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Molten Salt Reactor to close the fuel cycle: example of MSFR multi-recycling applications

J. Guidez[a*], E. Merle[b], D. Heuer[b], S. Bourg[a], G. Campioni[a], M. Allibert[b], S. Delpech[c], P. Gauthé[a], A. Laureau[b], J. Martinet[b], J. Serp[a]

[a] CEA, Direction de l'Energie Nucléaire, France
[b] LPSC-IN2P3-CNRS / UGA / Grenoble INP, Grenoble, France
[c] IPN Orsay, IN2P3-CNRS, Orsay, France
[*] Corresponding Author, E-mail: joel.guidez@cea.fr

Abstract

On October 1, 2018, 452 nuclear water reactors were operating in the world mainly of second generation (PWR or BWR). The first third generation reactors, EPR and AP1000, have just started in China, and 2 two other ones should start next year in Europe. All these reactors produce spent fuel. Some countries have decided to manage this spent fuel as a final waste. In France, a reprocessing of this fuel is carried out in order to separate the uranium and the plutonium remaining in this spent fuel (96% of the total mass of the spent fuel) and reused as MOX fuel. Only 4% of the spent fuel is sent in to the final waste. Reprocessed uranium is stored as a strategic resource whereas plutonium is reused once in MOX fuel. Today, only a mono-recycling of the Pu plutonium is implemented, because of the difficulties to use it a second time in the REPPWR, mainly due to its isotopic composition.

Only a reactor with a fast neutron spectrum, for example a Sodium Fast Reactor (SFR), makes the multi-recycling of the plutonium efficient by taking advantage of a higher ratio of fission vs over capture. This strategy was validated by reprocessing MOX assemblies from the French Phenix SFR, and by remanufacturing fresh fuel from the recycled Pu that was irradiated again in Phenix. This experiment demonstrates the feasibility of this actinides recycling by fast reactors.

Molten Salt Reactors (MSR) with a fast neutron spectrum would also be potentially able to burn this waste while producing energy. The final performance obtained by burning plutonium or final waste is fairly comparable for any fast reactor. But on the other hand, the MSRs would allow this operation to be achieved in one step, through a continuous multi-recycling and without cumbersome operations to make new fuel assemblies, which entails considerable time and money savings. A concept of fast MSR of 3000 MWt, the MSFR (Molten Salt Fast Reactor) has been studied in France since 15 years by the CNRS and then further developed within European projects. The main technical choices for this reactor, used for this simulation, are explained.

Several options are given on the way to use the MSFR as a burner of final products today available after reprocessing. In the first case, the reactor is used as burner of available depleted uranium and plutonium oxide recovered from the reprocessing. These products are fluorinated and introduced into the reactor to achieve criticality. The evolution of the composition of the salt during the operation is given as well as the periodic additions made, and the corresponding consumption by TWh produced. The evolution of the composition of other specific elements that could alter the physico-chemical properties of the salt or the neutronic performances of the reactor (such as the lanthanides) are also given since a specific treatment of the salt has to be developed to extract them.

The same exercise is repeated in a second case with the assumption that a reprocessing of a spent MOX has been made that gives a new composition of the plutonium.

In Finally in a third option, all the minor actinides (americium, curium and neptunium) are recovered and recycled together, with uranium and plutonium.

The possible consumption of waste by TWh produced is determined. Changes in the composition of salt over time give also an idea of the quantities of products to be extracted by salt treatment.

In conclusion, this paper gives quantitative estimations of the possibilities to operate a MSR in the U/Pu fuel cycle using only the available products obtained by reprocessing the spent nuclear fuel of a fleet of second- or third-generation water reactors.

KEYWORDS:

Multi-recycling, waste reduction, fast reactors, Molten Salt Reactor, breeder and burner

1) Introduction

The aim of this paper is to study the neutronic operation of a molten salt reactor using the products available at the La Hague reprocessing plant, as part of the current operating cycle of the French fleet.

This simulation is based on the concept of molten salt fast reactor, the MSFR, initially defined at the CNRS and subject of the EVOL and SAMOFAR European projects. The main parameters of the reactor are specified in this paper.

We are in the context of neutronic evolving calculations to verify the criticality of the concept, the consumptions made, and the necessary reloads. The evolutions of the main products are given, including the products to be extracted during operation by reprocessing.

