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A COMPARISON OF CURIUM, NEPTUNIUM AND AMERICIUM TRANSMUTATION FEASIBILITY

ABSTRACT

Minor actinides transmutation is the process of decreasing the long term radiotoxicity of the nuclear spent fuel by submitting it to a neutron flux so as to achieve fission of the heavy nuclides concerned. In the case of a closed fuel cycle, minor actinides are the main contributors to the spent fuel radiotoxicity after a few centuries. The isotopic vector of the minor actinides feed to be transmuted depends heavily on the fuel cycle considered: PWRs with UOX fuels will mainly lead to neptunium and americium production while MOX fueled reactors will produce mainly americium and curium. Americium is the main element currently considered for transmutation due to its relatively short half-life and important production. On the other hand, neptunium is seen as a secondary candidate for transmutation due to its very long half life and low activity while Curium has transmutation is generally ruled out due to the important activity of curium isotopes. Two modes of transmutation in fast reactors are generally opposed, namely the homogeneous approach in which minor actinides are directly mixed with the fuel while in the heterogeneous approach, the minor actinides are loaded in dedicated targets. It is shown in this paper that the impacts on the fuel cycle of heterogeneous americium transmutation are similar to the one of homogeneous curium transmutation. It is further shown that given the quantities of curium in the fuel cycle, only a limited number of reactors would be required to effectively transmute the curium production of fast reactors with americium bearing blankets. Curium transmutation thus appears a feasible option in a completely closed fuel cycle without significantly higher fuel cycle impacts than with only americium transmutation. It is finally verified that neptunium transmutation can be achieved regardless of the approach considered.

INTRODUCTION

In the case of a closed fuel cycle where plutonium is multi-recycled in fast reactors, minor actinides, namely americium, neptunium and curium are the main contributors to the long-term radiotoxicity of the spent fuel after a few centuries. A complete removal of those nuclides from the waste could lead to reduction of the radiotoxicity of the long-lived waste by at least one order of magnitude [1]. Additionally, as they contribute to most of the decay heat of the spent fuel by alpha-decay, their removal from the waste would limit the total heat load of the waste packages, which would increase their packing ratio in a final deep geological repository and thus limit the size of such repository [2].

Minor actinides production strongly depends on the irradiation history of a spent fuel and on the type of reactor in which it was used. An UOX LWR will thus mainly produce neptunium from captures on ^{235}U and ^{241}Am from decay of ^{241}Pu while MOX fueled reactors will mainly lead to the production of americium and curium isotopes from captures and decay of plutonium isotopes. The production level will be higher in a thermal reactor due to the non favorable capture to ratio in this spectrum compared to a fast reactor [3].

After reprocessing and recovery from the spent fuel, minor actinides can be turned into fission products by submitting them to a neutron flux. Two main approaches have been discussed to load minor actinides into a reactor core [4]:

- The homogeneous approach in which minor actinides are directly mixed with the reactor fuel up to a few percent. As they are located in the center of the core, the minor actinides are under a high neutron flux and the transmutation process is efficient. However, their loading has a hardening impact on the neutron spectrum in the core which negatively modifies the core behavior during various incidental transients. Additionally, it "pollutes" the entirety of the fuel cycle with minor actinides which are strong alpha and neutron emitters.
- The heterogeneous approach, in which minor actinides are loaded in dedicated targets, generally located at the core periphery. As they are located in a low-flux zone, they do not modify the core behavior during transient. However, due to this lower flux level, the transmutation process is less efficient, which is generally compensated by increasing the amount of minor actinides loaded in the blankets and their residence time. This in turn complicates the handling of the fresh and irradiated assemblies due to their high content in minor actinides. However, this approach completely separates the blankets fuel cycle from the standard fuel assemblies' fuel cycle, which offers a greater flexibility for the implementation of the transmutation process.

Regardless of the approach chosen, it is necessary to multi-recycle minor actinides bearing fuels as the transmutation rate, which is the fraction of minor actinides loaded effectively transmuted during irradiation of a fuel assembly is limited around 30 to 50 % depending on the approach chosen, which is insufficient to remove all the minor actinides from the waste. Consequently, the impacts of minor actinides transmutation on the fuel cycle will also be determinant in the implementation of a given transmutation strategy [5].

After a short description of the transmutation process and specificities of each minor actinide, their behavior in each approach will be analyzed and compared with regards to their transmutation performances, back and front end fuel cycle impacts and core behavior impacts in the case of homogeneous minor actinides transmutation.

NEPTUNIUM TRANSMUTATION

The element neptunium has one isotope with a half-life greater than 3 days, which is $^{237}_{93}Np$. With a half life of 2.144 million years, it is a long-lived radionuclide which is a by-product of nuclear reactors operation. ^{237}Np (or neptunium as it will be referred to henceforth) has two main production routes:

- Successive neutron captures on ^{235}U, which yields ^{236}U and ^{237}U, which finally decays with a half-life of 6.75 days to ^{237}Np. This reaction is preponderant in thermal reactors.
- (n,2n) reaction on ^{238}U which is a threshold reaction around 6 MeV. This reaction is preponderant in fast reactors, where it makes up to 90 % of the neptunium production.

$$^{235}_{92}U + n \rightarrow {}^{236}_{92}U + n \rightarrow {}^{237}_{92}U \xrightarrow{\beta} {}^{237}_{93}Np$$
$$^{238}_{92}U + n \rightarrow {}^{237}_{93}Np + 2n$$

Homogeneous incorporation of neptunium in the fuel has several impacts in fast reactors, which are:

- A decrease in the absolute value of the coolant void worth, due to an earlier increase in the fission cross section of neptunium compared to uranium with regards to the neutron energy. Consequently, during a coolant voiding accident, the hardening of the spectrum leads to a reactivity increase. [6]
- A decrease in the absolute value of the Doppler feedback, which is explained by an increased capture in the 30 keV range by neptunium nuclei. This leads to a reduction of the flux in the lower energy region below 1 keV which contributes the most to the Doppler Effect. [6]
- An increase in the breeding gain due to higher neptunium capture cross section level compared to uranium.
- A slight decrease in the delayed neutron fractions (around to 3%).

