



Towards the thorium fuel cycle with molten salt fast reactors



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ABSTRACT

There is currently a renewed interest in molten salt reactors, due to recent conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts. It has been recognized as a long term alternative to solid-fueled fast neutron systems with a unique potential (large negative temperature and void coefficients, lower fissile inventory, no initial criticality reserve, simplified fuel cycle, wastes reduction etc.) and is thus one of the reference reactors of the Generation IV International Forum. In the MSFR, the liquid fuel processing is part of the reactor where a small side stream of the molten salt is processed for fission product removal and then returned to the reactor. Because of this characteristic, the MSFR can operate with widely varying fuel compositions, so that the MSFR concept may use as initial fissile load, ^{233}U or enriched uranium or also the transuranic elements currently produced by light water reactors. This paper addresses the characteristics of these different launching modes of the MSFR and the Thorium fuel cycle, in terms of safety, proliferation, breeding, and deployment capacities of these reactor configurations. To illustrate the deployment capacities of the MSFR concept, a French nuclear deployment scenario is finally presented, demonstrating that launching the Thorium fuel cycle is easily feasible while closing the current fuel cycle and optimizing the long-term waste management via stockpile incineration in MSRs.

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1. Introduction

The Generation-IV International Forum (GIF) for the development of new nuclear energy systems has established a set of goals as research directions for nuclear systems (US DOE, 2002): enhanced safety and reliability, reduced waste generation, effective use of uranium or thorium ores, resistance to proliferation, improved economic competitiveness. Molten Salt Reactors (MSRs) are one of the systems retained by this forum in 2002.

The CNRS has been involved in molten salt reactor studies since 1997. Starting from the Oak-Ridge National Laboratory Molten Salt Breeder Reactor project (Whatley et al., 1970), an innovative concept called Molten Salt Fast Reactor or MSFR (Nuttin et al., 2005; Mathieu et al., 2006, 2009; Forsberg et al., 2007; Merle-Lucotte et al., 2008, 2009a,b) has been proposed. This concept results from extensive parametric studies in which various core arrangements, reprocessing performances and salt compositions were investigated with a view to the deployment of a thorium based reactor fleet on a worldwide scale. The primary feature of the MSFR concept versus that of other older MSR designs is the removal of the graphite moderator from the core (graphite-free core), resulting in a breeder reactor with a fast neutron spectrum and operated

in the Thorium fuel cycle, as described in Section 2 of this paper. The MSFR has been recognized as a long term alternative to solid fueled fast neutron systems with a unique potential (excellent safety coefficients, smaller fissile inventory, no need for criticality reserve, simplified fuel cycle etc.) and has thus been officially selected for further studies by the Generation IV International Forum as of 2008 (GIF, 2008, 2009; Boussier et al., 2012; Renault et al., 2009).

In the MSFR, the liquid fuel processing is an integral part of the reactor where a small sample of the molten salt is set aside to be processed for fission product removal and then returned to the reactor. This is fundamentally different from a solid-fueled reactor where separate facilities produce the solid fuel and process the Spent Nuclear Fuel. The MSFR can be operated with widely varying fuel compositions thanks to its on-line fuel control and flexible fuel processing: its initial fissile load may comprise ^{233}U , ^{235}U enriched (between 5% and 30%) natural uranium, or the transuranic (TRU) elements currently produced by PWRs. The characteristics (initial fissile inventory, safety parameters, and deployment capabilities) of each of these MSFR starting modes are detailed in Section 3, while the transition from today's second and third generation reactors to the Thorium fuel cycle is illustrated in Section 4 through the deployment capacities of a MSFR park in the context of France.

