

Estimation of the vitrified canister production for PWR fleets integrating MSR with different fuel cycle strategies

L. Tillard^{1*}, L. Mesthiviers^{2*}, E. Capelli¹, D. Heuer², R. Maréchaux¹,
Y. Meridiano¹, E. Merle², B. Morel¹, I. Morlaes¹, G. Senentz¹

¹Orano, 92320 Châtillon, France

²LPSC/IN2P3/CNRS, Université Grenoble-Alpes, Grenoble INP, 38026 Grenoble Cedex, France

*Corresponding Authors, E-mail: lea.tillard@orano.group, mesthiviers@lpsc.in2p3.fr

KEYWORDS: Molten Salt Reactor, Equilibrium Scenario, Waste Management

MSRs are very promising systems that could be deployed in a PWR fleet to manage Pu and Minor Actinides (MAs). This study focuses on the analyses of two equilibrium scenarios where PWR fleets loaded with UOX fuel or UOX and MOX fuels integrate MSRs. This MSR concept starts and is supplied with Pu and MAs coming from UOX Spent Fuels (SF) or Pu extracted from MOX SF reprocessing mixed with MAs from both UOX and MOX SF. For the scenario conditions analyzed in this article, it is shown that a 5-year cooling time before vitrification allows the optimization of the MSR waste vitrified canister (CSD-V) production as the first CSD-V limit reached is the in-glass Fission Product (FP) incorporation content, while considering ideal separation process. The fleet composition is then determined to balance Pu and MAs streams. The CSD-V production of the whole fleet is quantified to illustrate the gain of MA transmutation in reactors. Finally, the impact of non-ideal MA separation efficiency on CSD-V toxicity is studied, showing that the separation efficiency of all MAs together has a strong impact on the waste radiotoxicity, and the gains on the vitrified waste toxicity provided by the transmutation of americium only.

Introduction

Current environmental challenges, associated to the current worldwide energy transition, require an increasingly requisite role for electronuclear energy as part of a diversified mix of low-carbon energy sources. However, to be more sustainable, the development of new fuel cycle options that would enable a future closed fuel cycle reducing the use of natural resources while recycling the heavy elements and minimising the radioactive waste production becomes more and more relevant. The basis of any such strategy is the reprocessing of spent nuclear fuel, and the extraction of valuable elements, especially trans-uranic elements (TRU) that could be used to build new fresh fuel.

Pressurised Water Reactors (PWRs) fuelled with UOX (U oxide fuels) are the most common reactors in operation nowadays. A first recycling strategy should consider the reprocessing of spent UOX fuels to recover the uranium and the plutonium (major actinides). Then, the plutonium may be used in MOX fuels (mixed oxide fuels composed of Pu ex-UOX and depleted U) such as in France for example: this is a plutonium mono-recycling strategy. The next step would be the multi-recycling of plutonium, meaning the reprocessing of spent MOX fuels to recover the plutonium for fresh fuel fabrication. One of the investigated options is the MIX fuel (Pu and enriched U mixed oxide fuels), see [Cou 21], where the degradation of the plutonium isotopic composition is balanced, to some extent, with ²³⁵U enrichment increase. However, plutonium recycling in PWR inevitably increases Minor Actinide (MA) generation, MAs which are sent to the wastes during the reprocessing stages. Those are responsible for long-term decay heat and alpha radiation of the vitrified canisters, also called CSD-V, see [Til 21]. Hence, MA recycling options in reactors could be a real asset to help managing the wastes. A lot of R&D efforts are put in the exploration of advanced nuclear systems able to burn minor actinides, to produce energy and Fission Products (FPs) with significantly shorter half-lives. This is the so-called transmutation strategy. As Molten Salt Reactors (MSRs) are fuel input flexible, see [Mes 22], they are a very promising technology to meet these challenges effectively, when integrated in a PWR fleet.

This article presents a study of steady-state scenarios, where MSRs are considered for a second transmutation strata inside a PWR fleet. In this work current existing fuels only are considered as a starting point, thus PWRs can be loaded with UOX fuel or MOX fuel. Keeping the objective of looking at the complementarity between MSRs and an existing fleet, PWRs-MOX are fuelled with 30% of MOX assemblies and 70% of UOX assemblies, whereas PWRs-UOX are fuelled only with UOX assemblies. MSRs are respectively supplied with plutonium and MAs either coming from UOX spent fuel treatment

only (open fuel cycle strategy) or with plutonium extracted from MOX spent fuels and MAs coming from UOX and MOX spent fuel treatment (plutonium mono-recycling strategy). The symbiotic fleets of interest are described in the first part. Then, in the second part, the fast neutron spectrum chloride based MSR design, its composition evolution and the associated CSD-V production are detailed. The third part presents the composition of the symbiotic fleets at equilibrium and the associated vitrified high-level waste production. The number of MSRs to deploy is determined to recycle plutonium and MA streams coming from PWRs. High level produced wastes are compared with those of a plutonium multi-recycling PWR fleet. Finally, the impact of non-ideal MA separation efficiency on CSD-V production and their radiotoxicity is evaluated.

1) Description of the two symbiotic fleets and the study methodology

Two types of symbiotic fleets composed of PWRs and chloride MSRs are analysed in this article. In the first case of study, PWRs are loaded with UOX fuels only. In this scenario, MSRs are supplied with plutonium and MAs coming from UOX spent fuels, and the number of deployed MSR is calculated to incinerate all the plutonium and MAs produced in the PWR strata. This scenario is called PAU (*Pu and MAs ex-UOX*). In the second case, plutonium extracted from UOX spent fuel is mono-recycled in MOX fuels (that fuel 30% of PWR-MOX). MSRs supplied with plutonium from MOX spent fuels and MAs extracted from both UOX and MOX spent fuels are integrated to burn the whole PWR production as in the first case. This second scenario is called PAM (*Pu ex-MOX and MAs ex-UOX and ex-MOX*). Figure 1 illustrates the two scenarios of interest.

