A STUDY ON ONCE-THROUGH ACTINIDES TRANSMUTATION

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Abstract

Radiotoxicity of long-lived radionuclei in the nuclear waste can be reduced and shortened by transmutation technologies. However, these technologies convey some additional risks in the nuclear fuel cycle activities carried out at present. From this point of view, minimisation of nuclear waste manipulation and reprocessing in the transmutation strategy in of primary interest. Because of the foregoing reasons, a once-through transmutation scheme is studied in this paper, aimed at identifying the main contributors to long-term radiotoxicity and the most suitable neutronic ways to eliminate them. First, an analysis based on theoretical spectra is used to scan the full range of possibilities in neutron-induced transmutation. Some relevant conclusions are derived from this analysis, within the once-trough framework. After that, a physical reactor is studied to obtain a minimum radiotoxicity in a once-through strategy. Pebble Bed Reactors present excellent characteristics for this strategy, since the fuel elements can withstand very high burn-ups, and it is possible to obtain different neutron spectra by changing some design parametes, as the fuel mass and active radius of the pebble. A double step transmutation option is presented, one as critical and one as a sub-critical accelerator-driven system (ADS). Very good results in long-term radiotoxicity reduction are obtained in these cases.

Introduction

Transmutation is one of the principle concerns in the actual nuclear science community. [1-4] One of the principle uncertainties in the transmutation concept is the reprocessing of nuclear fuel during the transmutation phase. The problem arise in the safety considerations, since we are adopting an strategy where a risk for the population in the actual time is increased as compared to the solution of a Deep Storage Facility (DSF.) Economic considerations have to be taken into account, since the cost of reprocessing is going to be very high. Also, problems concerning Cm reprocessing due to the high energy release by alfa particles emission limit the maximum amount of Cm to be reprocessed per reprocessing step. In standard transmutation approaches, a fast spectrum is adopted for actinides elimination. In some scenarios, it has been reported that more than 10 reprocessing steps will be necessary for an actinides elimination of 99%. A possible solution to minimise this problem is to use a fuel element for transmutation purposes that can achieve very high burn-ups, so only one reprocessing step will be necessary [10] (before starting the transmutation phase.) The problem of k_{ef} depletion can be solved with the use of a sub-critical assemblie (ADS) and a proper recycling strategy. To achieve this goal, a fuel element capable of withstanding very high burn-ups is needed. Coated fuel particles used in HTGR have the maximum burn-up limit reported, above 800 MWd/kg. In these studies, a maximum value of 700 MWd/kg has been taken. The final radiotoxicity is going to depend on the isotopic composition of the remanent spent fuel. The initial composition of the waste fuel charged in the transmutator is described in Table 1.

Table 1. **Isotopic composition of actinides in the LWR discharged fuel, after 40 MWd/kg burn-up and 15y cooling**

Isotope	gr/ton U	Fraction	Isotope	gr/ton U	Fraction
^{236}Np	5.312E-04	4.575E-08	^{242m}Am	2.452E+00	2.112E-04
^{237}Np	6.514E+02	5.610E-02	^{243}Am	1.446E+02	1.245E-02
^{238}Pu	2.277E+02	1.961E-02	^{242}Cm	5.933E-03	5.110E-07
^{239}Pu	5.912E+03	5.092E-01	^{243}Cm	4.326E-01	3.726E-05
^{240}Pu	2.593E+03	2.233E-01	^{244}Cm	3.090E+01	2.661E-03
^{241}Pu	6.823E+02	5.867E-02	^{245}Cm	2.339E+00	2.014E-04
^{242}Pu	5.983E+02	5.153E-02	^{246}Cm	3.165E-01	2.726E-05
^{244}Pu	4.176E-02	3.597E-06	^{247}Cm	3.656E-03	3.149E-07
^{241}Am	7.651E+02	6.590E-02	^{248}Cm	2.440E-04	2.101E-08

In the study, it has been supposed an open cycle policy, where Pu isotopes are taken as waste fuel, not as a possible nuclear fuel as is the case in France or Japan, where reprocessing of Pu isotopes to form MOX fuel is the actual policy. Only transuranics isotopes are taken from the spent fuel.

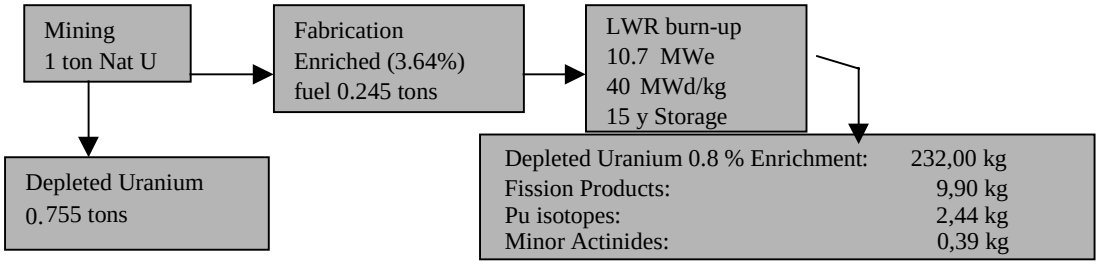
Characterisation of the problem

Radiotoxicity produced in actual Nuclear Power Plants comes form three different sources: actinides, fission products and activation products. Characterisation of all the elements contributing to the final radiotoxicity in the fuel cycle is extremely important, since this is the way to identify the main contributors to radiological hazard. Initially, the fuel cycle departs from the Natural Uranium that comes from mother nature (this radiotoxicity level should be accepted as a reference level.) The mass of every isotope present in a gram of Natural Uranium in secular equilibrium has been calculated starting from one gram of its standard composition (99.2890% $^{238}U_{92}$ and 0.7110% $^{235}U_{92}$). Results were obtained with Nuclides 2000 programme. [11] A total ingestion dose of 0.0197 Sv/gram is found.