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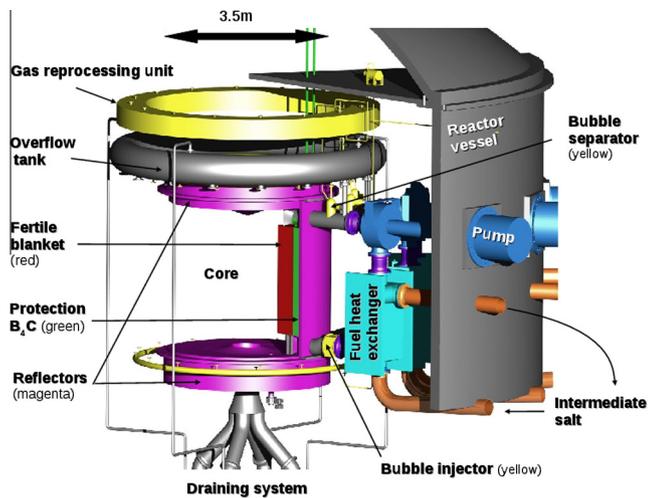


Fig. 1. Pre-design of the fuel salt circuit of the MSFR.

2. Molten Salt Fast Reactor (MSFR) concept

2.1. System description

The standard MSFR is a 3000 MWth reactor with a total fuel salt volume of 18 m³, with a mean fuel temperature of 750 °C. In order to allow exploration and discussions on possible ranges for physical and chemical parameters, basic drawings have been worked out in relation to the calculations. Fig. 1 describes one of the optimized geometrical configurations of the system. The core consists of a compact cylinder (height/diameter ratio =1) where the liquid fluoride fuel salt flows freely from the bottom to the top of the central component with no solid moderator. The return circulation of the salt (from the top to the bottom) is fragmented into 16 groups of pumps and heat exchangers located around the core (Brovchenko et al., 2012). The fuel salt completes a full cycle in 3–4 s. At any time, half of the total fuel salt volume is in the core and half in the external fuel circuit (salt collectors, salt-bubble separators, fuel heat exchangers, pumps, salt injectors and pipes).

The MSFR simulations have been performed using a binary fluoride salt, composed of LiF enriched in ⁷Li to 99.995% and a heavy nuclei (HN) mixture initially composed of fertile thorium and fissile material, ²³³U, enriched ²³⁵U and/or Pu and minor actinides. The (HN)₂F₄ proportion is set at 22.5 mol% (eutectic point), corresponding to a melting temperature of 565 °C. The choice of this fuel salt composition rests on many systematic studies (influence of the chemical reprocessing on neutronic behavior, burning capabilities, deterministic safety level, deployment capabilities) (Merle-Lucotte et al., 2009a,b, 2012).

This salt composition leads to a fast neutron spectrum in the core, as shown in Fig. 2 where the fast neutron spectrum of the simulated reference MSFR is compared to the spectra of 2 solid-fuel reactors: a Sodium-cooled Fast neutron Reactor (SFR) and a thermal Pressurized Water Reactor (PWR). The large Na capture cross-section appears clearly on the red curve at 2.8 keV, while the inelastic scattering cross-section of fluorine shows on the green curve between 0.1 MeV and 1 MeV.

The external core structures and the fuel heat exchangers are protected by thick reflectors made of nickel-based alloys, which have been designed to stop more than 99% of the escaping neutron flux. The radial reflector includes a fertile blanket (50 cm thick-red¹

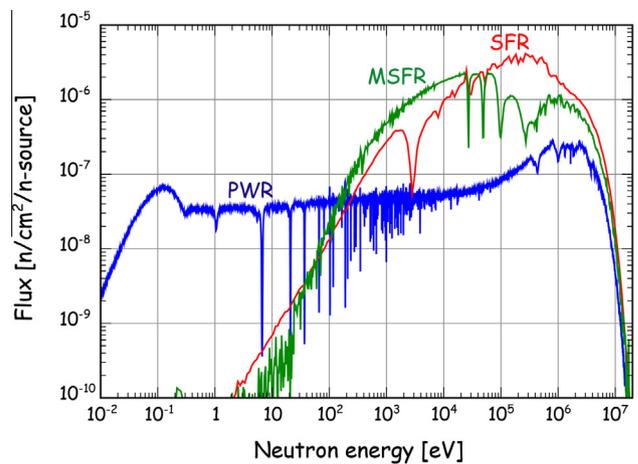


Fig. 2. Fast neutron spectra of the reference MSFR (green curve) and of a sodium-cooled fast neutron reactor (SFR – red curve) compared to the thermalized spectrum of a pressurized water reactor (PWR – blue curve). (For interpretation of color in this figure legend, the reader is referred to the web version of this article.)

area in Fig. 1) to increase the breeding ratio. This blanket is filled with a fertile salt of LiF–ThF₄ with initially 22.5 mol% of ²³²Th. This blanket is surrounded by a 20 cm thick layer of B₄C, which provides protection from the remaining neutrons.