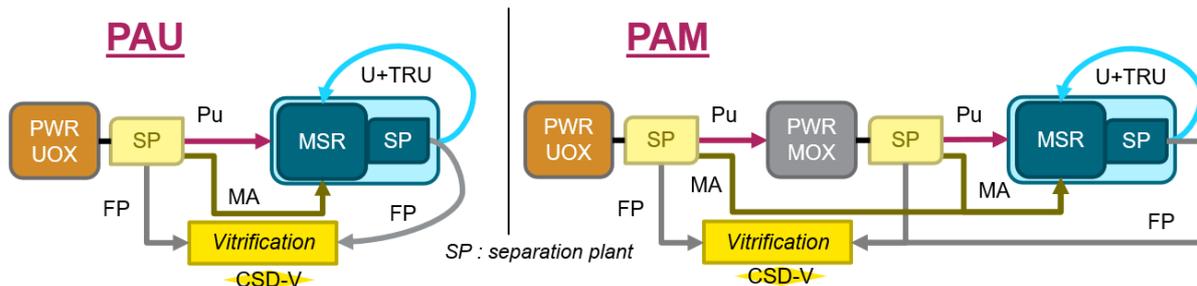


Figure 1: Schematic representation of the symbiotic fleets, "PAU scenario" (left) and "PAM scenario" (right)

The objective of this study is to estimate the vitrified canister production for both fleets considered at equilibrium [Bar 22]. This equilibrium describes the state when all PWR fresh fuel compositions are constants, and the MSR compositions is stable in time. To do so, several codes and tools are coupled as presented in Figure 2. There are three main stages. First, the PWR modelling step aims at determining PWR cooled spent fuel compositions according to different reactor and fuel characteristics using the CESAR code [Vid 12]. Then, the MSR modelling stage consists in full core depletion simulations enabling the calculation of extracted inventory with different reprocessing choices. MSR depletion simulations are performed with the REM code developed at LPSC/CNRS [Dol 14][Mer 10]. Those simulations aim at determining the plutonium and MA mass supply that guarantee the reactor criticality, given its isotopic composition. Finally, materials sent to the wastes are used to estimate the vitrified canister production and associated physic quantities with an Orano simplified non-industrial tool called AdViCE (Advanced Vitrified Canister Estimator) coupled with the SMURE package [Mep 22].

In these scenarios, the following hypotheses are considered. PWRs burn-up reaches 50 GWd/t for an electrical nominal power of 1670 MW_e, a load factor of 83% and a cycle time of circa 4 years lead to an initial heavy metal mass of about 120 tons, see [Cou 21]. UOX fuels, in this study, have an initial ²³⁵U enrichment of 4.2% (close to the CYCLADE fuel management, [Cou 17]); MOX fuels have a chosen initial Pu content of 9.6% (close to the PARITE fuel management but adjusted for a burn-up of 50 GWd/t, [Cou 17]). PWR spent fuels are cooled 5 years before reprocessing and FPs are then sent to vitrification. Within these hypotheses, CESAR calculations show that UOX spent fuel discharge per year contains circa 38 kg of MAs and 307 kg of plutonium and MOX spent fuels circa 62.5 kg of MAs and 624 kg of plutonium per year. In the PAM scenario, to mono-recycle plutonium in MOX fuel, circa 70% of PWR-UOX and 30% of PWR-MOX (30% of MOX fuel in one PWR-MOX) are required, hence MOX fuel assemblies represent 9% of all the PWR assemblies; this fleet is considered at equilibrium as all UOX spent fuels are treated for MOX fresh fuel fabrication.

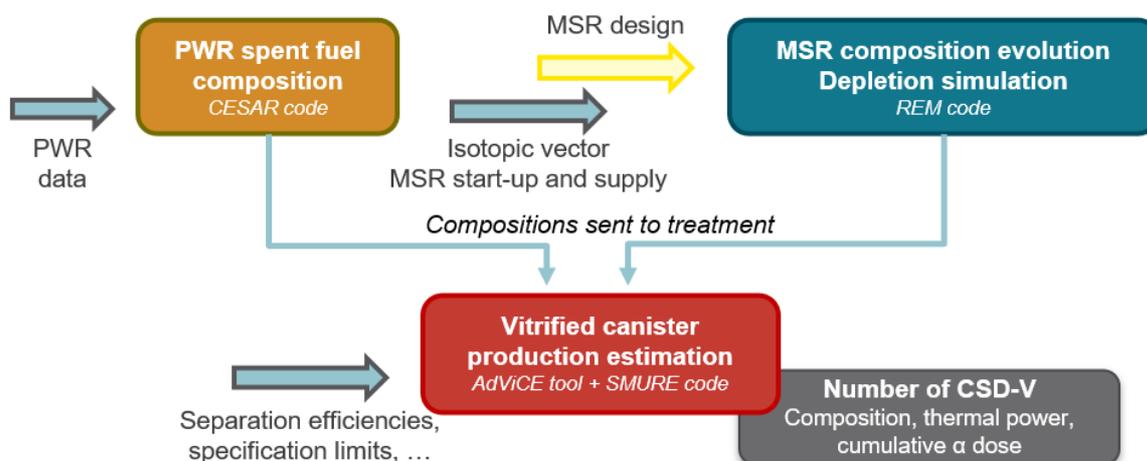


Figure 2: Schematic representation of the coupling methodology

In such open fuel cycle and plutonium mono-recycling cycle, plutonium contained in spent fuels is a valuable resource yet unused and MAs are usually considered as wastes to be vitrified. To value transuranic elements contained in PWR spent fuels, 300 MW_{th}-MSRs, with a fast neutron spectrum, are deployed in each PWR fleet. The MSR initial fissile load and the ongoing supply are a mix of plutonium and MA chlorides (see Figure 1). The isotopic vectors used for the MSR initial load and continuous supply are presented in Table 1 respectively for PAU and PAM scenarios. They are a direct consequence of the calculated PWR spent fuel compositions. Part of the salt is extracted every year from the MSR to be treated. The volume proportion of extracted salt for reprocessing is kept constant. Uranium and TRU elements present in the extracted salt are separated from the rest of the elements (solvent excluded) and reinjected into the core immediately or after 5 years of cooling. FPs extracted from the MSR salt treatment, but also gases and insoluble substances recovered in the bubbling system are sent to the vitrification. FP cooling period before their vitrification is analysed.

Table 1: Fuel isotopic vectors for MSR starting and supplying for PAU (left) and PAM (right) scenarios

PAU	Element	%mol	PAM	Element	%mol
Pu ex-UOX (89%)	Pu 238	3.1	Pu ex-MOX (77.2%)	Pu 238	4.21
	Pu 239	52.14		Pu 239	38.17
	Pu 240	25.2		Pu 240	32.44
	Pu 241	1.8		Pu 241	12.47
	Pu 242	7.76		Pu 242	12.71
MA ex-UOX (11%)	Np 237	49.14	MA ex-UOX + MA ex-MOX (22.8%)	Np 237	33.57
	Am 241	30.06		Am 241	38.34
	Am 242m	0.08		Am 242m	0.27
	Am 243	14.71		Am 243	19.27
	Cm 243	0.05		Cm 243	0.08
	Cm 244	5.45		Cm 244	7.61
	Cm 245	0.45		Cm 245	0.78
Cm 246	0.05	Cm 246	0.08		

Irradiation 50 GWd/t, cooling 5 years, CESAR calculations (see [Vid 12])

Reprocessing plants, in this work, perform ideal U, Pu and MAs (neptunium americium curium), extraction (efficiency of 100%). Only elements lighter than uranium (FPs, daughter products *etc*) are sent to the vitrification unit to produce CSD-V. The separation efficiencies used for non-actinide elements are a 99% efficiency for halogen elements and a 100% separation for gaseous elements, see [Til 21]. The impact of actinide separation efficiencies is studied in the last part of the paper.

