increasing the final transmutation efficiency.

Basically the EA is designed to work in a TRUs transmutation closed cycle [2]. This strategy implies that every EA discharge will be reprocessed. In the case of the EA this reprocessing would produce four waste streams: fission and activation products, remaining ^{232}Th, produced ^{233}U and remaining TRUs. The ^{233}U is separated for other purposes; the remaining TRU are recovered altogether and mixed with the adequate amount of ^{232}Th and fresh TRUs coming from LWR spent fuel. The manufactured fuel should be able to maintain the nominal initial subcritical level once loaded in the new EA core. The fission and activation products stream will be processed for proper storage or eventually some LLFF can be also eliminated in appropriated devices (e.g. the EA core periphery [2]).

Figure 11 **Time evolution of the k_s, beam intensity, energy gain and G_0 parameter for the case 3 of table 1**

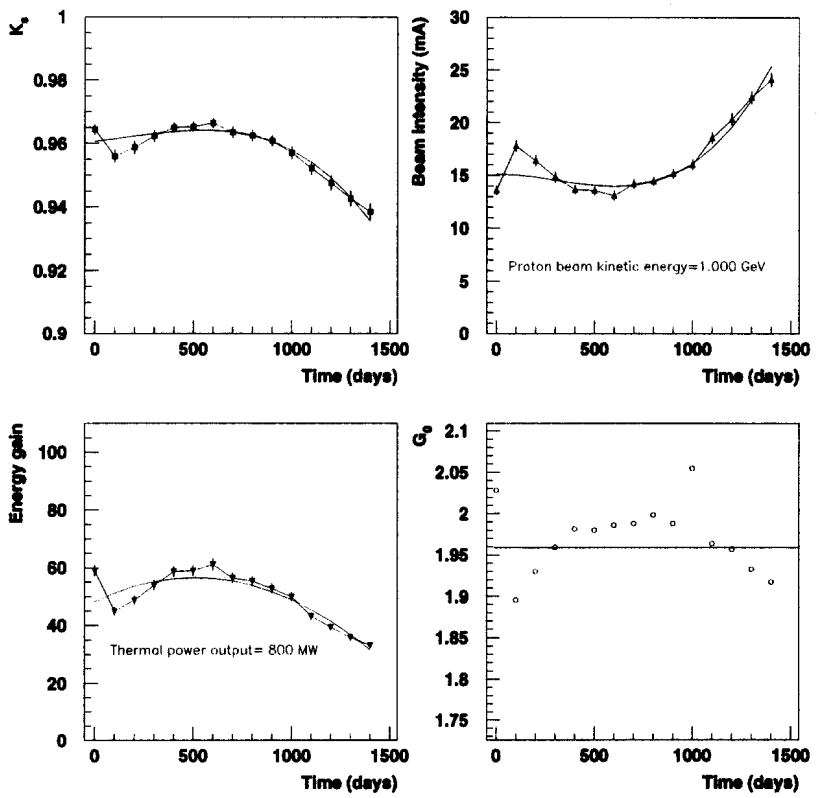


Figure 12 **Time evolution of the k_s, beam intensity, energy gain and G_0 parameter for the case 4 of table 1**