Neptunium-bearing pins have already been irradiated, in Russia in the DOVITA experiment [7] and in France during the SUPERFACT experiment [8]. The pins containing 2 vol % of Neptunium in SUPERFACT and 5 vol % in DOVITA did not exhibit any significant differences in terms of behavior under irradiation compared to the regular MOX pins. From these results, we can consider that neptunium volume fractions up to 5 % in the fuel can be considered without extensive modifications of the fuel design.

Furthermore, sample pins containing 45 % of Neptunium irradiated in SUPERFACT did not show extensive modification of their behavior compared to standard pins. Although there are not enough data to draw final conclusions on the behavior of pins containing a high fraction of neptunium, we assumed here that manufacturing and irradiation of such pins is possible.

Under irradiation, neptunium either undergoes fission or yields ^{238}Pu by neutron capture following the reaction:

$$^{237}_{93}Np + n \rightarrow \ ^{238}_{93}Np \overset{\beta}{\rightarrow} \ ^{238}_{94}Pu$$

^{238}Pu is a strong alpha-emitter with a half-life of 87.75 years and an associated specific decay heat of 567 W/kg. This high level of thermal activity will increase the complexity of manipulation and reprocessing of the irradiated neptunium-containing fuel. However, ^{238}Pu can be directly mixed with the existing plutonium feed and be reused as fuel as it is a net neutron provider in a fast spectrum.

Separation of the neptunium from the spent fuel has already been demonstrated and can be implemented industrially with a good expected separation factor [9]. As neptunium exhibits low specific activity level, it can be readily manipulated and manufacturing of neptunium-bearing pellets does not require additional precautions compared to MOX pellets.

The critical mass of a bare sphere of neptunium is estimated to be around sixty kilograms [10]. As such, separated neptunium from nuclear spent fuel can lead to a proliferation issue, which would be lessened by the transmutation of neptunium. However, leaving some of the ^{238}Pu produced with the separated neptunium would greatly diminish the proliferation risk as it would prevent its use for military purposes in a way similar to plutonium denaturation as discussed in [11]

Neptunium impact on the thermal load of the waste package, which is the main parameter influencing the repository size is nearly inexistent, as it has a very low specific activity. However, the amount of ^{238}Pu lost during reprocessing may increase the heat load of the waste packages. In terms of radiotoxicity, neptunium contribution is negligible up to several millions years, as it can be seen on Figure 1. This figure only shows the heavy nuclides radiotoxicity. The fission products radiotoxicity being negligible compared to the minor actinides contribution after a few centuries, they were not taken into account for this application.

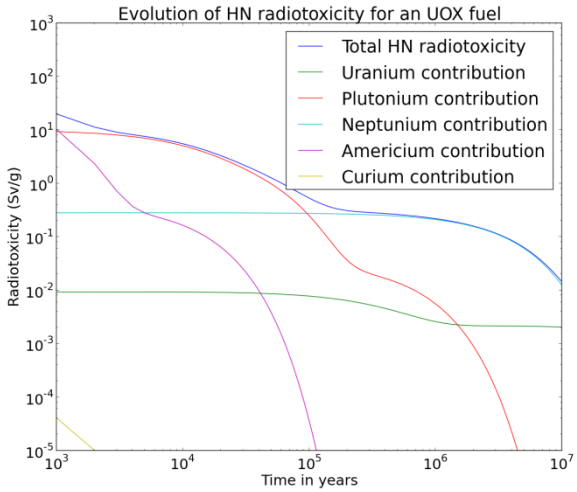

Figure 1 : Radiotoxicity by ingestion from ICRP 119 [12] for the MA vector of an irradiated UOX fuel at 33GWd/t from [12]

AMERICIUM TRANSMUTATION

Americium has three main isotopes that can be found in the fuel cycle as shown in Table 1. ^{241}Am comes from beta decay of ^{241}Pu with half-life of 14 years.

$$^{241}_{94}Pu \xrightarrow{\beta,14y} {}^{241}_{95}Am + {}^{0}_{-1}e + \bar{\upsilon}$$

Its production is strongly dependent on the isotopic vector of the plutonium used as fuel and on the reactor spectrum. Plutonium multi-recycled in SFR will have a content in ^{241}Pu as low as 3 %, which in turns leads to a low production of ^{241}Am, while current plutonium is closer to 8 % [13]. It makes up most of the minor actinides production in fast reactors, with generally more than 60 % of the mass. The amount of ^{241}Am in the fuel cycle is strongly dependent on the cooling and reprocessing time of the spent fuel. The longer this cooling time, the higher the fraction of ^{241}Pu that has decayed and the higher the production of ^{241}Am.

Table 1 : Isotopes of americium

Isotopes	Half Life (y)	Daughter-nuclei (by α decay)	Production way
241	432.2	Np 237	^{241}Pu decay
242m	141	Pu 238	Capture on ^{241}Am
243	7370	Pu 239	Capture on ^{242}Pu

Due to its alpha decay, it has a small thermal load of 0.11 W/g. This decay is accompanied with a weak gamma ray generally around 60 keV emitted by the ^{239}Np produced. Consequently, americium-containing fuels must be handled in gloves-boxes or even hot-cells, depending on the quantity considered.

^{242}Am and its stable isomer ^{242m}Am are produced by capture on ^{241}Am.

$$^{241}_{95}Am + ^{1}_{0}n \rightarrow ^{242(m)}_{95}Am$$

The actual proportion of each nuclei produced depends on the incoming neutron energy as it can be seen on Figure 2. ^{242}Am decays with a 16h half-life to ^{242}Cm in 82.7 % of the case and directly to ^{242}Pu by electronic capture in the remaining 17.3 %. ^{242m}Am is unusually a metastable state more stable than its related ground state and it nearly always transition to ^{242}Am by gamma emission. ^{242m}Am has a very high fission cross-section which means it is found in relatively low quantity in the minor actinides production of a reactor, between 1 and 2 %.