2) Review of the French fuel cycle and related issues

The enrichment of the uranium needed to manufacture the fuel of the 58 PWRs of the French fleet leads to an annual production of about 7000 tons of depleted uranium (depU) per year. About 340 000 tons are now stocked, available and unemployed.

The reprocessing of the PWR fuel leads to an annual production of about 10 tons of Pu and 950 tons of reprocessed uranium (URT). These 10 tons of Pu, mixed with 110 tons of depU allow manufacturing annually 120 tons of MOX fuel used today in twenty-two 900MW PWR reactors. Until 2013, the URT was re-enriched to produce a fuel called URE, used in four 900MW reactors. The 800 tons of URT / year without employment were stored. The resumption of URE fuel fabrication is announced for 2023.

The reprocessing of used MOX is currently not carried out because the uranium and plutonium that would be obtained would not have the isotopic quality allowing their effective use in PWRs. They are currently stored, waiting for reprocessing for a fourth-generation reactor capable of burning them.

It is known that fast neutron reactors are able to burn these products after separation, and in particular fast sodium reactors whose industrial feasibility is demonstrated. In this paper, we look how a MSR could theoretically perform this same operation.

Several cases of calculations are looked at using the different available materials, i.e. the depleted uranium, the uranium and the plutonium obtained after the reprocessing of the PWR assemblies, the uranium and the plutonium obtained after reprocessing of the used MOX, as well as the actinides obtained in this reprocessing if one decided to separate them.

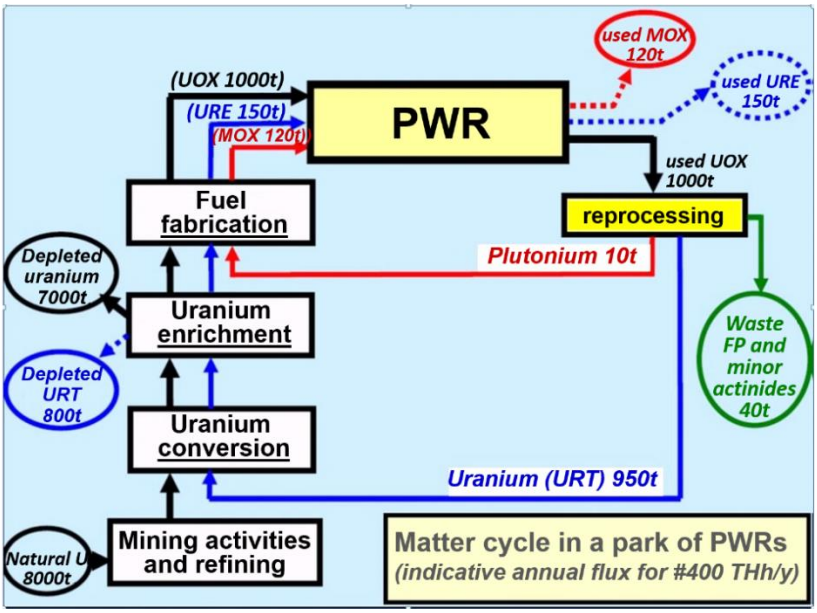

Figure 1: Cycle of materials in the French 400 TWh/year fleet of pressurized water reactors

It may be noted that this actinides separation could allow a closure of the cycle with final waste whose activity would be strongly reduced.

3) Methods and parameters used for these studies

Code used

Molten salt reactors have different ways of operation and management than solid fuel reactors. Tools have been specifically designed to simulate these reactors within the CNRS /LPSC. The code used for the studies presented in this paper is the coupling of a probabilistic neutronic code to a material evolution code developed to be adapted to the operating characteristics of a liquid fuel reactor (REM code [2]). This tool, developed over the past twenty years, has already been validated on the MSRE and MSBR historical concepts [3] and then via a European benchmark on the MSFR reactor [4].

This code works by assuming an addition of plutonium and uranium during reactor operation, via a salt control unit, to maintain criticality.