This radiotoxicity value will be taken as our **reference level** for transmutation applications. Since all ingestion dose calculations have been carried out with **ORIGEN** code, [12] all the radiotoxicity results obtained in the calculations are in m^3 of Water, and the **reference level for Natural Uranium** will be **$1.572 \cdot 10^7$ m^3 water/ton.**

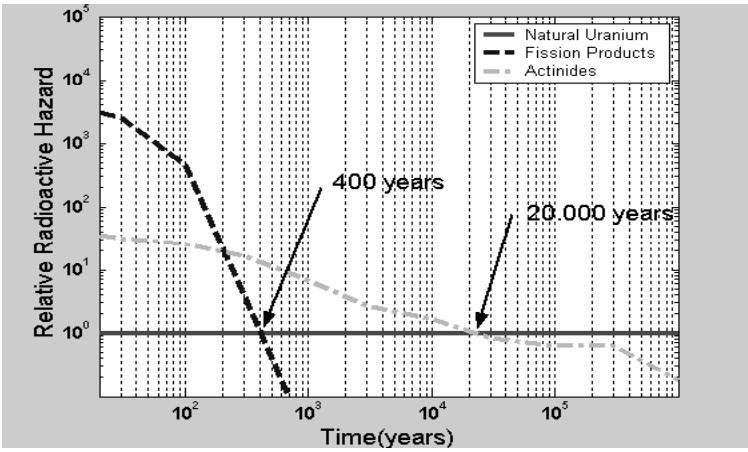
Our studies will focus on L.W.R. reactors (Pressure and Boiling reactors) enriched with 3.64% of $^{235}U_{92}$. Results will always be normalised to one tone of Natural Uranium needed to feed the nuclear cycle. The chart of the cycle is given in Figure 1.

Figure 1. **Nuclear waste produced by 1 ton of natural uranium**



The radiotoxicity evolution of the spent fuel can be represented for fission products and actinides in comparison with the radiotoxicity of one tone of natural uranium removed from earth to feed the nuclear cycle. Results are shown in Figure 2 (where radiotoxicity has been expressed in all cases as m^3 of water to attain maximum allowable concentrations.)

Figure 2. **Relative ingestion Effective Committed Dose (ECD) (relative to natural uranium level) of fission products and actinides**

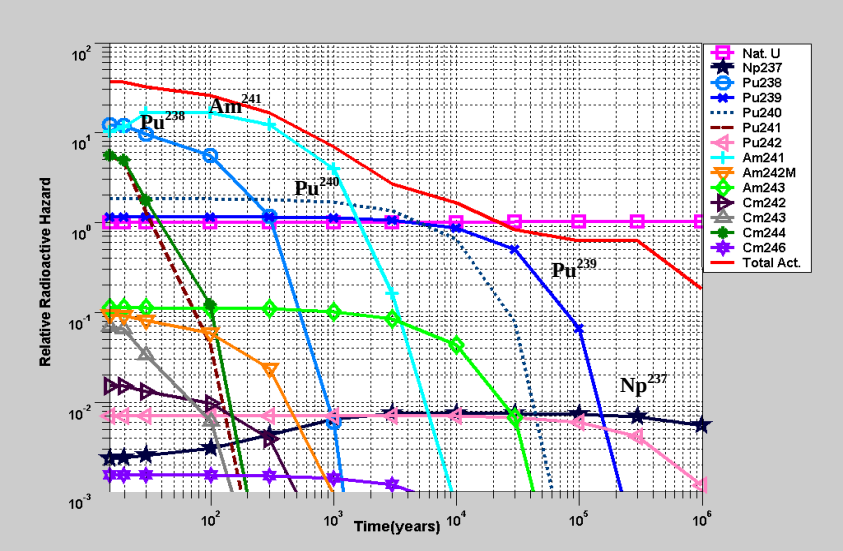


For tables and figures of Relative Radioactive Hazards, a reference level is used, which corresponds to the radiotoxicity of 1 ton of natural uranium (i.e., $1.572 \cdot 10^7$ m^3 of water.) All values for radiotoxicity refer to this level. For every nuclei, the radiotoxcicity is related to the mass generated in the nuclear fuel cycle starting from 1 ton of natural uranium.

From this figure, it can be seen that the main long-term problem is the actinides inventory. Actinides reach the reference radiotoxicity level (of Natural Uranium feeding the fuel cycle) after 20 000 years. Fission products reach the same radiotoxicity level in 400 years, a time span along which the control and confinement of those products could be done by technological systems.

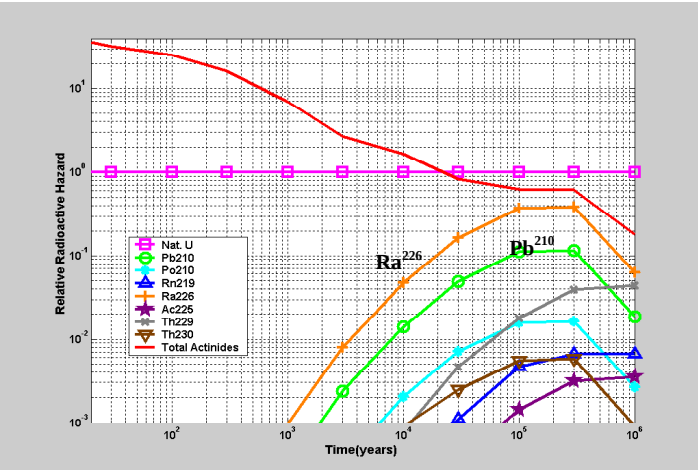
From this result, it is advisable to study in detail the actinides contribution to the total committed dose. Figure 3 shows the radiotoxicity evolution of the actinides discharged from the reactor, relative to Natural Uranium radiotoxicity needed to feed the nuclear cycle. From Figure 3, it can be seen that the higher contributors to the ingestion dose are ^{238}Pu, ^{241}Am (which is a daughter of ^{241}Pu), ^{240}Pu, ^{239}Pu and ^{237}Np.