The main physic limits used in the AdViCE tool to determine the number of CSD-V produced for a specific inventory are a maximal 3-kW thermal power at production time, a maximum of $2.5 \cdot 10^{19}$ alpha disintegrations per gramme of glass cumulated over 10^5 years and the glass chemical composition *i.e.*, a FP plus actinide maximal content of 18.5 wt% and a platinoid maximal content of 3 wt%. Besides, each glass canister weights 400 kg.

2) Focus on the chloride MSR with Pu and MAs supply

To increase the TRU content as much as possible in the 300 MW_{th}-MSR concept considered here, a fast neutron spectrum and a chloride salt are chosen (NaCl-MgCl₂). It has a melting point as low as 750 K for any ActiNide (AN) proportion up to 38.5%, which is the limit of solubility on the eutectic line [Ben 08]. The choice of isotopic vectors (see Table 1) has a tremendous impact on the reactor reactivity [Mes 22]. Consequently, the fuel salt composition chosen in the PAU scenario is 59.5NaCl-32MgCl₂-8.5(AN)Cl₃ and in the PAM scenario is 52.5NaCl-28.2MgCl₂-19.3(AN)Cl₃ for the same 2 m³ core size (power density of 150 MW_{th}/m³). The plutonium and MA supply the MSR, the daily mass added being adjusted to ensure the criticality of the system during its operation time under the nominal temperature conditions. Besides, a chlorine enrichment of 99% of ³⁷Cl is considered achievable and economically acceptable to avoid too much ³⁶Cl production, which is radioactive and volatile and can hardly be contained long enough in the wastes, the decay period of ³⁶Cl being $3.01 \cdot 10^5$ years. The geometry developed for the neutronic study is shown in Figure 3. The steel-based reflectors and the B₄C neutron protection limit the flux in the heat exchangers while avoiding too much thermalisation of the spectrum. All MSR composition evolution are calculated over 98 years with a B₄C protection renewal every 3 years and a complete fuel salt cleaning in 18.3 years.

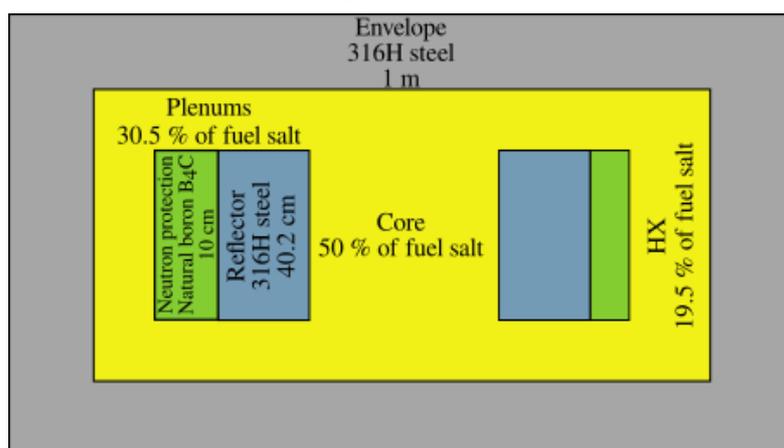


Figure 3: Schematic representation of the reactor neutronic geometry

a. Isotopic composition evolution in the system

Considering an ideal immediate actinide reinjection in the MSR core and an ideal actinides separation process (100% efficiency) allowing the total extraction of FPs from the core, solvent element excluded, Figure 4 shows the evolution of the main actinides in the fuel salt for PAU and PAM scenarios. Uranium and TRU have different evolutions as the supply isotopic compositions are different but, for both, the equilibrium is reached after about 40 years of operation. In the PAM scenario, neptunium and americium quantities decrease until stabilization while curium inventory stays relatively stable. On the other hand, in the PAU scenario, americium and curium quantities increase while neptunium inventory decreases. The behaviour difference is essentially due to the difference of initial isotopic composition in the core. The high production of uranium (mainly ²³⁴U) comes from the decaying of ²³⁸Pu highly produced in the MSRs considered in the PAM scenario, due to a higher ²⁴²Cm production caused by the higher capture rate of ²⁴¹Am. The plutonium mass evolution is similar, but its isotopic composition differs. Figure 5 presents the plutonium isotopic vector evolution for both scenarios showing the fissile isotopes being considerably consumed and fertile isotopes increasing in proportions. The decrease of actinide fissile quality is somehow compensated by the plutonium mass increase.

At equilibrium, the composition remains constant by definition. Consequently, the whole supply of TRU elements from the PWR strata is then burnt in the MSR. When considering all the main actinides, the MSR can fission 11.7 tons in total over 98 years of operation, corresponding at equilibrium to approximately 120 kg of actinides burned per year.

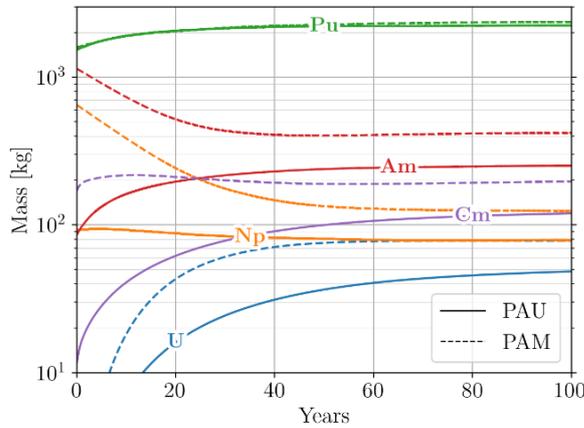


Figure 4: Main actinide mass evolution in the MSR for PAU scenario (full line) and PAM scenario (dotted line)

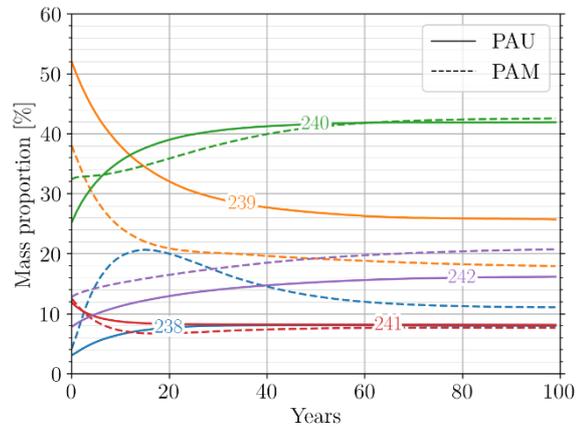


Figure 5: Plutonium isotopy in the MSR for PAU scenario (full line) and PAM scenario (dotted line)