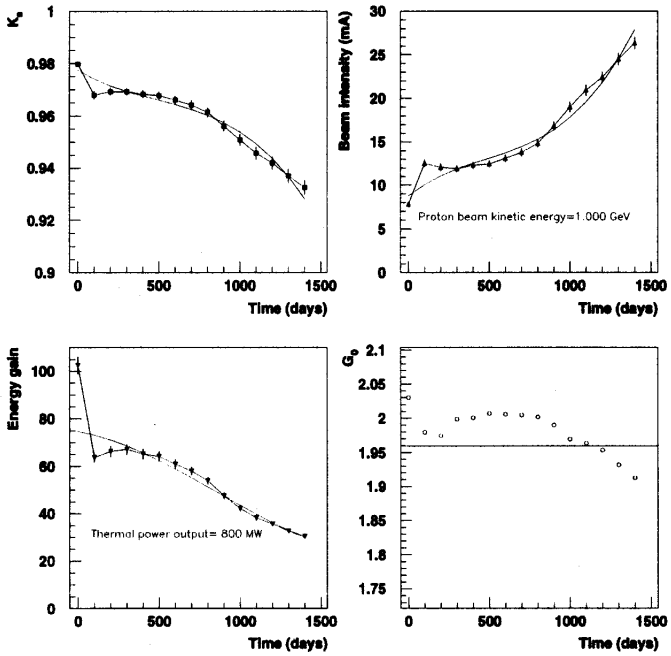


Table 6 **Time evolution parameters of the main transuranics in the 800 MW_{th}-25 years decay time EA model**

Model 3	800 MW		25 years decay time		
Isotope	**Initial (g)**	**Final (g)**	**kg /kg initial/ GWdt**	**Elimination constant (y)**	**Elimination constant (GWdt)**
^{237}Np	1.27E+05	6.21E+04	-4.01E-03	5.36E+00	1.78E+02
^{238}Pu	3.52E+04	9.62E+04	1.36E-02	—	—
^{239}Pu	1.19E+06	5.10E+05	-4.49E-03	4.53E+00	1.50E+02
^{240}Pu	4.95E+05	4.74E+05	-3.37E-04	8.75E+01	2.91E+03
^{241}Pu	9.43E+04	7.97E+04	-1.22E-03	2.28E+01	7.58E+02
^{242}Pu	1.15E+05	1.13E+05	-1.27E-04	2.35E+02	7.82E+03
^{241}Am	2.27E+05	1.19E+05	-3.73E-03	5.96E+00	1.98E+02
^{242}Am	0.00E+00	4.34E+01	—	—	—
^{242*}Am	2.10E+02	7.26E+03	2.63E-01	—	—
^{243}Am	2.27E+04	3.97E+04	5.89E-03	—	—
^{242}Cm	5.48E-01	9.38E+03	1.34E+02	—	—
^{243}Cm	6.20E+01	8.12E+02	9.49E-02	—	—
^{244}Cm	2.67E+03	3.94E+03	3.72E-03	—	—
^{245}Cm	2.83E+02	8.09E+02	1.46E-02	—	—
^{246}Cm	3.20E+01	9.52E+01	1.55E-02	—	—
^{247}Cm	3.18E-01	4.19E+00	9.55E-02	—	—

Table 6 (cont.) **Time evolution parameters of the main transuranics in the 800 MW_{th}-10 years decay time EA model**

Model 4	800 MW		10 years decay time		
Isotope	**Initial (g)**	**Final (g)**	**kg /kg initial/ GWdt**	**Elimination constant (y)**	**Elimination constant (GWdt)**
^{237}Np	1.14E+05	5.46E+04	-4.07E-03	5.22E+00	1.74E+02
^{238}Pu	3.66E+04	7.52E+04	8.23E-03	—	—
^{239}Pu	1.10E+06	4.64E+05	-4.53E-03	4.43E+00	1.48E+02
^{240}Pu	4.57E+05	4.38E+05	-3.25E-04	9.05E+01	3.02E+03
^{241}Pu	1.80E+05	9.86E+04	-3.54E-03	6.37E+00	2.12E+02
^{242}Pu	1.06E+05	1.08E+05	7.93E-05	—	—
^{241}Am	1.21E+05	7.33E+04	-3.09E-03	7.64E+00	2.55E+02
^{242}Am	0.00E+00	2.75E+01	—	—	—
^{242*}Am	2.10E+02	4.29E+03	1.52E-01	—	—
^{243}Am	2.11E+04	3.73E+04	6.01E-03	—	—
^{242}Cm	5.48E-01	5.81E+03	8.29E+01	—	—
^{243}Cm	8.21E+01	5.00E+02	3.98E-02	—	—
^{244}Cm	4.39E+03	4.68E+03	5.19E-04	—	—
^{245}Cm	2.62E+02	1.03E+03	2.30E-02	—	—
^{246}Cm	2.97E+01	1.14E+02	2.21E-02	—	—
^{247}Cm	2.94E-01	4.77E+00	1.19E-01	—	—