In these neutronic calculations, an extraction of the fission products is simulated. The gaseous fission products (He, N, Ne, Ar, Kr and Xe), the noble metals (Nb, Mo, Tc, Ru, Rh, Pd, Ag) as well as Sb and Te are extracted in a very short time (about 30 s) either by bubbling or by deposition on a trap for metals. The lanthanides, Y, Zr and the alkaline and alkaline earth elements (Na, Mg, K, Ca, Rb, Sr, Cs, Ba, Fr, Ra) are extracted with an hypothesis of characteristic time of 1 year.

Parameters of the MSFR concept of studies

The power considered for the “reference MSFR” used here is 3000 MWth, which can produce about 1400 MWe [5,6]. This is the power retained for most paper reactors under study (Russian concept MO-SART [7], etc. ..) to allow comparisons on a typical case. The MSR concept used for the calculations presented in this article has been optimized as breeder in the thorium fuel cycle. The idea here was to check the ability of a fast spectrum MSR to both burn and breed in the U/Pu fuel cycle before further studies on a MSR dedicated to such an application. The points that will be optimized are thus mentioned all along the article as they appear in the studies.

- **Choice of salt**

The fuel salt is circulating in the MSFR and acts also as the coolant. This places specific constraints on the liquid used as fuel in such a reactor. There are two main salt families that can be used: chloride and fluoride.

Chloride salts have several qualities: they have a harder spectrum, they are able to operate at slightly lower temperatures and the Pu solubility is higher. For example, they were selected by two American start-ups who are aiming at the incineration of waste (Terrapower and Elysium).

However, they have some disadvantages:
- If an enrichment of the natural chlorine in ^{37}Cl is not carried out, one obtains a significant production of ^{36}Cl, which is a problematic waste because of its period (300 000 years) and its difficult containment in final waste.
- Lack of experimental feedback on its functioning and on corrosion problems.
- The harder neutron spectrum induces a larger amount of irradiation damages (dpa) on the structures.

Nevertheless the choice of salt for such applications is under working and was not optimized in this study.

Fluoride salts have been used for the studies presented here, as for the Russian project MOSART [1], with the corresponding experience feedback available from the MSRE. In this family, several salts are available. In particular the FLiBe salt with a certain percentage of beryllium allowing lower operating temperatures. In our calculations, a pure LiF (Lithium Fluoride) salt was considered as a solvent, corresponding to the reference MSFR concept,

- **Choice of temperatures and specific powers**

For this simulation, the current data of the reference MSFR (333 MWth/m^3 of salt in the core) and a core input / output temperature rise of 100°C with a mean fuel temperature of 700°C were used.

- **Solubility U / Pu / Actinides**

In the presented calculations, we imposed a proportion of 22.5 mol% of heavy nuclei to be on a eutectic which makes it possible to lower the temperatures. In this salt, the maximum U / Pu solubilities are known. The solubility limit of Pu is of the order of 6 to 8% at reactor operating temperatures [8].

- **Reprocessing**

During the reprocessing the extractions are driven by the redox potentials, which imposes an order of extraction which is not the one desired. Some extracted elements (uranium / actinides...) are then to be returned to the fuel salt. At the end of the extraction, elements such as lanthanides and zirconium will constitute the final waste. In our case, during the reprocessing the uranium will be extracted in the first steps (for return to the fuel salt), requiring a necessary increase of the temperature of the mixture to continue the reprocessing in liquid phase. Here again, an optimization of the salt is necessary, for a fuel reprocessing which is not yet finalized today.

Conclusion

The calculations were made on the basis of the initial parameters of the MSFR, but as mentioned a certain number of parameters could be optimized in the context of the use of the reactor considered here, both as regenerator in the U / Pu cycle and as incinerator. This optimization was not performed as part of this study.

4) Results of calculation case #1

We have chosen to use the widely available depleted uranium, and the plutonium from the reprocessing of spent UOX PWR fuel (also available but used today for the manufacture of PWR MOX).

Other choices, as using URT also available, or only plutonium in a burner option, will be investigated also in the near future. Indeed the presence of fertile uranium in the fuel salt brings a breeding aspect with creation of plutonium, which reduces its consumption and thus the capacity of incineration. The idea here is to study a mixed concept combining both burning and breeding capabilities. It should also be noted that the use of blankets would make it possible to imagine breeder options.

In the simulation code, fertile material (here depleted uranium) and fissile material (here Pu and minor actinides) are added periodically to keep the core critical while maintaining the proportion of heavy nuclei initially chosen (22.5%) which impacts the physicochemical properties of the salt which is also the coolant.