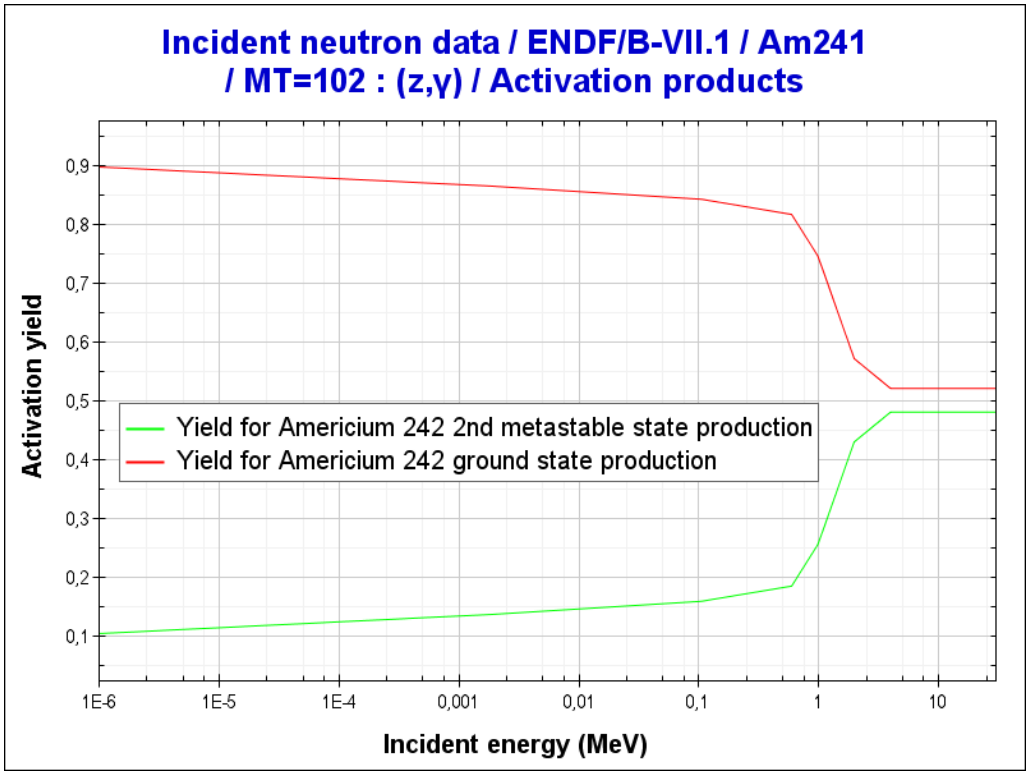

Figure 2 : Capture yields on ^{241}Am. Red : Am242, Green : ^{242m}Am

Finally, ^{243}Am is the longest lived isotope of Americium and is produced by neutron capture and subsequent decay on ^{242}Pu as shown in the equation below. Consequently and similarly to the ^{241}Am case, its production can vary by up to a factor two depending on the plutonium isotopic vector of the fuel. ^{243}Am yields ^{239}Np by alpha decay, which is a strong gamma emitter and thus contributes to the radiotoxicity of americium.

$$^{242}_{94}Pu + ^{1}_{0}n \rightarrow ^{243}_{94}Pu \xrightarrow{\beta,5h} ^{243}_{95}Am + ^{0}_{-1}e + \bar{\upsilon}$$

Americium is the main contributor both to minor actinide fuel cycle inventory and medium-term (between 100 and 10 000 years approximately) radiotoxicity in the case of a closed fuel cycle as visible in Figure 3. Consequently, Americium transmutation is a good option to reduce both parameters. However, it is less straightforward than neptunium transmutation as it leads to Curium by neutron capture, which is also a minor actinide and is less convenient to handle. As americium is generally consider as the best candidate for transmutation, it will be used as a comparison point here.

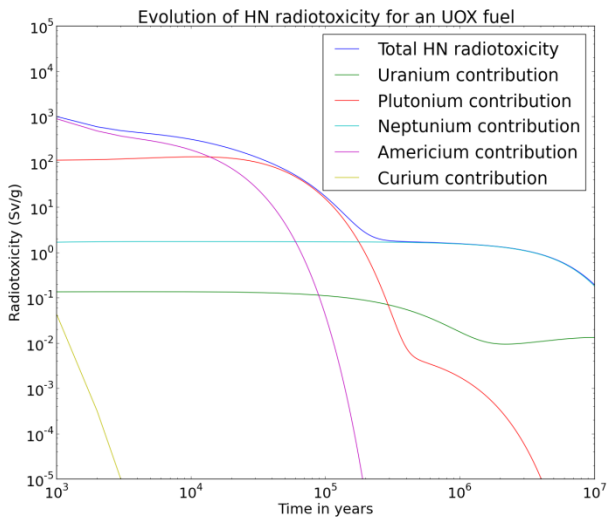

Figure 3 : Radiotoxicity by ingestion from ICRP 119 [10] for the MA vector of an irradiated MOX fuel at 48 GWd/t

CURIUM TRANSMUTATION

Curium is created by successive capture on plutonium and americium isotopes as shown in the equations below. The main isotopes that can be found in the spent fuel are given in Table 2.

$$^{242}_{95}Am \xrightarrow{\beta} ^{242}_{95}Cm + ^{1}_{0}n \rightarrow ^{243}_{95}Cm$$

$$^{243}_{95}Am + ^{1}_{0}n \rightarrow ^{244}_{95}Cm + ^{1}_{0}n \rightarrow ^{245}_{95}Cm + ^{1}_{0}n \rightarrow ^{246}_{95}Cm + \cdots$$

Table 2 : Isotopes of curium

Isotopes	Half Life (y)	Daughter-nuclei (by α decay)	Production way
242	0.45	Pu238	^{242}Am decay
243	29.1	Pu239	Capture on ^{242}Cm
244	18.1	Pu 240	Capture on Am 243
245	8.5e3	Pu241	Capture on ^{244}Cm
246	4.8e3	Pu242	Capture on ^{245}Cm
247	1.6e7	Am243	Capture on ^{246}Cm
248	3.5e6	Pu240	Capture on ^{247}Cm

^{244}Cm is the main curium isotope found in the spent fuel, as it is produced from ^{243}Am which is readily available for neutron capture and as its absorption cross-section is low leading to its accumulation in the fuel. Similarly to ^{242}Cm, it is a strong thermal and neutron emitter with a specific heat load of 2.84 W/g. With its longer half-life, it is going to be more of an issue with regards to the storage in the final repository. It also has a high spontaneous fission probability, leading to a high intrinsic neutron source.