Figure 3. **Evolution of relative ingestion Effective Committed Dose (relative to natural uranium level) for actinides discharged from LWR after 15y cooling**



If these initial actinides are not transmuted, they produce different disintegration chains. The principle contributors to the final dose of these daughters coming from the disintegration chains are represented in Figure 4:

Figure 4. **Evolution of relative ingestion Effective Committed Dose (relative to natural uranium level) for radioactive isotopes (daughters) coming from actinides disintegration**



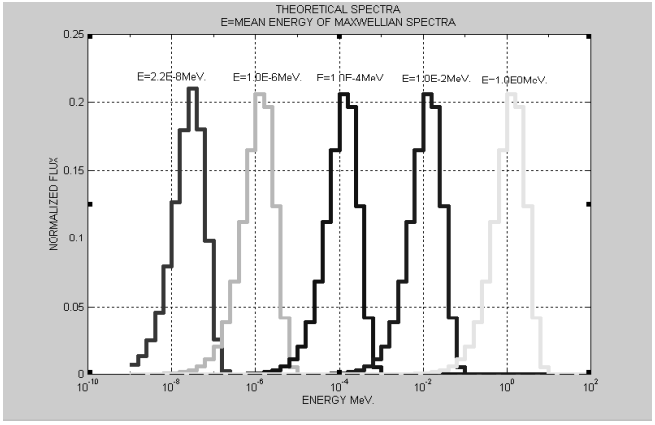
Those isotopes mainly come from three different chains. Isotopes with a higher effective dose contribution are **^{226}Ra** (which comes from ^{242}Pu disintegration chain, ^{238}Pu chain, and appears also in natural uranium (^{238}U chain) and **^{210}Pb** (which comes from the same chains.)

Radiotoxicity is thus dominated in medium and long term by **Pu isotopes** and descendants. The objective for transmutation is then to eliminate Pu isotopes without Am and Cm built up during burn-up.

Optimum spectra for a "once through strategy"

Finding an optimum spectrum for transmutation purposes has been discussed in different forums and benchmarks. [13-14] Since the way to transmute all isotopes in the fuel is to fission them, it seems obvious that the best spectrum to achieve this purpose is with a fast one (since some Cm isotopes produced in a thermal or epithermal spectra have very low absorption cross-sections in the thermal range). Additionally, the ratio of fission versus capture cross-sections is much higher in fast spectrum than in the rest of spectra. To feature these results by means of a theoretical exercise, 5 different theoretical spectra have been chosen. Spectra used in these calculations are depicted in Figure 5.

Figure 5. **Maxwellian spectra**



It is obvious that such spectra do not properly feature any nuclear reactor, because the spectrum shape mainly depends on the fission spectrum (rather hard) and the moderation process, not always reaching well-thermalised ranges. Maxwellian distributions only represents thermalised spectra at different temperatures, which is not a physical case for reactors. However, the systematic of using a Maxwellian spectrum at different temperatures is very useful to identify the importance of every range of energy in the quest of eliminating the most offending transuranics.

Effective cross-sections for all isotopes appearing during actinides transmutation have been calculated with each Maxwellian spectrum, using the MCNP code. [15] The spectral index σ_f/σ_c and the σ_f cross-section are given in Table 2 for important isotopes for transmutation purposes. The higher the spectral index, the better for transmutation purposes, since no higher actinides are created during the burn-up of the actinides. However, for a constant specific power in the fuel, all the spectra have the same transmutation capabilities, since we have the same number of fissions per time and unit mass. The problem with thermal spectrum is that some isotopes are nearly impossible to transmute (some Cm isotopes) with a regular neutron flux level, because of the low cross-sections of these isotopes for this spectrum.

From this Table 2, it can be seen that the higher spectral indexes are found for fast spectrum, since the fission cross-sections are usually higher than the capture cross-section for all isotopes. Theoretically this is the best spectrum for transmutation, but problems related with the number of reprocessing steps needed, high level and energy flux, as well as safety considerations have to be taken into account.

Table 2. **Effective cross-sections of important isotopes for transmutation purposes**