For the same reason, the code continuously extracts a number of neutron poisons (lanthanides, zirconium ... see section 3b), which will be the final waste.

The isotopic composition of depleted uranium is 0.25% of ^{235}U The isotopic composition of Pu is that obtained after reprocessing a spent UOX PWR fuel, after arbitrary but classical aging of 10 years, separation and then aging for 5 years. The isotopic composition obtained is as follows:

Isotope	Atomic proportion [%]
^{238}Pu	1.6%
^{239}Pu	61.2%
^{240}Pu	26.3%
^{241P}u	3.78%
^{242}Pu	6.15%
^{241}Am	1.01%

At startup, the fuel contains 35 tons of depleted uranium and 9.2 tons of Pu. This composition has been adjusted to have an exactly critical heart. The core is then supplied with depleted uranium (fertile material) and with Pu of identical isotopic composition to that of starting .d

Under these conditions, the evolution of the composition in the salt is given in figure 2. We see that the main balances are put in place before the first twenty years.

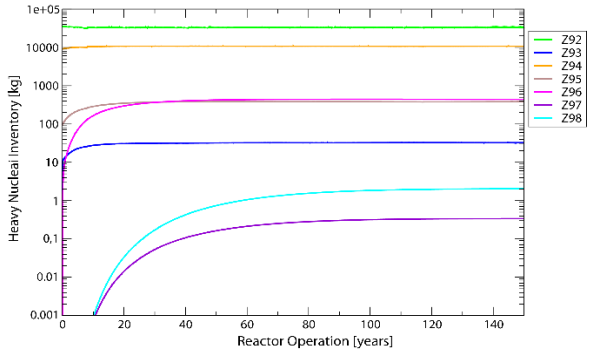

Figure 2: Evolution of the heavy nuclei inventory for the case using Pu from used UOX

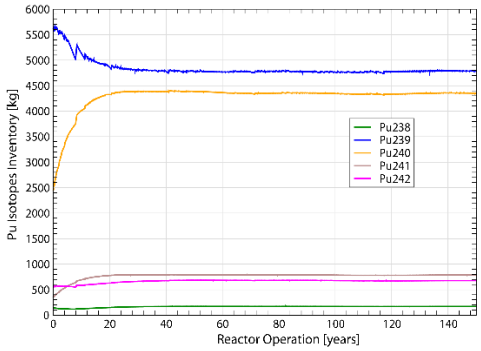

Figure 3: Evolution of the Pu isotope inventory for the case using Pu from used UOX

In the same way (cf figure 3) the inventories of Pu compositions stabilize in the first years. Consumptions of U and Pu are presented in section 7, and are about 1.15 tons / year of depleted uranium and only about 30 kg / year of plutonium.

5) Results of calculation case #2

In this second study, we start from the assumption of a reprocessing of the used MOX available today where we do not separate the minor actinides from the fission products. So we recover only U and Pu, with a degraded isotopy but usable in the fast MSR. Here the reactor will be started up and fed with depleted uranium as fertile material and with Pu as fissile material with the isotopic vector of Pu from spent MOX, aged 10 years then separated and aged 5 years (leading to the production of ^{241}Am from ^{241}Pu decay). The isotopic composition obtained is as follows:

Isotope	Atomic proportion [%]
^{238}Pu	4.0%
^{239}Pu	32.5%
^{240}Pu	35.4%
^{241}Pu	7.35%
^{242}Pu	12.7%
^{241}Am	8.1%

Here again the parameters of the data could be changed in future systematic studies, either on the choice at the level of uranium (one could have taken the uranium resulting from the reprocessing, or the URT, ...), or on the time of MOX aging.