^{245}Cm is also produced in kilogram quantities as it comes from ^{244}Cm which is present in significant quantities in the fuel. It shares a significant neutron emission with its parent isotope ^{244}Cm. However, heavier isotopes of Curium are not found in significant quantities in general (up to a few grams each in fast reactors) as they require numerous successive captures to be produced. Nevertheless, they have a non negligible contribution on the neutron source of the spent fuel as they usually have a high spontaneous fission rate.

Curium production is less than 10 % of the total minor actinides production but it drives the short-term radiotoxicity along with fission products (up to a few hundred years) and is the main contributor to decay heat and neutron source. Curium has very limited applications, the main one being the construction of X-ray spectrometer for space probes such as Curiosity.

In terms of radiotoxicity, its short term contribution is relatively important but its long term contribution is close to zero as most of the isotopes produced during irradiation are short lived. This is illustrated in Figure 3, where the radiotoxicity of the minor actinides vector from a MOX fuel is plotted.

The main rationale behind curium transmutation is the decrease of the final waste package heat load. Indeed, curium is a very strong contributor to final decay heat of the waste. Higher spacing between the waste packages is necessary to maintain adequate temperature in the deep geological repository if curium is loaded into the waste package. The current approach is to store the packages containing curium for 120 years in order to await its natural and then send them underground. A reduction in the curium amount in the waste would allow a reduction of this storage duration, and thus limit the inventory of waste stored on surface. It would also limit the final volume to be excavated underground and the total size of the storage facility as discussed in [2]. Curium transmutation is generally considered as un-practical due to its very high impacts on the neutron source.

THE HETEROGENEOUS APPROACH

In this part, the transmutation performances and fuel cycle impact of a « standard » blanket will be compared for each of the three minor actinides. A 4100 EFPD irradiation in an homogeneous core 3600 MWth SFR V2B as designed by CEA, EDF and AREVA will be considered here [14]. A fuel volume fraction of 38.6 % will be considered with 20 at% of minor actinide loaded in the blanket assemblies. The minor actinides isotopic vector considered in this study are shown in Table 3. The curium vector corresponds to the composition of the curium expected to be available around 2035 in the industrial scenarios considered in [13]

Element	^{237}Np	^{241}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm	^{245}Cm	^{246}Cm

Mass Fraction (%)	100	0	0	0	0	0	0	0
Mass Fraction (%)	0	75	25	0	0	0	0	0
Mass Fraction (%)	0	0	0	0,30	1,07	78,23	19,18	1,22

Table 3 : Composition of the minor actinides feed used in this study

BACK END

The first point to study is the characterization of the fresh blanket. Their decay heat level and neutron source level are summarized in Table 4. The main conclusion to draw here is that neptunium is not an issue from the back end point of view, while a fresh curium-loaded assembly is nearly thirty times hotter than americium assembly, while its neutron source is equivalent to the one of a freshly irradiated americium target.

	Specific power (W/kg)	Total power per assembly (kW)
Curium	0,52	74,08
Americium	0,02	2,47
Neptunium	0,00	0,00

	Specific neutron source (n/s/g)	Total neutron source per assembly (1e10 n/s)
Curium	1,80E+06	25
Americium	0	0
Neptunium	0	0

Table 4 : Decay heat and neutron source of the fresh blanket assemblies for neptunium, americium and curium loading

TRANSMUTATION PERFORMANCES

The second point of interest is the study of the transmutation performances through the specific consumptions and the mass balances in the blankets. The results are shown in Table 5. In terms of consumption, americium and neptunium behave similarly, with the only difference that neptunium transmutation does not produce americium or curium but only plutonium. On the other hand, Curium transmutation appears more efficient. This is due to the fact that during the 11 years irradiation, a significant part of the ^{244}Cm will decay considering its 18.8 half-life, thus adding to the total consumption of curium. Interestingly enough, a small production of americium and neptunium can be noted. Neptunium production is due to (n,2n) reactions on ^{238}U nuclei, the fast neutrons necessary for this reaction to take place being supplied by fissions of ^{245}Cm, while the americium is produced by capture on ^{240}Pu coming from the decay of ^{244}Cm.

Specific consumption in kg/TWeh | Loaded minor actinide	Np	Am	Cm
Pu	8,55	7,56	9,64
Np	-6,7	0,2	0,04

Am	0	-6,61	0,13
Cm	0	0,99	-10,52

Table 5 : Specific consumption in the blankets for neptunium, americium and curium loading

In terms of mass balance, similar conclusions can be reached as it is shown in Table 6. In all cases, a strong plutonium production can be observed. The production has two sources, namely breeding on ^{238}U and the actual transmutation process. Its isotopic composition is of interest regarding both the proliferation resistance of the blankets and their cooling behavior.

		Np	Am	Cm
Initial composition (kg)	Am	0	2375	0
	Np	2370	0	0
	Cm	0	0	2378
	Pu	0	0	0
Final composition after 5 years (kg)	Am	0	1433	19
	Np	1413	29	5
	Cm	0	141	878
	Pu	1220	1079	1376

Table 6 : Mass balance in the blankets for neptunium, americium and curium loading

The isotopic composition of the final plutonium is shown in Table 7. Significant differences can be observed in this table. Breeding from ^{238}U represents mostly fifty percent of the produced plutonium, in the form of ^{239}Pu and a small part of ^{240}Pu, with the remaining part being strongly different between the cases. For neptunium, it is mostly entirely composed of ^{238}Pu coming from captures on ^{237}Np. ^{238}Pu is also produced in the americium case through captures on ^{241}Am and the following decay of the produced ^{242}Cm. A small amount of ^{242}Pu is produced, mainly through decay of ^{242g}Am. Finally, for the curium case, an important production of ^{240}Pu can be observed due to the decay of ^{244}Cm. These differences will have an impact on the front end of the blanket fuel cycle, as it will be discussed in the next paragraph.