A simulation of this strategy applied to the EA model 2 (800 MW_{th} EA) has been performed under the considered hypothesis for six more burnup cycles. The first cycle was of 1400 days while the following six cycles was simulated for a burnup period of 1500 days. The resulting cumulative TRUs removals as a function of the number of cycle are presented in figure 13. As can be seen, there is substantial cumulative elimination of ^{239}Pu, ^{241}Am, ^{240}Pu, ^{237}Np and ^{242}Pu which grow almost linearly with the number of cycles. The ^{243}Am inventory increases from a production of 17.4 kg at the end of the first cycle up to 128.7 kg of cumulative production and the end of cycle seven.

Special behaviours come into view for the ^{238}Pu and the ^{241}Pu evolutions: both at the end of cycle number three starts to disappear, beginning with two production cycles. The ^{238}Pu is the main product of ^{237}Np conversion by neutron irradiation and the ^{241}Pu is produced by neutron capture in ^{240}Pu. These results indicate that both actinides have reached equilibrium ratios with their parents after the end of cycle three. In the case of ^{242}Cm and ^{244}Cm there is a linear increase of their cumulative production. Nevertheless, the produced masses are not comparable in scale with the transmutation rates of ^{239}Pu, ^{241}Am, ^{240}Pu and ^{237}Np.

The characteristic elimination constants for several TRUs of interest appear in figure 14. As can be seen, for ^{237}Np, ^{239}Pu and ^{241}Am these constants are nearly cycle independent, with average values of 0.19 $year^{-1}$ for ^{237}Np, 0.21 $year^{-1}$ for ^{239}Pu and 0.16 $year^{-1}$ for ^{241}Am. The differences with average values are in the three cases less than 5%. Therefore, if the loaded masses at the beginning of every cycle are near the same, as it is the case, the elimination achieved for the same burnup is the same, and the cumulative elimination is a linear function of the number of cycles, as figure 13 has revealed.

Conclusions

The aim of this study is to analyse the effects of the thermal power output level and the TRUs loaded in the fuel on the EA performance. Also a first approach to define the transmutation efficiency of an EA operated in closed cycle is presented.

When comparing 200 MW and 800 MW EA systems its neutron multiplication constant can be maintained in an acceptable operation range for long burnup periods (up to 110 GWdt in average) in both cases. This is due to the ^{233}U breeding from the ^{232}Th loaded within the EA fuel. Therefore, in both cases accelerator maximum to minimum beam intensity ratios are limited to less than 2. From the point of view of TRU transmutation, the characteristic elimination constants for ^{237}Np, ^{239}Pu and ^{241}Am are near to constant in units of GWdt (~180 GWdt, ~150 GWdt and ~195 GWdt respectively). This result implies that higher thermal power generation needs less burnup periods (in days) to achieve near to similar ^{237}Np, ^{239}Pu and ^{241}Am elimination.

The use of the different proposed TRUs mixtures for this study in the EA fuel load has slight effect on the performance from the neutronics point of view (e.g. the accelerator maximum to minimum beam intensity demand). The key is once again the ^{233}U-breeding time evolution that should compensate reactivity decrease due to fissile TRUs burn up. The transmutation performance could depend on the initial mass loaded of the actinide under study, as the ^{241}Am case reveal in this study.

On the other hand, a closed fuel cycle strategy can be designed for the EA with substantial cumulative elimination of ^{239}Pu, ^{241}Am, ^{240}Pu, ^{237}Np and ^{242}Pu which grow almost linearly with the number of cycles .