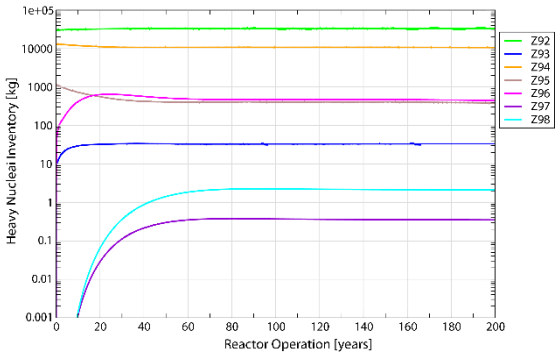

Figure 4: Evolution of the heavy nuclei inventory for the case using Pu from used MOX fuel

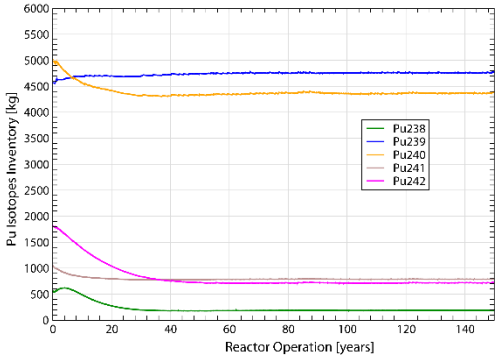

Figure 5: Evolution of the Pu isotope inventory for the case using Pu from used MOX fuel

At startup, the fuel contains 30.2 tons of depleted uranium and 13 tons of Pu. This composition has been adjusted to have an exactly critical core. Figure 4 then gives the evolution of the heavy nuclei compositions in the fuel salt for this second case.

The evolution of plutonium inventories is presented in Figure 5. The conclusions are identical to that of the previous study.

6) Results of calculation case #3

For this third study, we consider the theoretical case where the reprocessing of used MOX would take place with separation of the minor actinides. In this case, the fissile material could be a mixture of Pu and MA coming from used MOX fuel. It should be noted that this option corresponds to the implementation of a minor actinide/fission products separation technique that is feasible from the research results, but is not in place today. The main interest of such a separation is to reduce the toxicity of final waste, down to a period of 300 years, after which time this toxicity would correspond to that of natural uranium. This could help revisiting some long-term storage options and also improving social acceptance.

We then look in our calculations how these actinides can be integrated into the MSFR cycle. Here the reactor will be fed with a fertile material still essentially of depleted uranium and as fissile material the fissile isotopes of Pu and minor actinides (isotopic vector from a used MOX fuel aged 15 years). The isotopic composition obtained for the fissile material is as follows (in atomic percentages):

Isotope	Atomic proportion [%]
^{238}Pu	3.57
^{239}Pu	29.0
^{240}Pu	33.0
^{241}Pu	6.54
^{242}Pu	11.3
^{244}Pu	2.44E-03
^{236}Np	3.17E-06
^{237}Np	0.64
^{239}Np	3.11E-06
^{241}Am	9.99
^{242}Am	0.04
^{243}Am	3.55
^{243}Cm	0.02
^{244}Cm	1.65
^{245}Cm	0.59
^{246}Cm	0.09
^{247}Cm	2.84E-03
^{248}Cm	2.95E-04

At start-up, the fuel contains 28.5 tons of depleted uranium, 13.4 tons of Pu, 100 kg of Np, 2.1 tons of Am and 380 kg of Cm. This composition was adjusted to have an exactly critical core by fixing the proportion of heavy nuclei (U, Pu and MA) to 22.5 mol% and respecting the isotopic compositions given above. Here again other calculation hypotheses will be considered in coming systematic studies, such as the choice of uranium or the quantities of minor actinides to be injected or the cooling time of spent MOX fuel. Figure 6 shows the evolution of the heavy nuclei compositions in the fuel salt of the fuel circuit.

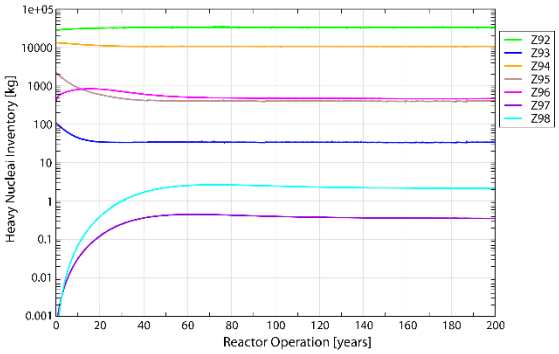

Figure 6: Evolution of the heavy nuclei inventory for the case using Pu + MA from MOX used fuel