The continuously effective actinide supply is cumulated per year and shown in Figure 6. Supply is larger at the beginning to compensate the formation of FPs that capture neutrons in the MSR considered in the PAU scenario and thus, accumulation of matter is necessary. For the PAM scenario, Figure 6 shows that the supply needed at the beginning is less important, because of the substantial masses of neptunium and americium available at the start-up, which will be first consumed. Plutonium and MA supplied quantities at equilibrium and cumulated over 98 years are presented in Table 2. As for the supply, the continuously extracted FP quantities are summed per year to be sent to the vitrification stage. It comprises FPs from the salt treatment as well as gases and insoluble substances extracted from the bubbling system. Figure 7 illustrates the contribution of both sources in the FPs production. It should be pointed out that the extracted mass is smaller than the supply mass leading to a global increase of the fuel salt mass. This figure shows that the masses of FPs do not differ much, which is not surprising as the amount of FPs is directly related to the thermal power. Besides, as the fission yields are pretty similar, there is no change in the non-soluble fraction of FPs. Extraction of insoluble and gaseous FPs is stable while soluble FP extraction raises. Due to the increase of soluble FP amount in the salt with time, their concentration increases, so do their extraction. Table 2 presents the FP quantities from the extracted salt treatment and from the bubbling system, at equilibrium and cumulated over 98 years.

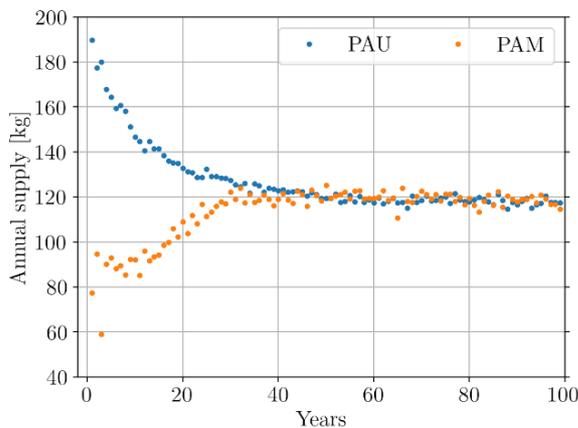


Figure 6: Annual TRU supply in the MSR for PAU scenario (full line) and PAM scenario (dotted line)

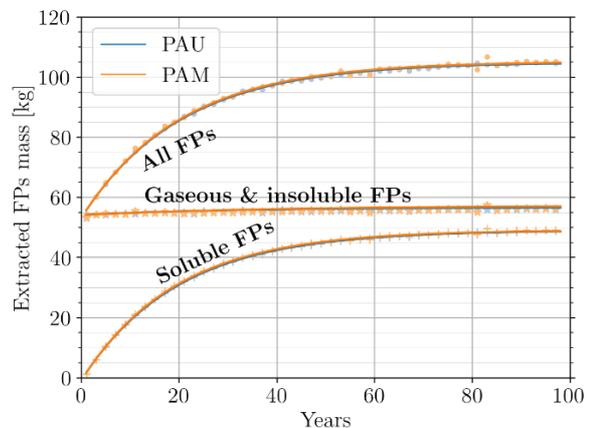


Figure 7: Annual FP extraction for the PAU scenario (blue) and the PAM scenario (orange)

Table 2: Actinides supplied quantities and waste element extracted quantities, at equilibrium and the cumulative sum over 98 years of MSR operation, for PAU and PAM scenarios

Scenario	Pu supply	AM supply	FPs from the chemical treatment	FPs from the bubbling system
PAU	106.3 kg/y at eq. 11.3 t over 98 y.	13.1 kg/y at eq. 1.4 t over 98 y.	48.7 kg/y 3.9 t over 98 y.	55.8 kg/y 5.4 t over 98 y.
PAM	93.5 kg/y at eq. 8.7 t over 98 y.	27.7 kg/y at eq. 2.6 t over 98 y.	48.8 kg/y at eq. 3.9 t over 98 y.	56.0 kg/y at eq. 5.4 t over 98 y.

b. Impact of cooling time delaying the actinide reinjection

The hypothesis of immediate actinide reinjection in the MSR core is an ideal unrealistic situation because of the residual thermal power. Hence, the extracted fuel salt should be cooled before the ideal separation process. Therefore, to evaluate the impact on the reactor isotopic composition evolution and assess whether this separation cooling is needed, a delay of 5-year is used. It is worth noting that with a 5-year Cooling Time (TC) before actinide separation and reinjection, as the fuel salt is cleaned in 18.3 years, almost a third of the salt is outside the core: the total mass of salt needed is considerably more important. These new simulations are compared with previous simulations with instantaneous reprocessing *i.e.*, for direct actinide reinjection.

Plutonium and Americium isotopy evolutions for PAM scenario are shown respectively in Figure 8 and Figure 9 with and without cooling. The main impacts of the cooling time parameter on inventory are the increase of ^{241}Am proportion due to ^{241}Pu decay outside the core and the lower ^{238}Pu proportion during the 20 first years of operation due to ^{242}Cm decay outside of the core.

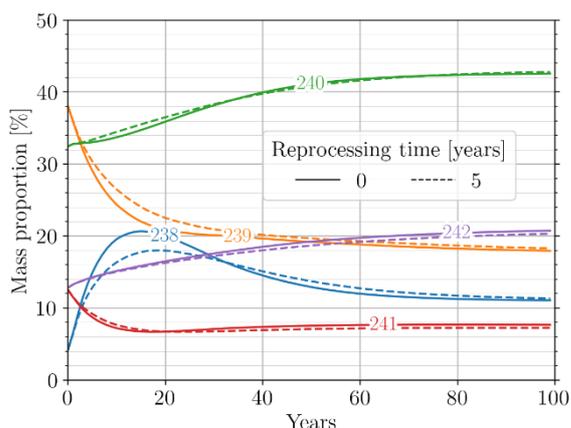


Figure 8: Plutonium isotopy evolution for TC = 0y and TC = 5y for the PAM scenario

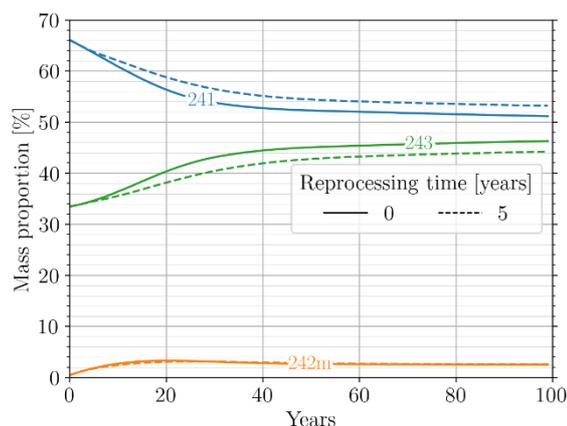


Figure 9: Americium isotopy evolution for TC = 0y and TC = 5y for the PAM scenario

The 5-year of cooling before actinide reinjection induces a higher fresh fuel injection in the system to compensate the fuel stored for cooling outside the core as illustrated on Figure 10. It is worth noting that after circa 18.3 years, both supply curves meet: there is a sufficient mass of fuel for the system to operate (inside the core and outside for 5 years of cooling). Thus, for PAM scenario, the cumulative plutonium supply increases from 8.7 tons over 98 years to 9.5 tons due to the 18.3 first years. After 40 years of operation, at equilibrium, the supply remains the same no matter the cooling time. The 5-year cooling time before actinide reinjection however does not impact the FP extracted quantities: as the MSR power is constant, the FP production remains the same and so do the extracted mass of FPs. New simulation results are superimposed with the ones visible in Figure 7.