Figure 13a **Cumulative elimination for several TRUs of interest as a function of the number of cycle**

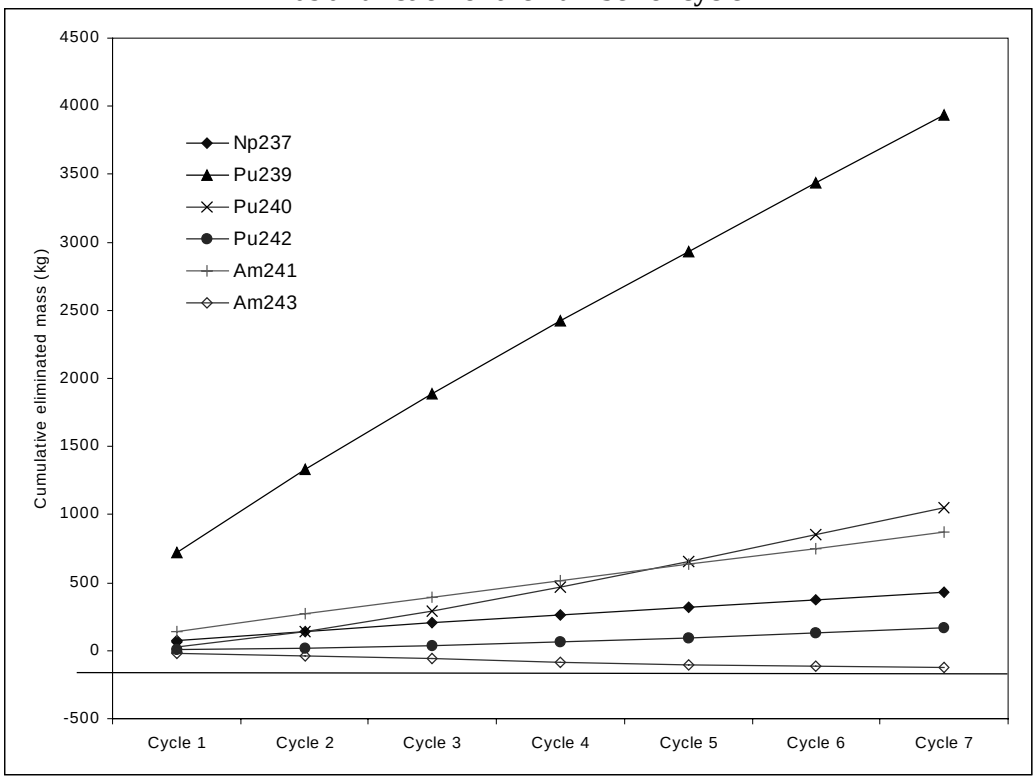


Figure 13b **Cumulative elimination for several TRUs of interest as a function of the number of cycle. (Note that there is a factor 40 reduction is the Y axis scale comparing with figure 13a)**