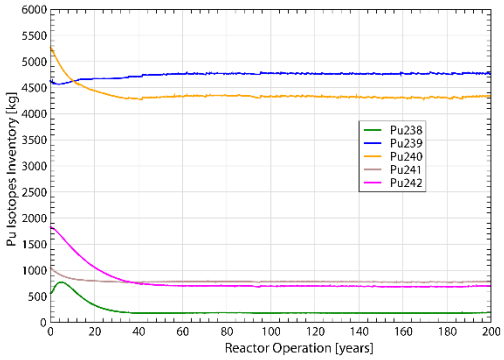

Figure 7: Evolution of the Pu isotope inventory for the case using Pu + MA from MOX used fuel

The evolution of the composition of the Pu isotopes is shown in Figure 7. Here again the conclusions are identical to those of the first 2 studies as to the equilibrium of the inventory of heavy nuclei in the fuel salt.

7) Assessment of U / Pu consumption in the three calculation cases presented

Figure 8 shows the depleted uranium consumption in the three cases presented. We see that this consumption is identical in all three cases. This is normal because it corresponds to the injection of the fertile matter that does not affect the criticality level required but is led by the power produced by the reactor. We therefore have an annual consumption of depleted uranium of about 1.15 tons for an MSR of 1400 MWe.

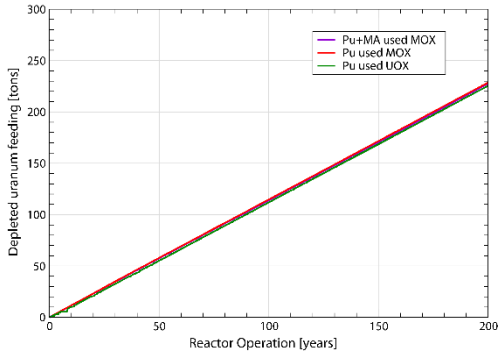

Figure 8: Feeding in fertile matter (depleted uranium) for the three studies presented

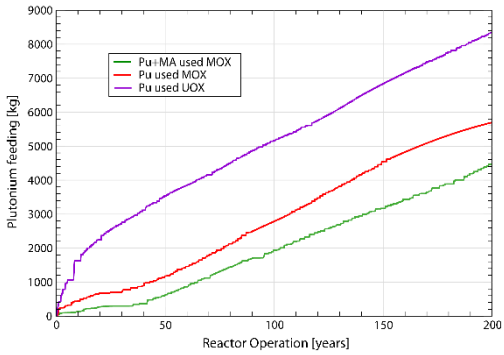

Figure 9: Feeding in plutonium (see each study for the corresponding isotopic composition)

Figure 9 gives the consumption of corresponding Pu in the three cases studied. It can be seen that the initial isotopic composition of Pu plays an important role in the establishment of the neutron equilibrium in the core and that in the case of Pu from a PWR fuel a higher Pu supply is required in the first years. The Pu supply is higher in the first years for the first case (Pu of the used UOX) compared to the other two cases using Pu from spent MOX because the isotopic vector of Pu is degraded in the first case given the high proportion of ^{239}Pu injected, element consumed quickly. In both cases based on Pu from MOX, the presence in the reactor does not degrade the isotopic vector similarly, on the contrary the presence of minor actinides leads to the production of fissile elements by neutron capture.

On the other hand, the feeding slopes are approximately identical at about 32 kg / year. This amount of external Pu used to feed the reactor, whose composition is different for the 3 cases considered but which is very small compared to the whole Pu consumption per year (around 1 ton per GWe mainly produced from ^{238}U) explains why the Pu isotopic compositions tend to the same values at equilibrium in the 3 calculations (see figures 3, 5 and 7).