One advantage of a liquid fuel is that its configuration can be modified with no fuel handling, simply by passively draining the fuel salt by gravity into tanks located below the reactor. Two fuel configurations are thus available with no external intervention:

1. the fuel configuration in the reactor, optimized for heat production, corresponding to the critical core;
2. the fuel configuration in the draining tanks, designed to optimize heat and neutron evacuation. This is a sub-critical configuration that allows passive and active heat extraction. In PWR parlance, this is equivalent to a hot shutdown with subsequent removal of the fuel to the fuel building after a few days.

During normal reactor operation, this draining procedure will lead to MSFR shutdown, sub-criticality being reached quickly and easily. In case of an accident or incident leading to a loss of heat sink, the fuel will still be cooled in the draining tanks, the residual decay heat being thus extracted within months.

Fuel salt cleaning (Delpech et al., 2009; Ghetta et al., 2010; Doligez, 2010) involves two processes: (1) the mechanical extraction of rare gases and some noble metals via an on-line bubbling process; (2) the removal of other fission products via batch processing of small fuel salt samples (typical rate ~10–40 l/day) at an on-site facility near the reactor.

2.2. Simulation tools and methodology

Our numerical simulations rely on the coupling of the MCNP neutron transport (Briesmeister, 1997) with a home-made materials evolution code REM (Heuer et al., 2010; Doligez et al., 2009; Nuttin, 2002; Matthieu, 2005).

The probabilistic MCNP code evaluates the neutron flux and the reaction rates in all the parts (called cells) of the simulated system. This requires a precise description of the geometry and the characteristics of all materials involved (temperature, density, elements, isotopes, proportions), together with the interaction cross-sections of each isotope present in the reactor.

¹ For interpretation of color in Figs. 1 and 6, the reader is referred to the web version of this article.

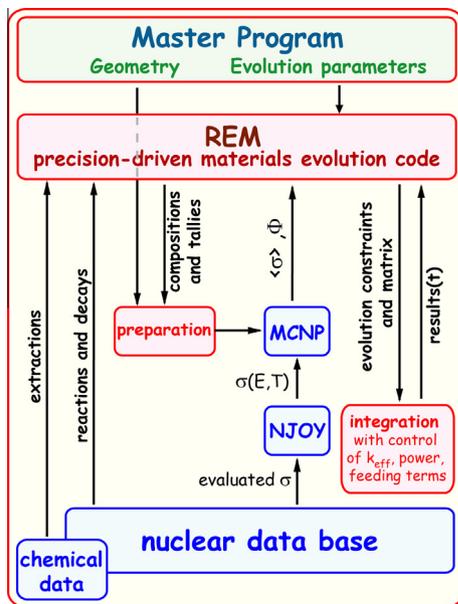


Fig. 3. Coupling scheme of the MCNP neutron transport code with the in-house materials evolution code REM.

These calculations are static, for a given and fixed state of the system. Following the reactor operation over time also requires simulating the temporal evolution of the system. The neutronic code thus has to be coupled with an evolution code, as shown in Fig. 3.

The evolution code, REM, solves the Bateman equations to compute the evolution of the materials composition isotope by isotope within the cells as a function of the nuclear reactions and decays occurring in the system and of external parameters like fuel reprocessing or composition adjustment. These last parameters are implemented through specific removal constants equivalent to decay constants. Our simulations consider several hundreds of nuclei (heavy nuclei, fission products, structural materials etc.) with their neutron reactions and radioactive decays.