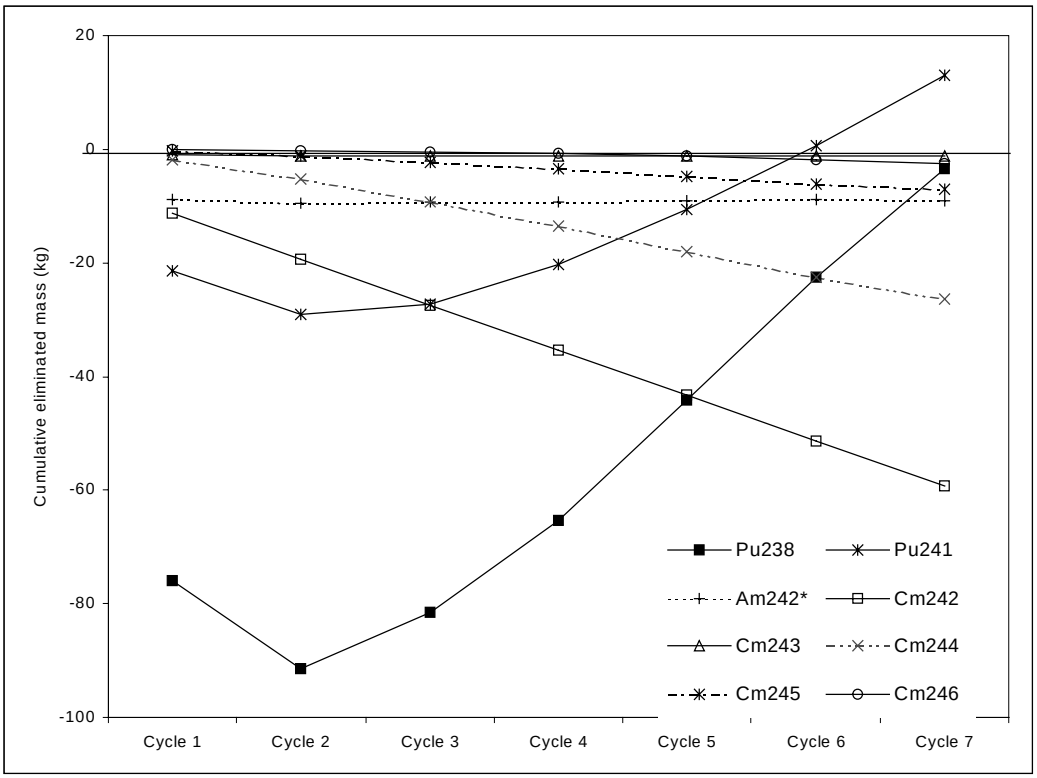
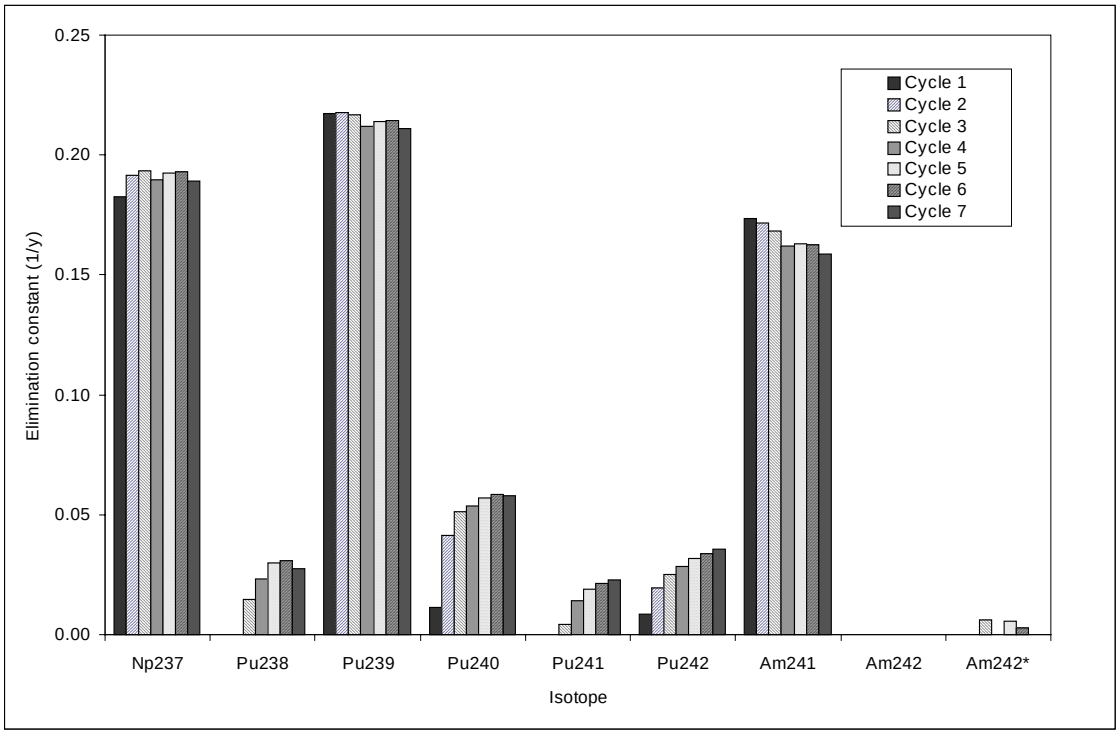


Figure 14. **Transmutation constants for the main TRUs of interest as a function of cycle number**



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