Plutonium isotopic vector (%)	Np	Am	Cm
^{238}Pu	50	33	0
^{239}Pu	46	49	55
^{240}Pu	4	9	42
^{241}Pu	0	0	3
^{242}Pu	0	8	0

Table 7 : Plutonium isotopic vector in the blankets for neptunium, americium and curium loading

Finally, a last point to be discussed here is the gas production in the blankets during irradiation, which plays an important role in the design of the blanket assemblies [15]. As it can be seen in Table 8, the gas production in the neptunium case is relatively limited compared to the Am or Cm cases. For neptunium, gas production comes from fission of ^{238}Pu or ^{239}Pu and to a lower extent from alpha-decay of ^{238}Pu. On the other hand, for the americium case, most of the gas production comes from the alpha-decay of ^{242}Cm. Finally, for the curium case, the gas production is evenly distributed between fission gases coming mainly from ^{245}Cm fissions and helium coming from ^{244}Cm and ^{242}Cm decay. Overall, the design margin for the curium case is more limited due to the increased gas production. For the neptunium case, the gas production is not an issue in terms of design.

Gas production	Np	Am	Cm
Fission gases (cm^3/g)	0,99	0,84	3,35
Helium (cm^3/g)	0,26	4,19	3,83
Total (cm^3/g)	1,25	5,02	7,18

Table 8 : Gas production in the blankets for neptunium, americium and curium loading

FRONT END

The evolution of the decay heat of the assembly during cooling is shown in Figure 4. Neptunium exhibits a lower level decay heat as it comes mostly from ^{238}Pu which has a low specific decay heat. However, consequently, the target assembly can be handled without any limitations (if we consider a 40 kW limit for short-term handling). Similarly, sodium washing is not limited if a 7.5 kW limit is considered with a few weeks necessary to reach this limit. However, if this limit is decreased to 2.5 kW, the cooling time jumps to more than 60 years due to the relatively long half life of ^{238}Pu (87 years).

The americium case exhibits an important short term decay heat followed by a sharp drop due to ^{242}Cm production during irradiation. Long term decay heat is dominated by ^{244}Cm, ^{238}Pu and ^{241}Am and the cooling time to 7.5 kW is close to 10 years. The cooling time to 2.5 kW is similar to the one of the neptunium due to the contribution of the long-lived ^{238}Pu and remaining ^{241}Am

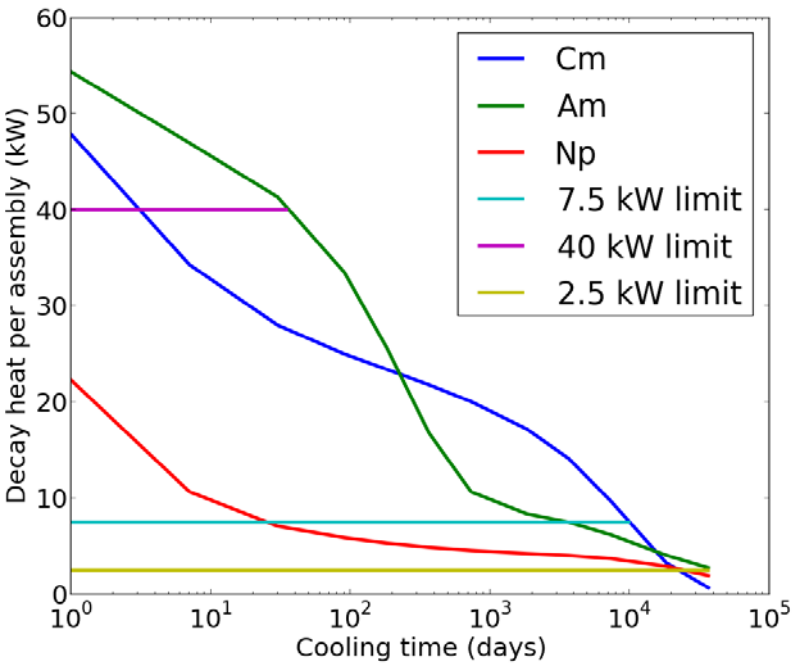

Figure 4 : Evolution of the target assembly decay heat for neptunium, americium and curium loading

Finally, the curium case exhibits a stronger short term decay heat mainly due to ^{244}Cm, which decreases with the decay of this nucleus. Due to important quantities still present in the blankets, the cooling time to 7.5 kW or 2.5 kW is higher than for the americium or neptunium case.

Regarding neutron source, it can be observed in Figure 5 that neptunium loaded targets do not exhibit any neutron source as they do not contain neutron emitting nuclei. On the other and, since ^{244}Cm and heavier curium isotopes are the main responsible for neutron source in the blankets, it can be verified that the curium case yields the highest neutron source. Its neutron source after five years of cooling is nearly fifty times higher than the one of a standard MOX assembly, making its handling significantly harder.