The simulations of the reactor's evolution take into account the input parameters (power released, criticality level, chemistry etc.), by continuously adjusting the materials composition and thus the neutron flux of the system, via multiple interactions between the neutronic and the evolution tools. The REM code is indeed a precision-driven code, i.e. it has been designed to determine the reactor evolution while controlling the precision of the results at each step of this evolution. The resolution of the Bateman equations is constrained by several variables to keep the simulated reactor's physical parameters constant during the evolution. These include the total power (with a one percent or so precision) and the reactivity (with a huge precision of some 10 pcm, much smaller than the computational uncertainty of this parameter under MCNP). The numerical integration of the Bateman equations is done using a Runge–Kutta method.

3. MSFR starting modes

To produce power, a fission nuclear reactor requires fissile material. Generation 2 or 3 reactors (PWR, CANDU, EPR etc.) being under-breeder systems, i.e. using more fissile material than they produce, need to be regularly re-fueled with fissile material all along their operation time. On the contrary, breeder generation-4 reactors (SFR, MSFR, GFR etc.) require only one (or two in the case of solid fuel reactors) initial fissile material load. They then produce at least the fissile material they need to be operated during their entire lifespan. Molten salt reactors require only one fissile

load since no fuel re-fabrication is necessary and the fuel salt composition is controlled on-line without stopping reactor operation whereas 2 loads are necessary for solid-fueled reactors with one fissile load inside the reactor while the other one is in the reprocessing/fuel manufacturing process.

The only natural fissile material on earth is ^{235}U (0.72% of natural uranium), which can be used directly as enriched uranium in breeder reactors for their initial fissile load, or which can be loaded in generation 2 or 3 reactors to produce either ^{239}Pu by irradiating ^{238}U , or ^{233}U by irradiating ^{232}Th . To deploy the Thorium fuel cycle in MSFRs, we have thus investigated the following solutions:

- MSFR directly started with ^{233}U as initial fissile material, assuming this ^{233}U may be produced in fertile blankets of other reactors (SFR...) or by irradiating ^{232}Th in an Accelerator Driven System (ADS) for example. Once an initial park of the MSFRs based on the Th– ^{233}U cycle is launched, ^{233}U will also be produced in MSFRs which are breeder reactors, allowing the deployment of such ^{233}U -started MSFRs in a second phase even if no ^{233}U is produced elsewhere.
- Using as initial fissile material the plutonium produced in current PWRs or in future EPRs or, even better, the mix of transuranic elements (TRU) produced by these Generation 2–3 reactors.
- Starting the MSFR with enriched uranium as initial fissile material, with an enrichment ratio less than 20% due to proliferation resistance issues.
- A mix of the previous starting modes. For example, ^{233}U may be produced by using special devices containing Thorium and Pu–Mox in current PWRs or in future EPRs.

Typical configurations of the MSFR corresponding to the different starting modes are detailed in the following paragraphs. The geometry of these MSFR configurations is identical to that presented in Section 2.1.

The fertile salt is always composed of LiF–ThF₄ with 22.5 mol% of heavy nuclei, just as the fuel salt is made of LiF–(HN)F₄ with 22.5 mol% of heavy nuclei among which Thorium as fertile matter.

3.1. ^{233}U started-MSFR

The characteristics of the reference MSFR configuration started directly with ^{233}U as initial fissile material are given in Table 1.

Its initial heavy nuclei inventory per GWe comprises 3.26 tons of ^{233}U and 25.6 tons of ^{232}Th . Fig. 4 (dashed lines) illustrates the evolution of the heavy nuclei inventoried in the fuel salt during the operation of this reactor, up to equilibrium. The proportion of minor actinides in the salt remains low: around one percent at equilibrium.

Regarding safety issues, the feedback coefficient of this configuration, equal to -5 pcm/K, is largely negative and remains stable during the reactor lifespan, as shown in Fig. 5.