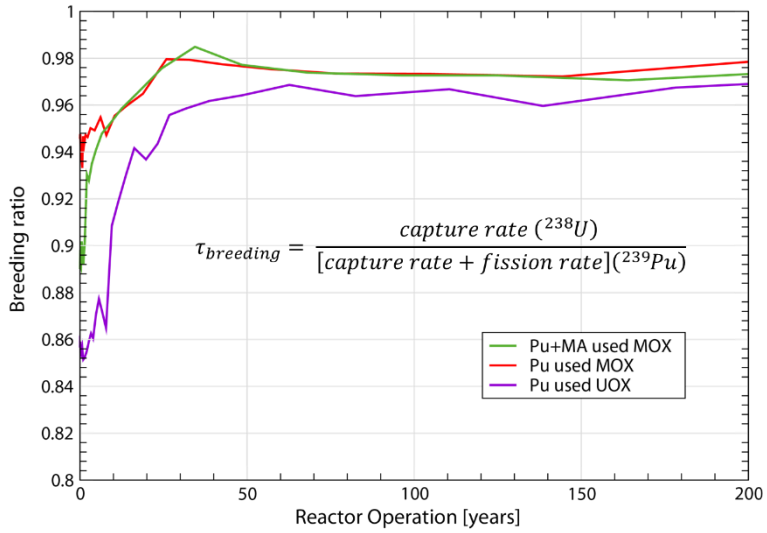

Figure 10: Breeding ratio of the core for the 3 studied cases

Finally associated with these feeds, Figures 10 and 11 show the rates of regeneration and incineration of these three cases studied, which correspond to almost regenerative reactors (regeneration rate between 0.96 and 0.98) while allowing to burn injected transuranic elements.

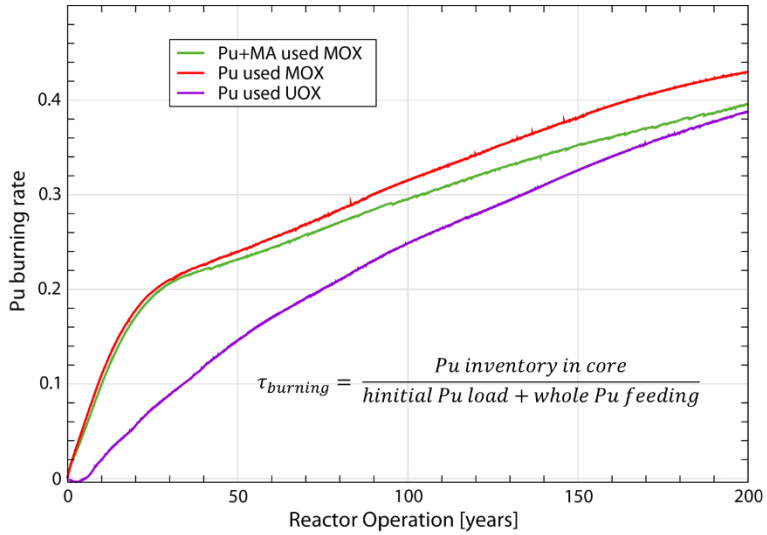

Figure 11: Burning rate of the reactor for the 3 cases studied

8) Summary of extractions

To maintain criticality, the code assumes a continuous extraction of lanthanide or zirconium type poisons. It is a "theoretical" extraction that does not correspond to a reprocessing scheme defined and validated. In particular, the extraction coefficients are "ideal" at 100% which is never the case. Figure 12 gives the evolution of the compositions in the fuel salt, with the extraction hypotheses retained. It can be seen that the concentrations of the elements which are extracted (lanthanides, zirconium, alkalis, etc.) are stabilized quite rapidly. Figure 13 gives the quantities of products extracted from the mixture during the operation of the reactor.

We see an extraction roughly equivalent in mass to the reactor feed. But the elements obtained (metals, lanthanides, alkaline earths, zirconium...) are final wastes with a significantly reduced activity.