Isotope	Cross-Sect	0.022eV	1eV	100ev	10keV	1MeV
^{239}Pu	σ_f [barn]	**1.097E+03**	**3.850E+01**	**3.680E+00**	**2.640E+00**	**1.700E+00**
T½=24,100y	σ_f/σ_c	2.890E+00	1.650E+00	1.300E+00	1.420E+00	2.000E+01
^{240}Pu	σ_f [barn]	**9.300E-02**	**7.700E-01**	**7.500E-02**	**1.100E-01**	**9.800E-01**
τ½=6,560y	σ_f/σ_c	2.000E-04	2.000E-04	1.300E-03	7.000E-03	7.500E+00
^{241}Pu	σ_f [barn]	**1.500E+03**	**2.440E+02**	**5.130E+01**	**5.100E+00**	**1.700E+00**
τ½=14.y	σ_f/σ_c	2.600E+00	2.600E+00	3.500E+00	4.400E+00	1.310E+01
^{242}Pu	σ_f [barn]	**1.400E-03**	**2.300E-04**	**3.900E-03**	**1.600E-03**	**8.000E-01**
τ½=3.74ey5	σ_f/σ_c	5.500E-05	1.100E-06	1.600E-03	1.100E-02	6.700E+00
^{241}Am	σ_f [barn]	**4.800E+00**	**4.300E+00**	**3.300E-01**	**2.900E-02**	**8.600E-01**
τ½=43y	σf/σc	5.000E-03	5.900E-03	7.100E-03	5.900E-03	2.300E+00
^{244}Cm	σ_f [barn]	**9.000E-01**	**1.000E-01**	**9.300E-01**	**8.100E-02**	**1.180E+00**
τ½=18.1y	σ_f/σ_c	5.800E-02	3.700E-02	2.600E-02	3.600E-02	5.100E+00

To compare the different theoretical spectra defined above, we have to depart from the same power level, at least initially. The neutron flux level necessary to obtain this constant power (which means a constant actinides transmutation at the beginning) is going to be defined by the well known formula:

$$\textbf{Power Density (W/cm}^3\textbf{)} = C\cdot\sum_i N^i\cdot\sigma_f^i\cdot\phi \qquad (1)$$

Where 'i' refers to all the isotopes in the fuel, and C can be calculated from the energy release from fission reactions (≈210 MeV/fission for ^{239}Pu and ^{241}Pu.) We have calculated this neutron flux level for an **initial** power density of **353 W/cm^3** (for a mass density of the fuel in the active zone of 0.141 gr/cm^3, which corresponds to a pebble fuel element design charged with 2 gr of actinides per pebble and 1.5 cm of active zone, 5 kW per pebble.) The results for the five theoretical spectra are included in Table 3.

Table 3. **Neutron flux level for 353 W/cm^3 power density**

Mean energy	Neutron flux level
E1 = 2.2E-08 MeV	Φ1 = 4.772963E13
E2 = 1E-06 MeV	Φ2 = 1.468360E14
E3 = 1E-04 MeV	Φ3 = 1.418104E15
E4 = 1E-02 MeV	Φ4 = 1.830872E16
E5 = 1E-00 MeV	Φ5 = 2.266231E16

In the case of thermal and low epithermal spectrum, only ^{239}Pu and ^{241}Pu isotopes have significant fission cross-sections. This means that the power can be calculated by the following approximate formula.

$$\textbf{Power (W/cm}^3\textbf{)} = C\cdot(^{239}N\cdot\sigma_f^{239} + {}^{241}N\cdot\sigma_f^{241})\cdot\phi \qquad (2)$$

As energy increases, the fission cross-section decreases sharply for these isotopes, but the rest of them, initially with low fission cross-sections, have a non negligible value. Nevertheless there is still a

big difference in the values of these cross-sections, so the harder the spectrum, the higher the neutron flux level needed to achieve this power (which does not mean a higher neutron concentration, since the neutron flux is the product of density times velocity.)

The problem of thermal and low-epithermal spectra to transmute all the isotopes from the spent fuel is clear. Initially, in a thermal spectrum, 57% of the mass initially charged are ^{239}Pu and ^{241}Pu isotopes, and they are the only isotopes with a non negligible fission cross-section. This means that the initial neutron flux level is going to depend on the fission cross-section of both isotopes, particularly ^{239}Pu (which is 51% of the total actinide mass.) For a thermal spectrum, the fission cross-section of ^{239}Pu is very high, so the neutron flux level is low. The destruction rate of any isotope in the reactor is given by next equation, if captures and radioactive decays are neglected for the moment as source terms.

$$\frac{dN}{dt} = -\sigma_{abs} \cdot N \cdot \Phi \tag{3}$$

If the absorption cross-section of any isotope is much lower than the fission cross-section of ^{239}Pu (which gives de neutron flux level), the destruction rate is very low. This is the case of ^{244}Cm isotope in a thermal spectrum, where we find:

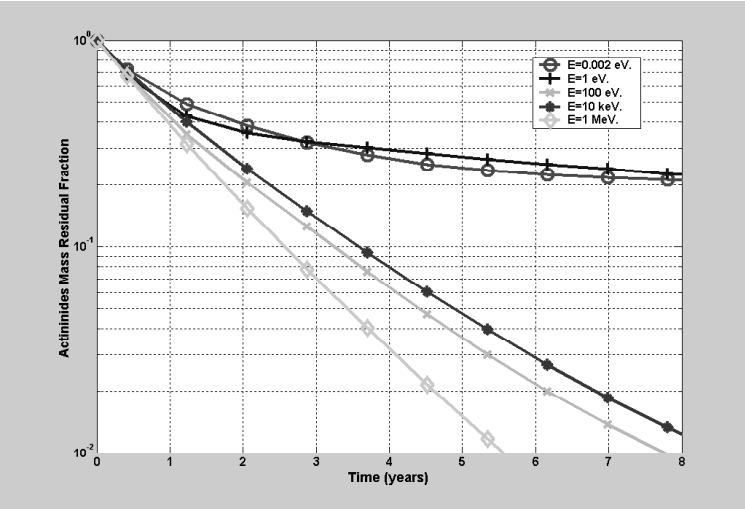
$$\frac{\sigma_{abs}^{Cm244}}{\sigma_{f}^{Pu239}} = 0.015 \tag{4}$$

The elimination rate of some Cm isotopes in the reactor is going to be about two orders of magnitude lower than those of Pu isotopes. This gives us an idea of the difficulty to transmute Cm isotopes in a thermal and low-epithermal spectrum. This is not the case for fast spectrum.