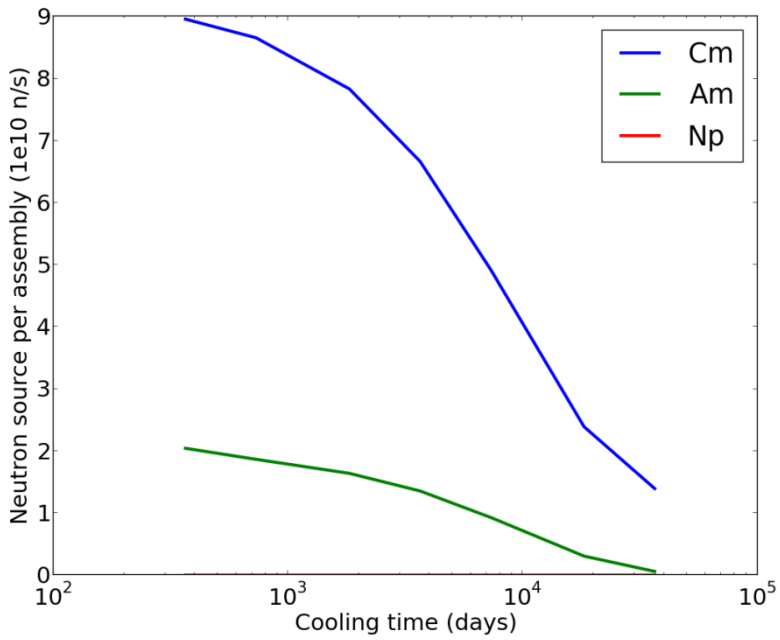

Figure 5 : Evolution of the target assembly neutron for neptunium, americium and curium loading

Regarding the heterogeneous transmutation of neptunium and curium compared to americium, it can be concluded that neptunium is a suitable candidate, even better than americium as the irradiated targets are less active and do not emit neutrons, while curium heterogeneous transmutation cannot be considered realistic due to the high heat load for fresh targets and the very high neutron source of the irradiated targets. Decay heat of the irradiated targets is similar to the americium case, making it a non-limiting factor.

THE HOMOGENEOUS APPROACH

For the homogeneous approach, a reference core similar to the one available in [15] was used. This core is an innovative heterogeneous design with axial heterogeneities such as an upper sodium plenum or a inner fertile blanket. The residence time of the fuel was modified to reach a mean burn up of 200 GWd/t. Various geometrical parameters of the core design are given in Table 9. 5 at% of neptunium, americium and curium were loaded in the cores and their

respective performances were compared in terms of minor actinides consumption, impacts on the core transient behavior and fuel cycle impacts.

Core description	Parameter	Unit
Power	3600	MWth
Inner core height	80	cm
Inner fertile layer	20	cm
Outer core height	90	cm
Core radius	320	cm
Assembly fuel volume fraction	40%	%
Assembly sodium volume fraction	30	%
Number of batch	7	-
Cycle length	435	Jepp

Table 9 : Description of the core used for this comparison

TRANSMUTATION PERFORMANCES

As previously, the transmutation performances will be analyzed through the specific consumption (Table 10) and the mass balance in the core (Table 11). It can be seen that neptunium consumption is the highest in the homogeneous configuration, while curium and americium consumption are quite similar. The difference between the two can be explained by the very limited neptunium production in the core during irradiation compared to americium or curium. Curium production being more limited than the production of americium during irradiation, curium consumption is consequently higher.

Specific consumption (kg\Tweh) | Actinide loaded	Np	Am	Cm
Np	-14,7	0,3	0,2
Am	3,8	-11,0	3,3
Cm	0,7	2,7	-13,3

Table 10 : Specific consumptions in the core for neptunium, americium and curium loading

Regarding the mass balance, the cases of neptunium and americium are quite similar regarding the plutonium inventory. On the other hand, curium loading leads to a diminution by around 20 % of the initial plutonium content in the core as ^{245}Cm is a very good fissile nucleus. ^{244}Cm also contributes significantly to the core neutron balance by breeding ^{245}Cm during irradiation. Concerning the minor actinides, the conclusions are similar than for the specific consumption.

		Np	Am	Cm
Initial composition (kg)	Np	2438	0	0
	Am	101	2545	84
	Cm	0	0	2446
	Pu	12835	12927	10640

Final composition after 5 years (kg)	Np	860	33	26
	Am	398	1252	336
	Cm	103	382	1149
	Pu	10475	10288	9033

Table 11 : Mass balance in the core for neptunium, americium and curium loading

IMPACTS ON THE CORE TRANSIENT BEHAVIOR

The core calculations will be carried out using the ERANOS deterministic code for fast reactor [16] with the JEFF 3.1 nuclear data library [17]. A 2-D RZ description of the core will be considered with calculations carried out using the diffusion approximations. Sodium void worth will be calculated by voiding the entirety of the fissile and fertile layer along with the upper gas expansion plenum and sodium plenum.

Regarding the transients calculations, three transients will be evaluated:

- An Unprotected Loss Of Flow (ULOF), which corresponds to a stop of the primary pumps without insertion of the control rods.
- An Unprotected Loss of Heat Sink (ULOHS), which corresponds to a stop of the secondary pump while the primary pumps are still running, effectively removing the core heat sink without insertion of the control rods.
- An Unprotected Transient OverPower (UTOP), which corresponds to reactivity insertion in the core. We will consider here a slow insertion of reactivity due to the malfunction of a control rod drive mechanism and the consequent extraction of a control rod without insertion of the others.

An ULOF corresponding to an exponential decrease of the primary circuit flow rate up to an asymptotic value of 10 % of the nominal flow with a half-time of 24 s was considered, along with an ULOHS corresponding to a linear total cancellation of secondary flow in 40 s. For the UTOP, a 150 pcm insertion in 250 s was considered. The maximal sodium or fuel temperature during the transients was calculated and is recorded in Table 12.

These transients have been chosen as they are considered as conservative, in a sense that they represent hypothetical highly damaging situations and are thus a good measure of the core behavior during any similar transients. The calculations were carried out using the MAT4DYN dynamic code, which is a mono channel code with point kinetics developed at CEA at the beginnings of the 2000s [18]. A constant exchange coefficient was taken for the gap conductance at 5000 $W/m^2/°C$.

The description of the power, Doppler and sodium thermal expansion axial profiles was simplified to calculate the core transient behavior. An axial power profile from the inner 60 cm of the core was extracted and the average power per region (inner fuel, lower axial blanket, inner axial blanket) was computed. The power profile was then flattened over the assembly height so as to keep the same total power but averaged per medium. However, in the mesh corresponding to the higher fuel temperature (usually slightly above the top of the inner axial blanket), the power was adapted so that the ratio of the maximal power to the average of the fissile part was conserved. This was done to keep the information about the fuel maximum temperature, which is primordial for UTOP behavior. Doppler and sodium thermal expansion profiles were similarly averaged over each medium, with the sodium and plenum sodium thermal expansion behind added in an extra mesh on top of the fissile stack with zero power

generation. The impacts of these approximations were found to be below 1 % for the temperature estimator.