The cooling time before actinide reinjection has small impact on the actinide in-core composition (mainly visible on ^{238}Pu and ^{241}Am) but the quantity of salt needed for the MSR operation is strongly increased.

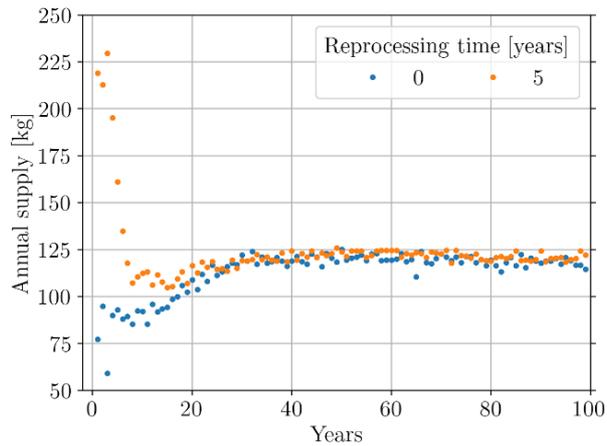


Figure 10: Annual TRU supply for the PAM scenario for $TC = 0y$ and $TC = 5y$

c. Impact of cooling time on MSR CSD-V production

To estimate the CSD-V production in this study, the simplified AdViCE tool is used. Inventory of FPs extracted from MSR are cooled before the vitrification. The shortest the cooling time before vitrification is, the highest is the thermal power at CSD-V production time. As ideal actinides separation is performed while reprocessing the salt, no heavy elements are sent to the wastes, hence the alpha radiations are negligible. A competition between the thermal power limit and the glass chemical composition (FPs including platinoids) occurs and the dominant limit is function of the cooling time before vitrification. For FPs extracted from the MSR in PAU and PAM scenarios, the CSD-V production is presented in Figure 11, on the left with 1 year of cooling, on the right with 5 years of cooling.

As shown in Figure 11, the CSD-V production are similar for both PAU and PAM scenarios. With a 1-year cooling time (left), the thermal power is the dominant limit due to short-life FPs, while after a 5-year cooling time the FP content becomes the dominant limit and allows a reduction of the CSD-V production (right). It should be noted that before the steady-state, the platinoid content limits the CSD-V production, while after circa 30 years of evolution, the FP content is restraining the vitrification. At equilibrium, circa 15.60 CSD-V/y (1-year cooling) and 1.90 CSD-V/y (5-year cooling) are produced per MSR. The cumulative glass container production for one MSR over 98 years, from the start to equilibrium, is 1560 (1-year cooling) and 180 (5-year cooling).

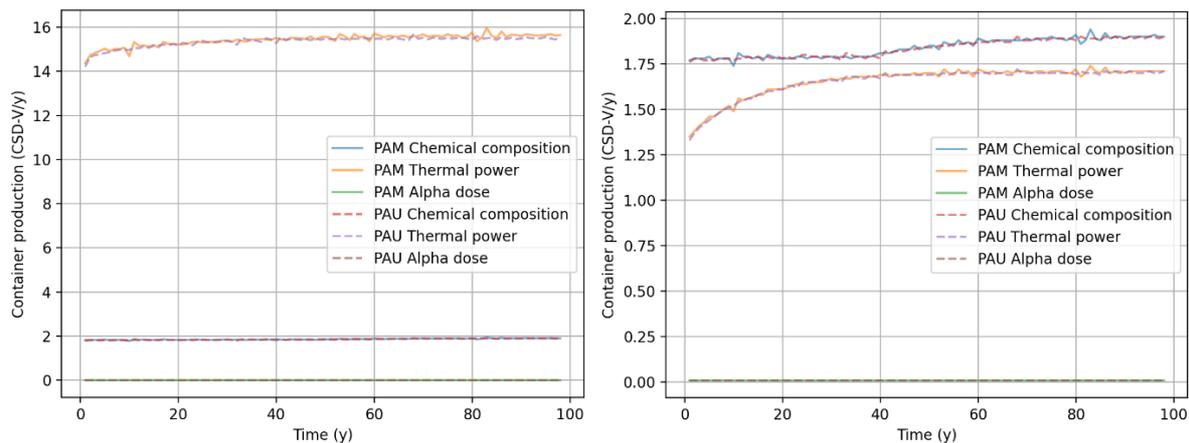


Figure 11: Produced CSD-V estimation per year per MSR, function of operation time with 1 year of cooling (left), after 5 years of cooling (right)

For the PAM scenario, the cooling time parameter is sampled between 1 and 30 years of cooling before vitrification. The CSD-V estimations are similar after 5 years of cooling as illustrated in Figure 12 where the 5-year, 10-year and 30-year the curves are superimposed; same conclusions for the PAU scenario. For the rest of the study, a cooling time before vitrification of 5 years is kept and thus consistent with PWR spent fuel cooling time before treatment.

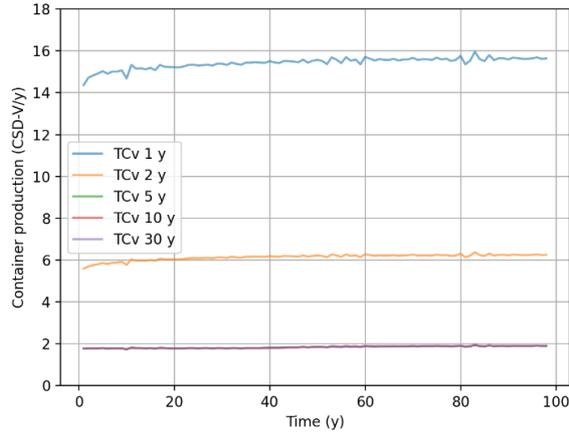


Figure 12: Produced CSD-V estimation per year per MSR with different Cooling Time before Vitrification (TCv) for the PAM scenario

The MSR CSD-V production is also estimated with a simplified module developed at LPCS/CNRS. This module uses only the three following constraints: thermal power at production time, cumulated alpha over 10^5 years and a FP plus actinide content. For PAU and PAM scenarios, after a cooling of 5 years the limit is also the FP content. Results are 176 CSD-V in total over 98 years and, at equilibrium, the CSD-V production reaches 1.80 CSD-V/y. Both methods give consistent matching values.