With **MCNP** calculated cross-sections, libraries for **ORIGEN** burn-up code have been prepared. This code carries out the calculation of actinides isotopic evolution inside the reactor. To compare the five different spectra, we have supposed an ***initial*** power density of 353 W/cm^3, obtaining the initial neutron flux level given in Table 3. This ***initial*** neutron flux level is maintained ***constant*** during calculations. The power obtained from the fuel (or the transmutation rates, which are related concepts) are not constant with time during burn-up, and depends on the neutron spectrum. These calculations correspond to a fuel element inside a reactor with a constant spectrum and neutron flux level. Figure 6 represents the elimination rate of all actinides (composition defined in Table 1) with a constant neutron flux level and spectrum.

From Figure 6, it can be seen that it is almost impossible to eliminate Cm isotopes with thermal and low epihermal spectrum, with the neutron flux level that gives the selected initial power density, since absorption cross-section of these actinides are much lower than the fission cross-section of ^{239}Pu. The spectrum that can achieve an optimum and constant burn-up rate is the fast spectrum. 99% of actinides can be eliminated within 5.5 years, with the presumed constant neutron flux level. In thermal and low epithermal spectra, it can be seen that the maximum burn-up achieved correspond to a 80% actinides elimination for the time span of our calculations. Of course, for a burn-up time going to infinity, the final residual mass tends to 0.

Figure 6. **Actinides elimination; mass residual fraction**



There is a direct correspondence between burn-up and actinides mass elimination. Supposing a 240 mean mass number for all the isotopes eliminated, and a fission energy released per reaction of 210 MeV, the burn-up equation related to actinides mass elimination is given by:

burn-up (MWd/kg)= 975.9·(1-RF); where RF is the actinides residual fraction.

Nuclear cycle scenario: no transuranic reprocessing in the transmutation phase

The goal of this paper is to **study the option of a minimum reprocessing strategy,** where the nuclear waste (after LWR discharge) is reprocessed, with transuranic (TRU) separation. After that, TRU are incinerated in a reactor with an optimum spectrum for this purpose. Direct disposal in a Deep Storage Facility of the fuel elements after transmutation is considered as the final step of the fuel cycle. In this scenario, a fuel element design capable of withstanding a very high burn-up is needed. This is the case of pebble-bed reactors fuel elements, which are the so-called Triso coated fuel particles. [16-18] It is composed of an inner region or kernel in which the fuel is embedded, and different graphite layers that can retain the gaseous fission products produced. When using Pu-fueled kernels without uranium, the Triso coated fuel particles can withstand burn-ups up to 700 MWd/kg.

In our study, we will take 700 MWd/kg as the maximum for our once-through strategy. This value correspond to near 75% of mass elimination, that is, a 25% of actinides residual fraction. The isotopic composition of this 25% of actinides is the fundamental variable to determine the final radiotoxicity (effective committed dose) of the spent fuel in the final repository. It is worth remembering the good confinement properties of the coated fuel particles, which can act as a barrier for radioactive isotopes release. TRU are introduced as the only fuel in the coated particles. After 700 MWd/kg, the irradiated coated fuel particles are disposed of in a deep storage facility. In the calculation of residual radiotoxicity, the following items must be taken into account:

1. **Initial radiotoxicity, as a reference value:**
   - natural uranium (reference level).

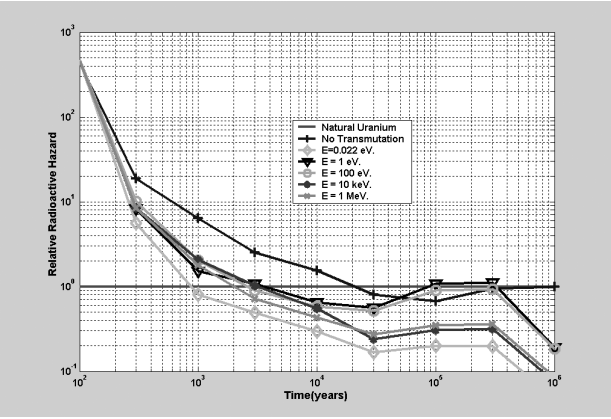
2. **Final radiotoxicity after transmutation:**
   - fission products;

- Pu and MA from reprocessing;
- fission products after transmutation;
- actinides after transmutation.

3. **Final radiotoxicity without transmutation:**
   - fission products;
   - Pu and MA (total).

In Figure 7, residual radiotoxicity after 700 MWd/kg for the different five transmutation spectra are depicted in terms of relative radiotoxicity, using the value of the total radiotoxicity of Natural Uranium as reference level. Radiotoxicity in case of no transmutation is also given. To obtain this burn-up value, the initial fuel has been irradiated with a constant neutron flux level for all five cases, for an **initial** power density of 353 W/cm^3, and the time necessary to achieve that burn-up.

Figure 7. **Relative ingestion Effective Committed Dose (relative to natural uranium level) of the nuclear waste after 700 MWd/kg transmutation of actinides from LWR spent fuel**



From Figure 7, it can be seen that the best results are obtained in the case of a well thermalised spectrum. Principle contributors to the final radiotoxicity after 700 MWd/kg burn-up are ^{238}Pu, ^{240}Pu, ^{241}Am, ^{243}Am. The isotopic evolution and the radioactive dose of these elements can be characterised independently until a **total** burn-up of 700 MWd/kg.