Americium has a positive effect on the ULOHS transient, barely any effect on the ULOF and an important effect on the UTOP transient. Neptunium shares a similar behavior with americium in this regard. More interesting is the behavior of the core when Curium is loaded, where with observes a degradation of the performances in ULOHS and UTOP along with a lower decrease in the UTOP performances. This is consistent with an actual increase in the Doppler Effect caused by the incorporation of curium in the fuel. From this analysis, it can be postulated that the production of Curium during irradiation has positive effects on the core behavior during a transient as it behaves like a fissile material and cancels some of the penalties created by the loading of americium.

	Ref	Np	Am	Cm
Maximal sodium temperature ULOF (°C)	1044	1052	1049	1056
Final sodium temperature ULOHS (°C)	775	761	763	795
Maximal fuel temperature UTOP (°C)	2370	2490	2482	2453

Table 12 : Evaluation of the core transient behavior depending on the minor actinides loaded

IMPACTS ON THE FUEL CYCLE

The decay heat and neutron source of the fresh and irradiated assembly was analyzed here to evaluate the impacts on the fuel cycle. The values for the fresh assemblies are given in Table 13. There is very little difference with the reference case for the neptunium and americium case. However, for the curium case, the decay heat and neutron source of the fresh assembly are significantly higher. This will require improvements in the transportation cask design in order to be able to transport these assemblies. On the other hand, it can be supposed that the three other assemblies can be transported safely using a similar cask design.

Fresh assembly	Ref	Np	Am	Cm
Heat load (kW)	0,07	0,07	0,11	1,38
Neutron source (n/s)	3,48E+03	3,27E+03	4,45E+03	4,57E+06

Table 13 : Heat load and neutron source of a fresh assembly with neptunium, americium or curium loading

Regardless of the minor actinides loaded, the decay heat of the spent assembly will be higher than for a regular one, as it can be seen in Figure 5. This increase is the most limited for the neptunium case, while the short term increase is the most important for the Am case to ^{242}Cm production. The impact on long term decay heat can be analyzed by evaluating the time necessary to reach a 7.5 or 2.5 kW washing limit, as shown in Table 15.

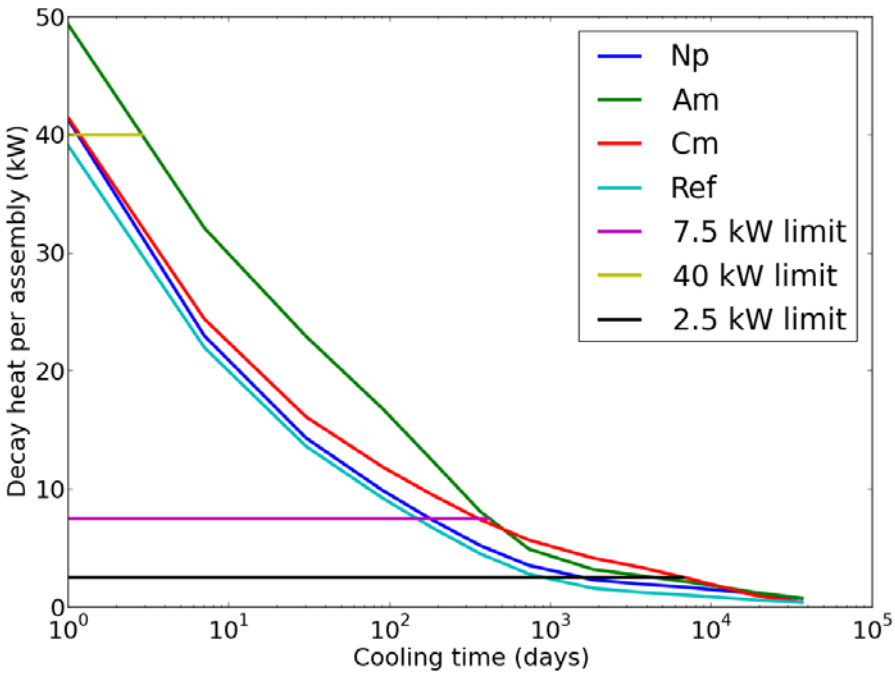

Table 14 : Evolution of the inner fuel assembly decay heat for neptunium, americium and curium loading

For the 7.5 kW limit, the reference case and the neptunium case are relatively similar as the contribution of ^{238}Pu to the decay heat remains smaller compared to the fission products contribution. The americium and curium case requires much longer cooling time due to the contribution of ^{244}Cm for both cases and ^{242}Cm for the americium case only.

For the 2.5 kW, the effect of the short-lived ^{242}Cm disappears and only the long-lived ^{238}Pu, ^{244}Cm and ^{241}Am are playing a role in the decay heat production. As the final content in ^{244}Cm is the highest for the curium case, this case requires the longest cooling time, followed by the americium case in which ^{244}Cm is produced by captures on ^{243}Am. Overall, if we consider a maximal cooling time of 5 years, minor actinides loading is not an issue if a washing limit of 7.5 kW is feasible. However, if a limit of 2.5 kW is considered, only neptunium loading is feasible in this case.

Case	Ref	Np	Am	Cm
Time to 7,5 kW (days)	148	178	416	357
Time to 2,5 kW (days)	932	1620	4611	6840

Table 15 : Cooling time to a given washing limit for neptunium, americium and curium loading

For neutron source, the neptunium and reference case are similar as neptunium addition does not lead to the production of any neutron emitting nucleus, as shown in Table 16 . On the other hand, the americium case and curium case exhibits higher neutron due to ^{244}Cm production, with the curium case being the case with the highest neutron source. It should nevertheless be mentioned that the neutron reached here in this case corresponds to the neutron source of an irradiated blanket assembly loaded with americium as studied in the previous part. Being able

to handle assemblies with such a level of neutron source is consequently necessary both for heterogeneous transmutation of americium or homogeneous transmutation of Curium.