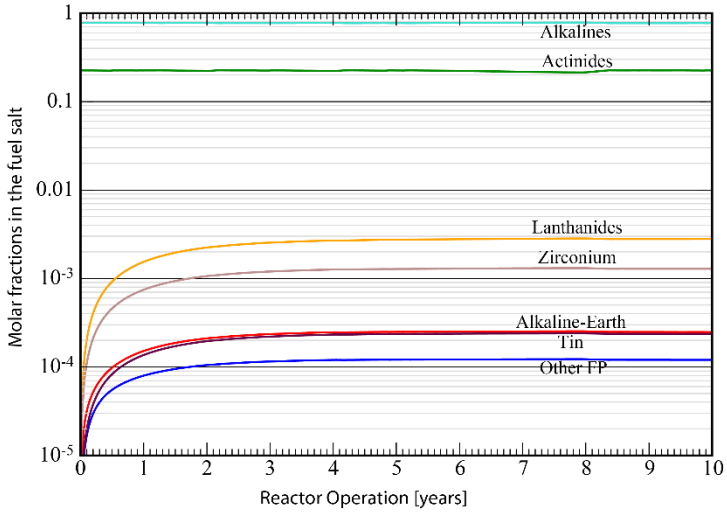

Figure 12: Molar proportions of the different elements in the fuel salt

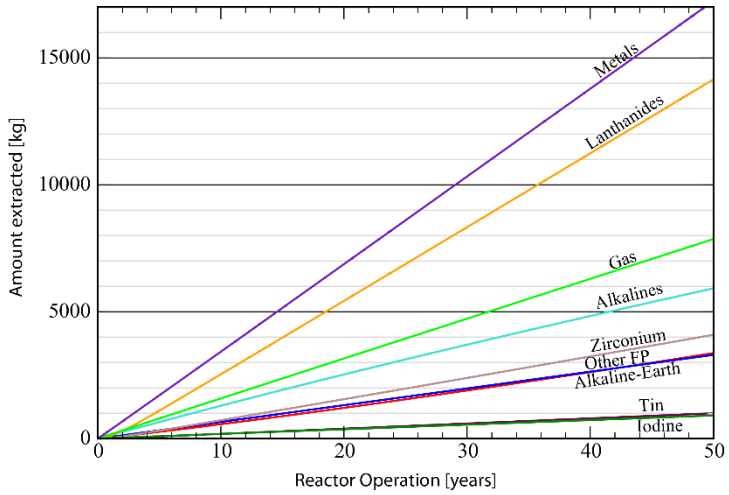

Figure 13: Fission products extracted by the processing units during the reactor operation

One can notice the reduction of the toxicity duration of the final waste. Figure 14 gives a calculation of the evolution over time of the radiotoxicity of this residual waste. The calculation is carried out after 30 years of output from the reactor. This waste is at a level of toxicity corresponding to natural uranium, after about 300 years.

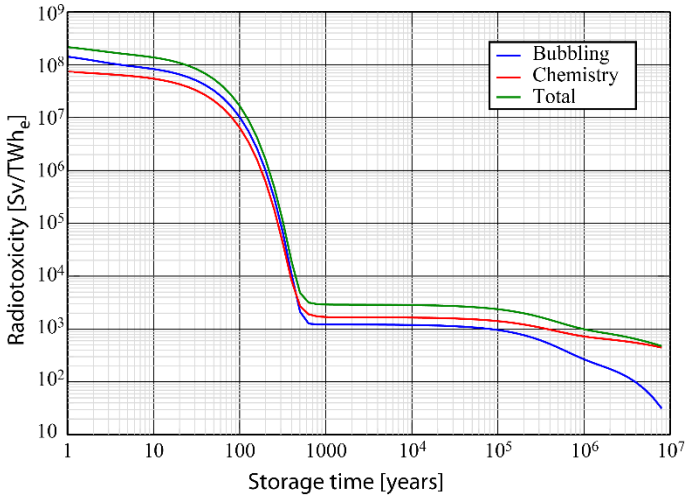

Figure 14: Time evolution of the final waste radiotoxicity

9) Conclusions

The neutronic calculations presented were carried out with the initial parameters of the existing “reference MSFR” without further optimization of them (choice of salt, specific power, etc.) vis-à-vis the mode of use of the reactor in the U / Pu cycle.

Three calculation cases are presented using depleted uranium with successively Pu from a reprocessed PWR used UOX fuel, then Pu from a used MOX, finally Pu and minor actinides from a used MOX fuel. The results show the great neutronic flexibility of use of this type of reactor with in all three cases a consumption of natural uranium of about 1.15 t / year and about 30 kg of plutonium per year. So we are almost in iso-breeder mode. Further studies are led to use plutonium without uranium to optimize a Pu burner version of the concept.

As Lavoisier said, "nothing is lost, nothing is created, everything is transformed". It is therefore necessary to extract quite the same quantities of final waste, i.e. about 1.15 tons per year of metals, alkaline earths and lanthanides.(in fact with E=m C2 , calculations show that only about one kilogram /year is converted in energy) It should be noted that this extraction is made theoretically by the code but does not correspond today to a reprocessing process defined and validated. This final waste then has activities and periods much lower. After about 300 years, this activity becomes equivalent to that of natural uranium, which has consequences for the storage options and would also allow better societal acceptability of this final waste.

Acknowledgment

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