3) Equilibrium scenario analysis for the symbiotic fleets

The purpose of MSR deployment in a PWR fleet is to reach equilibrium for plutonium and MA inventory in the whole fleet. Actinide disappearance in MSRs should then balance actinide production in PWRs. This equilibrium is reached if the supplied actinide mass for MSR operation corresponds to the mass of transuranic elements contained in PWR Spent Fuels (SF), see Equation 1, where M_{AN-MSR}^{supply} represents the annual mass of heavy nucleus needed for the operation of all MSRs in the fleet and M_{AN-PWR}^{SF} stands for the annual mass of all TRU element produced by PWRs (after 5 years of cooling). The actinide mass contained in PWR spent fuels for the PAU scenario is explained in Equation 2, while for the PAM scenario it is explained in Equation 3, where, M_{MA-PWR}^{SF-UOX} , M_{Pu-PWR}^{SF-UOX} , M_{MA-PWR}^{SF-MOX} and M_{Pu-PWR}^{SF-MOX} are respectively the annual production of MAs and plutonium in the UOX and MOX fuel assemblies.

$$M_{AN-MSR}^{supply} = M_{AN-PWR}^{SF} \quad (1)$$

$$\text{PAU:} \quad M_{AN-PWR}^{SF} = M_{MA-PWR}^{SF-UOX} + M_{Pu-PWR}^{SF-UOX} \quad (2)$$

$$\text{PAM:} \quad M_{AN-PWR}^{SF} = M_{MA-PWR}^{SF-UOX} + M_{Pu-PWR}^{SF-MOX} + M_{MA-PWR}^{SF-MOX} \quad (3)$$

In the PAU scenario, PWRs, presented in this article first part, are loaded with UOX fuel. Plutonium and MAs contained in UOX spent fuels, cooled 5 years, feed the MSR described in this article second part. As seen in Table 2, the annual plutonium and MA supply for one MSR is approximately 121 kg/year. It represents respectively 20.2 kg/TWh_e and 2.5 kg/TWh_e. Given the respective power of each reactor, the plutonium and MA equilibrium condition leads to a power produced by MSRs that represents 19% of the total, while 81% of the power is produced by PWR-UOX; the fleet would be composed of 76 300 MW_{th}-MSRs and 26 PWRs for a 400 TWh_e/y fleet. The number of MSRs to be deployed is important due to the low power of the concept considered here and the important plutonium mass to manage: an increase of the MSR power and the plutonium management in another system (for example in PWR-MIX) leads indubitably to a decrease of this number.

For the PAM scenario, the PWR stage is composed of 30% of PWR-MOX to recycle once all the plutonium contained in UOX spent fuels in MOX fresh fuels. To supply the MSRs deployed, 13.4 kg/TWh_e of plutonium come from MOX spent fuels and 3.8 kg/TWh_e of MAs are extracted from UOX and MOX spent fuels (35% of MAs from MOX spent fuels). Consequently, in the PAM scenario, the power proportion produced by MSRs represents 14% of the total, while 60% of the power is produced by PWR-UOX and 26% by PWR-MOX. For a total electrical production of 400 TWh_e/y, it means about 19 PWR-UOX, 9 PWR-MOX and 57 MSRs of 300 MW_{th}. For the same reasons as for the PAU scenario, the MSR quantity needed to balance plutonium and MA streams seems important but

could be decreased with higher unit power MSR. Table 3 presents the Pu and MA balances for the PAU and PAM symbiotic fleets.

Table 3: Pu and MA balance, with SF for Spent Fuel and FF for Fresh Fuel for PAU and PAM scenarios

Scenario	PWR-UOX	PWR-MOX	MSR
PAU	$SF_{UOX} \sim 1.77 \text{ t/TWh}_e$ $\rightarrow Pu_{SF} \sim 20.2 \text{ kg/TWh}_e$ $\rightarrow MA_{SF} \sim 2.5 \text{ kg/TWh}_e$	/	$Pu_{supply} \sim 20.18 \text{ kg/TWh}_e$ $MA_{supply} \sim 2.49 \text{ kg/TWh}_e$ $= U + TRU_{consumption}$
PAM	$SF_{UOX} \sim 1.31 \text{ t/TWh}_e$ $\rightarrow Pu_{SF} \sim 14.9 \text{ kg/TWh}_e$ $\rightarrow MA_{SF} \sim 1.8 \text{ kg/TWh}_e$	$SF_{UOX} \sim 0.46 \text{ t/TWh}_e$ $\rightarrow Pu_{SF} \sim 5.2 \text{ kg/TWh}_e$ $\rightarrow MA_{SF} \sim 0.6 \text{ kg/TWh}_e$ $SF_{MOX} \sim 0.20 \text{ t/TWh}_e$ $\rightarrow Pu_{SF} \sim 13.4 \text{ kg/TWh}_e$ $\rightarrow MA_{SF} \sim 1.3 \text{ kg/TWh}_e$ $Pu_{FF} \sim 18.8 \text{ kg/TWh}_e$	$Pu_{supply} \sim 13.36 \text{ kg/TWh}_e$ $MA_{supply} \sim 3.96 \text{ kg/TWh}_e$ $= U + TRU_{consumption}$

As shown in the second part of this article, 1.9 CSD-V/y are produced per MSR at equilibrium after a 5-year cooling of the salt for both PAU and PAM scenarios. Besides, AdvICE calculations, for the PWRs presented in the first part and an ideal heavy element separation process, allow the estimation of the canister production per ton of reprocessed spent fuels: 0.80 CSD-V/ t_{SF-UOX} and 0.75 CSD-V/ t_{SF-MOX} . At the fleet level, the PWR CSD-V production is dominant. Both fleets induce approximately the same production of about 1.8 CSD-V/ TWh_e (PAU: 1.78 CSD-V/ TWh_e , PAM: 1.81 CSD-V/ TWh_e).

The value obtained for the symbiotic fleets at equilibrium can be compared with a Pu multi-recycling fleet at equilibrium composed only of PWRs (without MSRs): 36.6% PWR-MIX and 63.4% PWR-UOX are required to stabilize the plutonium inventory, see [Cou 21]. In [Cou 21], PWR-MIX are fully loaded with MIX fuel which have an initial Pu content of 8 wt% and the main PWR characteristics are similar with the one presented in the first section of this article. Thus, for a fleet producing 400 TWh_e/y , there are 20 PWR-UOX and 12 PWR-MIX. In this PWR fleet, MAs are not separated from the FPs at the reprocessing stage and are sent to the wastes, consequently no MA transmutation is then considered in this last scenario. Keeping these reactor proportions for the vitrification stage, circa 1.28 CSD-V is produced per ton of spent fuels treated, value estimated using AdvICE calculations (0.88 CSD-V/ t_{SF-UOX} and 1.96 CSD-V/ t_{SF-MIX}). It means a production of circa 2.94 CSD-V/ TWh_e for the PWR fleet. The MSR deployment allow the decrease of a factor 1.6 of the CSD-V production per TWh_e , because MAs are kept in the fleet for transmutation. Table 4 synthetises the results.