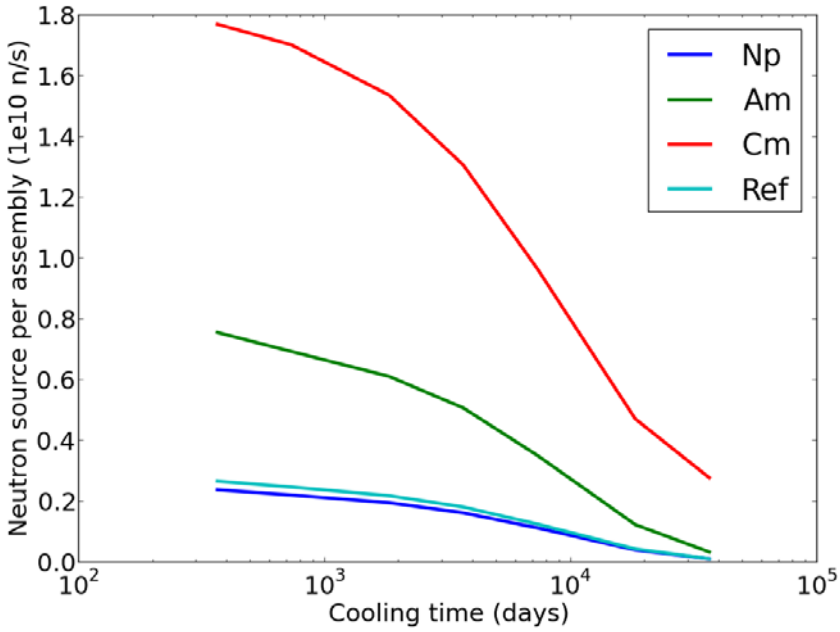

Table 16 : Evolution of the inner fuel assembly neutron source for neptunium, americium and curium loading

The impact on the fuel cycle of homogeneous transmutation of curium and heterogeneous transmutation of americium in this configuration are compared in Table 17. The two approaches yield similar impacts on the fuel cycle front end and back end decay end. Neutron production for curium-bearing assembly remains higher than for americium-bearing targets. Nevertheless, it can be concluded that homogeneous curium transmutation can be achieved using similar plants as for the heterogeneous americium approach.

Table 17 : Comparison of curium homogeneous transmutation and americium heterogeneous transmutation

Parameter	Americium hetereogeneous transmutation	Curium homogeneous transmutation
Fresh fuel decay heat (kW)	2,47	1,38
Fresh fuel neutron source (n/s/assembly)	0	4,57E+06
Irradiated decay heat @ 5 years (kW)	8,2	4,2
Irradiated neutron source @ 5 years (n/s/assembly)	1,60E+10	1,53E+10

SIMPLIFIED SCENARIO ANALYSIS

A simplified scenario analysis for all three nuclides can be carried out to highlight the feasibility of curium homogeneous transmutation compared to heterogeneous transmutation of

americium. The core production of 3600 MWth SFR V2B from [14] fueled with a low quality plutonium as shown in Table 18 will be considered. The corresponding production of minor actinides is shown in Table 19. We will consider a 32 reactor fleet generating around 400 TWeh per year corresponding to the reference level used in French scenarios studies [13].

Table 18 : Plutonium isotopic vector used in the simplified scenario study

	238Pu	239Pu	240Pu	241Pu	242Pu	241Am
Low quality Pu	3,57	47,4	29,7	8,2	10,4	0,78

Table 19: Minor actinides production of V2B fueled with plutonium from Table 18

Element	Production of reference core (kg/TWeh)	Production of curium loaded core (kg/TWeh)	Total mass balance for core with americium bearing blankets (kg/TWeh)
Np	0,42	0,2	0,62
Am	4,22	3,3	-2.51
Cm	0,55	-13,3	1.55

Given the values shown in Table 19, it is necessary to fit 20 cores with americium bearing blankets to achieve closure of the americium fuel cycle. This amounts to a total of 1680 blankets assemblies under irradiation. The total curium production of the fleet is then 37.6 kg/TWeh. It is then necessary to implement curium homogeneous transmutation in 3 reactors to achieve closure of the curium fuel cycle. This amounts to 1359 fuel assemblies under irradiation, with similar decay heat and neutron source profiles as americium bearing blankets. Considering that the irradiation time of blankets is twice as long as the one of standard fuel assemblies, it can be supposed that reprocessing of the curium bearing assemblies could fit between the reprocessing of blankets. This simple analysis requires further complete scenarios studies to evaluate it in more details, however it was shown that curium cycle closure was possible with a similar cost in terms of fuel cycle as for americium cycle closure using homogeneous transmutation in axially heterogeneous cores.

CONCLUSION

Compared to americium, it was shown that neptunium was a very good candidate for transmutation, either in the homogeneous or the heterogeneous approach. Its impacts on the fuel cycle are lower in each case but it has the same negative impacts on the UTOP transient than americium in a heterogeneous core.

Concerning curium, two main problems can be identified for the heterogeneous approach:

- The very high heat load and neutron source of the fresh assemblies
- The high neutron source of irradiated assemblies

The decay heat of an irradiated curium target is lower than the one of an Americium target, with a slightly lower cooling time. In terms of consumption, curium transmutation is more favorable due to the decay of ^{244}Cm during irradiation.

In the homogeneous case, curium was shown to have a limited impact on the UTOP transient as it behaves as a fissile nucleus. However, as its introduction in the fuel increases the Doppler Effect, it has a slightly negative impact on the flow transients. The fuel cycle impacts are comparable to the ones of an irradiated americium target. Regardless of the option chosen, the main limiting factors towards implementation of minor actinides transmutation remain linked to the fuel cycle.

It is shown that implementation of heterogeneous americium transmutation along with homogeneous transmutation of curium in a selected number of reactors could lead to a complete closure of the americium and curium fuel cycle without additional impacts on the fuel cycle compared to heterogeneous minor actinides transmutation only. More refined scenarios studies are necessary to evaluate the feasibility of this approach.

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