From Figure 3, it has been demonstrated that the principle contributors to the Effective Committed Dose of the spent fuel are Pu isotopes. If we study the independent contribution of Pu isotopes after a total burn-up of 700 MWd/kg of all the spent fuel, we obtain the results in radiotoxicity presented in Figure 8.

From these figures, it can be seen that better results are obtained with the very well thermalised spectrum. The explanation of this behaviour can be explained as follows. It can be seen that the residual waste is mainly ^{242}Pu (72% in mass) with a very thermal spectrum. ^{242}Pu has 3.74E+05 years half-life, which implies a low radiotoxicity level. ^{242}Pu build-up is produced because the capture cross-section of ^{242}Pu is much lower than fission cross-section of ^{239}Pu. The isotopic evolution of Pu isotopes ends at ^{242}Pu, due to the small capture cross-section of the latter. The spectral index relevant to this calculation is given in Table 4 and Figure 9 for the thermal and low epithermal spectra.

Figure 8. **Relative ingestion Effective Committed Dose (relative to natural uranium reference level) due to plutonium isotopes and descendants after total 700 MWd/kg burn-up in the transmutator**

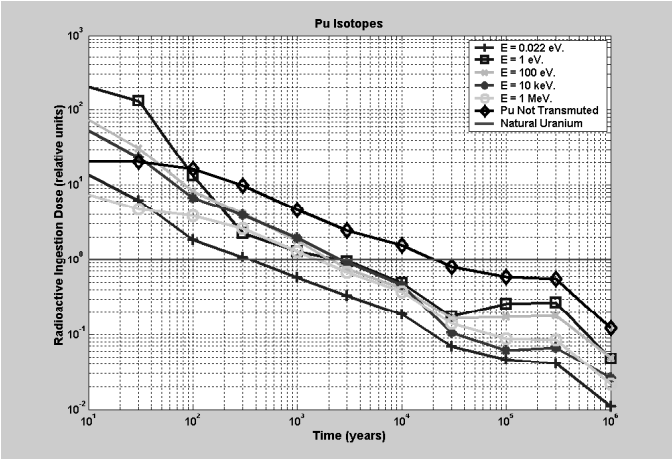
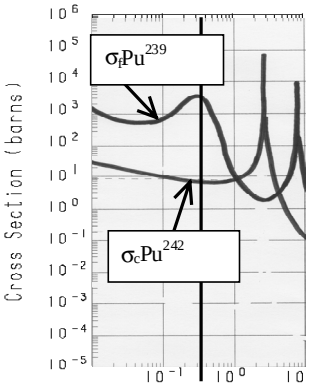


Figure 9. **^{242}Pu capture and ^{239}Pu fission cross-sections**



0.3 eV

Table 4. **Spectral index between ^{242}Pu capture and ^{239}Pu fission cross-sections**

Spectrum	$i = \frac{\sigma_c^{Pu242}}{\sigma_f^{Pu239}}$
E = 0.022 eV	2.57E-02
E = 1 eV	5.38E-01

It can be seen that the difference in the spectral index comes from the different position in energies of the resonance regions of ^{239}Pu and ^{242}Pu. From Figure 13, it can be seen that an optimum value exists for an spectral index, corresponding to 0.3 eV mean energy of neutrons. From the former data, it can be seen that the captures of ^{242}Pu are practically inhibited in the thermal spectrum. In the low epithermal one, captures of ^{242}Pu are enhanced (due to resonance), and then Cm isotopes are produced (^{244}Cm is 87% of total mass of residual TRU after 700 MWd/kg due to the low ^{244}Cm capture cross-section.) The radiotoxicity of ^{244}Cm is very high, due to the short half-life of the isotope ($T_{1/2}$=18.11 years.) It becomes ^{240}Pu by α disintegration, which is one of the higher contributors to final dose. This explains the poor results in the low epithermal spectrum. On the contrary, it is also important to highlight that ^{242}Pu disintegrates by α emission in ^{238}U, which is the initial isotope that starts the energy production cycle (massive presence in Natural Uranium Ore.) Therefore, ^{242}Pu seems to be a natural way to channelise actinides decay into natural material.

Pebble bed reactors application; optimum spectrum to minimise residual radiotoxicity

From the previous analysis, it can be seen that for a once-through transmutation scenario, with a burn-up of 700 MWd/kg of TRU, the optimum spectrum corresponds to the thermal case. Once identified the reason (^{242}Pu accumulation in the final residual mass), calculations must be performed to feature a physical system that could achieve this purpose. Fuel elements that can withstand very high burn-up values are Triso Coated Fuel Particles. [5-7,16-18] It has been identified that the best results for minimising radiotoxicity correspond to a minimum in the spectral index defined in Table 4, defined in terms of the cross-sections of ^{242}Pu and ^{239}Pu. For an energy of 0.3 eV (which corresponds to the energy of the first ^{239}Pu resonance) the spectral index gets a minimum value, so the ^{242}Pu accumulation becomes a maximum which is the main objective for radiotoxicity reduction. To obtain a similar spectrum in a pebble fuel element design, a thermal spectrum is needed. To taylor it, a heterogeneous design has to be modelled. In the case studied in this paper, Triso particles would only be loaded with transuranics in the fuel (e.g. actinides coming from LWR spent fuel.) To achieve an optimum spectrum for once through transmutation applications, it is needed to be as far as possible from the ^{242}Pu capture resonance. This objective requires a good neutron thermalisation. The pebble design suited for this purposes is the heterogeneous one, with fuel in the very inner region of the pebble. In this zone, 50% of Triso Coated Fuel Particles would be mixed with 50% of graphite to compact the Triso particles (it is not possible to use less graphite in the compactation process. [2] In our study, the 6 cm diameter pebble is taken as a constant parameter in the design, so the smaller the active zone, the bigger the moderator zone, and the higher the moderation effect in the spectrum. The final active radius depends on the fuel mass charged per pebble (which depends on the number of Triso coated fuel particles in the active zone.) For a thermal spectrum, a low fuel mass has to be charged. Three different cases have been studied, corresponding to 0.25 gr, 0.5 gr and 1 gr per pebble. It must be noted again that the fuel is composed by TRU discharged from LWR, only. If the mass charged in the fuel element is larger than 1 gr, the spectrum is low epithermal, and many neutrons appear in the resonance energy of ^{242}Pu. As a result of that the final radiotoxicity for a 700 MWd/kg max burn-up becomes high. We have compared radiotoxicity results for three different cases. The spectrum used to calculate average cross-section has been the one obtained in a reactor with characteristics defined in Table 5.