Table 4: Comparison between the symbiotic fleet multi-recycling Pu and MAs (U+TRU separation) and the PWR fleet multi-recycling Pu (U and Pu separation)

Scenario		Fleet composition Fraction of elec. prod.	CSD-V production
Symbiotic fleet Pu+MA multi-recycling	PAU	81.1% PWR-UOX 18.9% MSR	$\sim 1.78 \text{ CSD-V/TWh}_e$
	PAM	59.5% PWR-UOX 26.3% PWR-MOX 14.2% MSR	$\sim 1.81 \text{ CSD-V/TWh}_e$
PWR fleet Pu multi-recycling		63.4% PWR-UOX 36.6% PWR-MIX	$\sim 2.94 \text{ CSD-V/TWh}_e$

4) Impact of non-ideal MA separation efficiency

In the previous parts, actinide separation extraction process is considered as ideal for all elements: meaning that actinides are separated from other elements with an efficiency of 100% before being reinjected in the MSR core and thus only non-actinide elements are sent to the vitrification. The sensitivity of the number of CSD-V produced for MSR as well as the CSD-V radiotoxicity as a function of the MA extraction efficiency are quantified in this part to investigate the gain allowed by such a fuel

cycle with more realistic conditions.

Taking the PAU scenario, the composition sent from the core to the separation stage at 97 years *i.e.*, once the equilibrium is reached, is used to sample the separation efficiency impact on the MSR CSD-V production. A strong hypothesis is made here as it is considered that the composition of the fuel salt at equilibrium is not affected by the separation efficiency. This hypothesis may not be acceptable for all separation efficiencies, specially under 95%. This efficiency set the disappearance rate of each actinide in the fuel salt during operation. Consequently, a low efficiency induces a high actinide disappearance rate and then a low actinide inventory. The amount of MAs considered in the salt and sent to the wastes is somehow over-estimated with this methodology leading to conservative results.

With ideal MA separation, 1.89 CSD-V/y are produced after 5 years of cooling for this specific composition (equilibrium value of 1.90 CSD-V/y for this scenario, see part 2.c.). From this composition, the number of CSD-V produced is quantified using the AdVICE tool, in the following conditions: with no MA separation (MA separation efficiency: 0%), only americium extraction and reinjection in the fuel salt (the Am separation efficiency takes the following values: 95%, 99%, 99.9%, 99.99% and 100%), and finally MAs (Np, Am, Cm) are separated and reinjected with a 95%, 99%, 99.9%, 99.99% and 100% efficiency. Uranium and plutonium are extracted always at 100%. The corresponding MSR CSD-V production and the associated limit are presented in Table 5, considering the exact same limitation as earlier for the CSD-V quantity estimations.

Table 5: MSR CSD-V production for different MA separation efficiencies

Separation efficiency	CSD-V prod. per MSR per y.	CSD-V limit
All MAs: 0%	6.28	Alpha dose
Am only: 95%	5.35	Thermal power
Am only: 99%, 99.9%, 99.99%, 100%	5.33	Thermal power
All MAs: 95%	1.91	Chemical composition
All MAs: 99%, 99.9%, 99.99%, 100%	1.89	Chemical composition

Without MA transmutation 6.28 CSD-V/y are produced and the alpha dose is the first limit reached. Americium isotopes are major contributors to alpha radiations. When 95% of americium is extracted, the thermal power becomes the preponderant limit and CSD-V production is reduced of 15%. Once 95% of MAs is separated, the FP content in the canisters is limiting and the reduction reaches almost 70%. An improvement of the separation process from 95% to 99% has a visible impact on the number of CSD-V produced but from 99% to 100%, no gain in the canister generation is visible. This refinement does however impact the radiotoxicity of the CSD-V, which is an indicator of the harmful potential of the contained radionuclides quantity (inventory multiplied by ingestion dose factors).

The radiotoxicity is estimated for the different cases using the SMURE package [Mep 22] and the results are shown in Figure 13. The blue thick curve shows the results presented in the previous sections, when the MA separation is considered ideal and no TRU elements are vitrified. The thick brown curve represents the opposite case when all MAs are sent to vitrification.

Comparing the toxicity of CSD-V produced when 100% Americium is transmuted in MSR ($S_{\text{eff}} - \text{Am } 100\%$) with no transmutation (the Total curve), the gain is limited while considering the other separation efficiencies and so transmutation strategies. However, the factor of CSD-V toxicity reduction after 100 years of storage varies between 2 and 15 which is not negligible especially when considering the other americium effect on the CSD-V (*e.g.* on alpha radiations and residual heat). In addition to americium, the efficiency of neptunium and curium separation highly impacts the CSD-V toxicity. For a 95% efficiency, which is significantly below current uranium and plutonium separation efficiencies, the toxicity is reduced by a factor 20 after 10 000 years of storage in comparison with the no transmutation case, as it intersects the radiotoxicity of initial uranium needed for UOX fuels (see [Par 08]). With a 99% efficiency, the radiotoxicity is equivalent with initial uranium after 1 000 years of storage, while it becomes equivalent with an ideal MA transmutation from a 99.9% efficiency after 300 years only.

It should be reminded that the radiotoxicity estimation without actinides separation and the one with americium separation only may be biased as they rely on the same MSR fuel evolution simulation as the others *i.e.*, with ideal U and TRU extraction and reinjection in the core. Depletion simulations considering directly the separation efficiencies should be performed to validate these conclusions.