Table 5. **Transmuter main parameters**

Active radius	2.8 m
Active height	5 m
Reflector thickness	1 m
Reflector material	graphite
Number of pebbles	664 222 pebbles
Porosity (gas vol/pebble vol)	x = 0.39
Power per pebble	1 kWth
Total power reactor	664.22 MWth

This conceptual design has a low aspect ratio (height/diameter), which is bad for heat decay cooling in accidental cases, but this design was chosen only for checking the transmutation performance, which is not expected to change significantly if a different geometry is adopted. The active zone of pebbles has been taken as a homogeneous mixture of graphite and fuel, since Triso coated fuel micro-particles diameter is much lower than thermal neutron mean free path in that region. The corresponding active zone for each case is given in Table 6. Once the cross-sections have been calculated for this reactor (**MCNP** calculations), taking an average value for all active zones in the reactor, the isotopic evolution along the transmutation phase can be calculated. To obtain good results

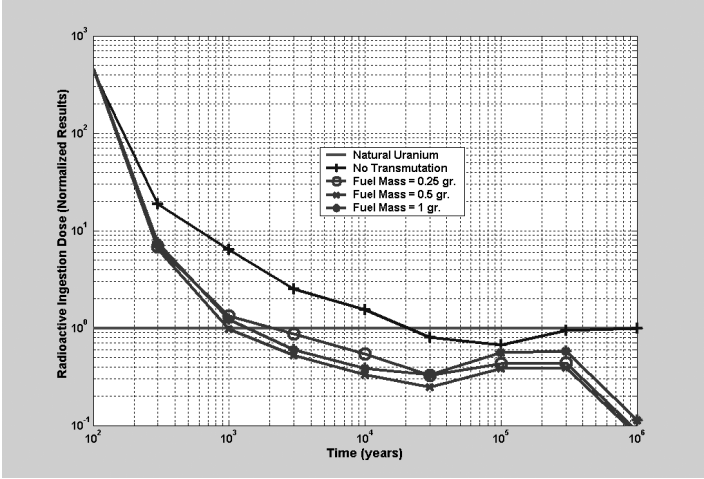
in radiotoxicity after 700 MWd/kg burn-up, it is necessary the spectral index of Table 6 to be as small as possible (to stop captures of Pu^{242} isotope, and not produce Cm isotopes.) This case corresponds to a fuel mass of 0.5 gr (see Table 6.)

Table 6. **Capture and fission cross-sections and spectral index for ^{242}Pu capture and ^{239}Pu fission, neutron flux level for 1 kW per pebble and active fuel radius**

Mass charged per Pebble	$i = \frac{\sigma_c^{Pu242}}{\sigma_f^{Pu239}}$	σ_f Pu^{239}	Neutron flux level 1 kW per pebble	Active fuel radius
Mf = 0.25 gr	1.63E-01	2.33E+02	3.63E+14	0.340 cm
Mf = 0.5 gr	4.61E-02	1.74E+02	2.41E+14	0.428 cm
Mf = 1 gr	1.19E-01	9.37E+01	2.23E+14	0.539 cm

The neutron flux level needed to achieve a certain power per pebble has also been calculated. For normalisation purposes, we have normalised the results to 1 kW per pebble. The results are presented in Table 6. Once cross-sections are obtained for the three cases, and neutron flux levels are calculated for 1 kW per pebble, the evolution of radiotoxicity has been calculated for the three different cases. Results are included in Figure 10, in terms of radiotoxicity relative to Natural Uranium Ingestion Effective Committed Dose.

Figure 10. **Relative ingestion Effective Commited Dose (relative to natural uranium ore) evolution of actinides after 700 MWd/kg burn-up for cases defined in Table 6**



From this figure, it can be seen that the optimum spectrum corresponds to Mf = 0.5 grams per pebble. This corresponds to the case of a minimum spectral index defined in Table 6. The residual radiotoxicity line crosses the reference level at less than 1 000 years. The spectrum corresponding to the case of Mf = 1 gr is the second one in crossing the line, which agrees with the spectral index results. The third one crosses the Natural Uranium reference line at 2 000 years. As a conclusion, from the point of view of residual radiotoxicity, the optimum spectrum for a 700 MWd/kg burn-up limit corresponds to 0.5 grams of fuel.

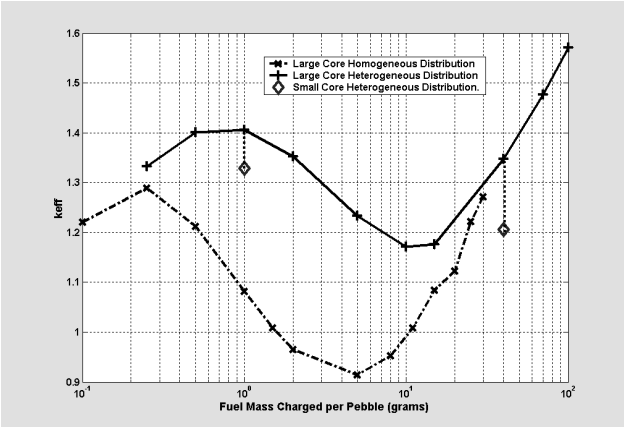
Accelerator-driven system to complete the transmutation phase

Once the thermal spectrum has been identified as the optimum one for transmutation applications in a once-through transmutation scenario, with a limit of 700 MWd/kg burn-up, it must be considered the type of reactor to carry out the transmutation phase. If a fast spectrum were adopted for transmutation applications, the Doppler effect would be positive for clean initial composition waste. This means that the transmutation in a sub-critical reactor (Accelerator-driven system or ADS) would be necessary (with a k_{eff} well bellow one), since a critical reactor with a positive Doppler effect can not be considered for licensing. [19-21] In ADS, one of the critical points is the accelerator itself, because of the cost, the reliability, and the uncertainty of achieving big currents (in the order of mA) of high energy protons (GeV) needed to drive the reactor by neutrons produced in spallation reactions. The scenario changes if the spectrum for transmutation is thermal (which is the optimum case for a once-through strategy.) In this case, the reactor presents a negative Doppler effect, due mainly to ^{240}Pu capture resonance. This enables one to burn the fuel in a critical reactor at least partially, until it is not possible to keep the reactor with $k_{eff}=1$. With a suitable pebble recycling strategy, high burn-up values could be achieved, but it seems really difficult to reach 700 MWd/kg final burn-up for all the fuel to transmute. Hence, the once-through transmutation phase will be done in two stages, the first one in a critical reactor, the second one in an ADS. [8,9] Some criticallity calculations have been computed with initial fuel, to find out what is the level of initial reactivity we can depart from. Conclusions from this study at reactor level can be summarised as follows (numerical results are shown in Figure 19.):

1. In the heterogeneous case (minimum fuel volume), high values of k_{eff} are achieved, with a maximum for 1 gram of TRU per pebble. In the epithermal region, k_{eff} decreases due to the high resonance of ^{240}Pu, and the smaller spectral index (σ_f / σ_c) for fissile isotopes. k-eff increases with harder spectrum, and higher mass content. In the heterogeneous mode, it is possible to drive partially the reactor as a critical one, and then use an ADS for final burn-up until 700 MWd/kg of TRU.
2. In the homogeneous distribution case (constant fuel volume corresponding to 2.5 cm), the values of k_{eff} are smaller, since the spectrum is always low epithermal. Captures of ^{240}Pu are higher, and k_{eff} decreases. However, the behaviour of k_{eff} with fuel mass has the same shape as in the heterogeneous case.
3. In the case of a smaller core, with a bigger aspect ratio, reactor safety features in the accidental case (decay heat removal) are improved, but k_{eff} decreases. An optimisation work has to be carried out to optimise both characteristics.
4. In the case of reprocessing after only 4 years cooling, k_{eff} increases due to the increase in ^{241}Pu fissile isotope. If a critical reactor is intended to be used in the first part of the transmutation phase, the faster the reprocessing, the higher the initial k_{eff} of the reactor.

The transmutation reactor could be driven as critical until k_{eff} becomes one. Even with a suitable pebble recycling strategy, the maximum burn-up will not likely achieve 700 MWd/kg (as it is necessary for this once-trough strategy.) An initial calculation of the maximum burn-up achieved in the critical reactor has been carried out. The burn-up is calculated homogeneously in all the reactor. Assuming homogeneous burn-up, a value of 351 MWd/kg for reaching k_{eff} one is found. This value could be improved in a detailed design, but it indicates that a second stage with transmutation in ADS is needed. Questions about safety considerations (Doppler effect negative during burn-up, etc) will be objectives of future studies. [22]

Figure 11. **k_{eff} dependence of fuel mass charged per pebble; initial TRU fuel composition; heterogeneous and homogeneous pebble design**



Summary

A potential scenario for nuclear waste transmutation has been defined and studied on the basis of avoiding further reprocessing during the transmutation phase. In this scenario, nuclear fuel unloaded from LWR would be reprocessed after a cooling period after discharge, the transuranics being converted into the fuel of a pebble fuel element, without addition of any other nuclear fuel. This type of fuel element was selected because it could withstand very long burn-up, of the order of 700 MWd/Kg (of TRU.) Those pebbles would not be reprocessed ever. Within this framework, residual radiotoxicity can be minimised by using a thermal spectrum, unlike in the general case of multiple TRU reprocessing during the transmutation phase, where fast spectra are needed to minimise the long-term radiotoxicity. Results of the once-through scenario studied in this paper can be explained in term of ^{242}Pu accumulation (which is enhanced in thermal spectra) not leading to higher-A actinide build-up (specially ^{244}Cm isotope.) In a study case for a given type of reactor, it was found that the residual radiotoxicity with the thermal spectrum would achieve the reference level of the natural uranium ore in less than 1 000 years. It is worth pointing out that a significant improvement would be achieved in that case, because the most offending nuclei (^{239}Pu, ^{240}Pu and ^{241}Pu) would be eliminated in a very high fraction, and the time span for the radiotoxicity to go down to the natural level of reference is reduced by a factor higher than 20 (from more than 20 000 years to 1 000 years.) Because of the neutronic features of the fuel and the reactor, a first stage of the transmutation phase could be done in dedicated critical reactors, with a second phase where ADS would be needed.

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