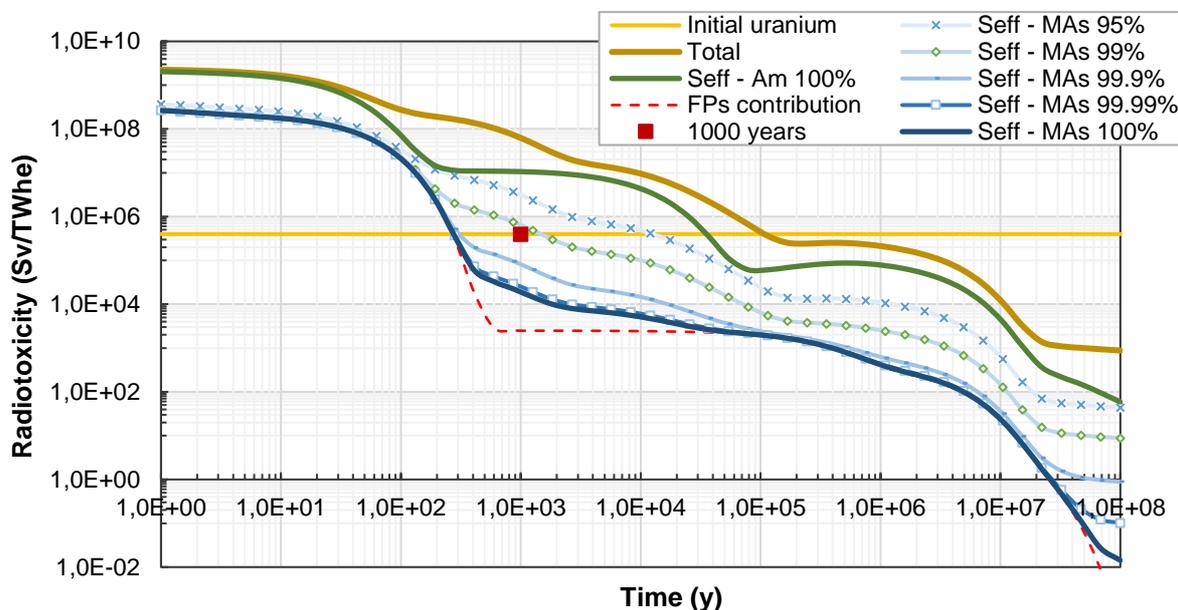


Figure 13: Estimation of the radiotoxicity evolution for MSR CSD-V: without MA separation (Total), without americium (Seff-Am 100%), with different MA separation efficiency (Seff-AMs x%)

Conclusion

This article analyses plutonium and MA management possibilities with MSR deployed in a PWR fleet, through an equilibrium scenario study. Two fuel cycle strategies are considered for the PWR strata: either PWR are fuelled only with UOX (PAU scenario) or plutonium extracted from UOX spent fuels is recycled once in PWR MOX (PAM scenario). For both symbiotic fleets an ideal actinide separation is performed and only FPs are sent to the wastes. This work focuses on a 300-MW_{th} MSR started and supplied either with plutonium and MAs coming from UOX fuel reprocessing (PAU scenario) or plutonium coming from MOX spent fuel reprocessing and MAs extracted from MOX and UOX spent fuels (PAM scenario).

The calculations show that plutonium and MA streams are balanced when MSRs produce for PAU and PAM scenarios respectively 19% and 14% of the total fleet electrical power (PWR-MOX electrical production represents 26% of the total in PAM scenario). In both scenarios the fleet produces about 1.8 CSD-V/TWh_e. In comparison, the production of a Pu multi-recycling fleet of PWR, where only U and Pu extraction is performed *i.e.*, without any MA transmutation, is about 2.94 CSD-V/TWh_e. In this PWR fleet, MAs are not managed and sent to the wastes. A reduction of about 40% of the CSD-V production per TWh_e is estimated while MSR recycling Pu and MAs are deployed.

Finally, a sensitivity study of the CSD-V production to the MA separation efficiency shows that the major gain is achievable when all MAs are transmuted (extracted and reinjected in the MSRs fuel salt) as it decreases the number of CSD-V by 70%. The extraction of only americium leads to a reduction of the number of CSD-V produced of 15% and their radiotoxicity diminution up to a factor 15. Moreover, it is shown that with at least a 99% efficiency for MA extraction a substantial decreasing of the CSD-V toxicity is observed. It becomes in fact equivalent with initial uranium toxicity needed for UOX fuels after less than 1 000 years. These conclusions should however be confirmed with more precise depletion simulations taking directly into account the separation efficiencies.

References

- [Bar 22] Barale, H., Laguerre, C., Sabatini, P., *et al.* (2022), “A new tool for the simulation of different nuclear fleets at equilibrium”, EPJ Nuclear Sci. Technol. 8 1, DOI: 10.1051/epjn/2021025.
- [Ben 08] Beneš, O. and Konings, R. J. M., (2008), “Thermodynamic evaluation of the NaCl–MgCl₂–UCl₃–PuCl₃ system”, Journal of nuclear materials, vol. 375, no 2, pp. 202-208.
- [Cou 17] Courtin, F., (2017), “Etude de l’incinération du plutonium en REP MOX sur support d’uranium enrichi avec le code de simulation dynamique du cycle CLASS”, Ph.D. Thesis, Ecole nationale supérieure Mines-Télécom Atlantique Bretagne Pays de la Loire – IMT Atlantique.
- [Cou 21] Courtin, F., Laguerre, C., Miranda, P., *et al.* (2021), “Pu multi-recycling scenarios towards a PWR fleet for a stabilization of spent fuel inventories in France”, EPJ Nuclear Sci. Technol. 7, pp. 23, DOI: 10.1051/epjn/2021022.
- [Dol 14] Doligez, X., Heuer, D., Merle Lucotte, E., *et al.*, (2014), “Coupled study of the Molten Salt Fast Reactor core physics and its associated reprocessing unit”, Annals of Nuclear Energy 64: 430–440.
- [Mep 22] Méplan, O., *et al.*, “MURE 2 : SMURE, Serpent-MCNP Utility for Reactor Evolution User Guide,” [Online]. Available: <http://lpsc.in2p3.fr/MURE/html/SMURE/UserGuide/UserGuide.html>. [Accessed 2022].
- [Mer 10] Merle-Lucotte, E., Heuer, D., Allibert, M., *et al.*, (2010), “Simulation Tools and New Developments of the Molten Salt Fast Reactor”, Contribution A0115, Proc. European Nuclear Conf. ENC2010 Barcelona.
- [Mes 22] Mesthiviers, L., Heuer, D., Merle, E., *et al.* (2022), “Actinide conversion capabilities in Molten Salt Reactors (MSR)”, Proceedings of Fast Reactor 22 Vienna.
- [Ora 22] Orano, “Contribution Orano au Débat Public sur le Plan National de Gestion des Matières et Déchets Radioactifs”, [Online]. Available: <https://pngmdr.debatpublic.fr/images/contributions/contribution-15-orano.pdf>. [Accessed 2022].
- [Par 08] Parisot, J.F. (Ed.), (2008), “Nuclear fuels”, Editions le Moniteur, for the Commissariat à l’énergie atomique.
- [Til 21] Tillard, L., Doligez, X., Senentz, G., *et al.* (2021), “Estimation of the vitrified canister production for a PWR fleet with the CLASS code”, EPJ Nuclear Sci. Technol. 7, pp. 21, DOI: 10.1051/epjn/2021020.
- [Vid 12] Vidal, J.M., Eschbach, R., Launay, A., *et al.*, (2012), “CESAR5.3: an industrial tool for nuclear fuel and waste characterization with associated qualification”, WM2012, Phoenix Arizona.