Finally, when considering the deployment capabilities of such a MSFR, this configuration corresponds to a breeder reactor producing 95 kg of ^{233}U in excess per year, corresponding to a reactor doubling time of 56 years as shown in Fig. 6 (green line). These values are obtained with the following reprocessing characteristics:

- a simulated batch reprocessing rate of 40 l of fuel salt per day, corresponding to a reprocessing of the whole core in 450 days.
- an on-line bubbling reprocessing with an extraction period of 30 s.

Because of the fast neutron spectrum, the fission product capture cross-sections are small, so that the neutronic characteristics of the reactor, such as the breeding ratio and thus the reactor

doubling time, are only slightly sensitive to the fission product extraction and the cleaning of the salt, as illustrated in Figs. 7 and 8 respectively for the batch and the on-line reprocessing rates.

3.2. MSFR started with transuranic elements

Plutonium and the minor actinides (neptunium, americium and curium) produced in Generation 2–3 reactors may also be used as initial fissile material in a MSFR. This would allow closing the current fuel cycle while launching the Thorium fuel cycle. The mix of minor actinides used as initial fissile load in these calculations is detailed in Table 2 (third column). It corresponds to a UOX fuel after one use in a PWR without multi-recycling, for a burnup of 60 GWd/ton and after 5 years of storage (De Saint Jean et al., 2000). The evolution of the heavy nuclei inventories for a TRU-started MSFR is displayed in Fig. 4 (solid lines).

The utilization of TRU elements to start the reactor increases the initial amounts of minor actinides compared to the ^{233}U -started MSFR. But at equilibrium, the fuel salt compositions of TRU-started and ^{233}U -started MSFRs are identical, the initial TRU being converted into ^{233}U .

Th, Pa, U and Np reach their equilibrium composition quickly, while a few dozen years are necessary to burn 90% of the Pu initial load and around a century for the Am and Cm elements. The in-core Cm inventory reaches a maximum of 390 kg (with 265 kg of ^{244}Cm) after 26 years of operation. After 200 years of operation, as shown in Fig. 4, the Cf inventory is not yet at equilibrium but decreases after a maximum of 900 g (with 8 g of ^{252}Cf) reached after around 110 years of reactor operation.

Note that both statistical and systematic uncertainties are propagated all along the evolution calculations, mainly to the heavier nuclei. The values obtained for the Cm inventory are thus not very precise, while the Cf inventory is known at best by a factor 2.

The deployment capacities of this MSFR configuration are better than those of the ^{233}U -started MSFR with a production of 120 kg of ^{233}U in excess per year during the first 30 years of operation, corresponding to a reactor doubling time of 30 years as shown in Fig. 6.

The limitation of this MSFR starting mode, which is otherwise very promising as far as waste reduction is concerned, lies in the Pu solubility limit in this lithium and thorium fluoride salt. Indeed, solubility issues of valence-3 elements (lanthanides and plutonium) in the fuel salt require special attention. The initial molar proportion of Pu in this configuration is as high as 5.5% (see Fig. 4), corresponding to 12,970 kg. It decreases during reactor operation. 0.2% of valence-3 elements are progressively added due

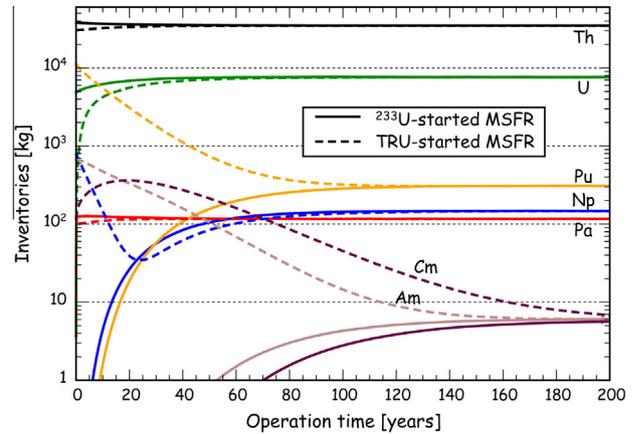


Fig. 4. Time evolution up to equilibrium of the heavy nuclei inventory for the ^{233}U -started MSFR (solid lines) and for the TRU-started MSFR (dashed lines).

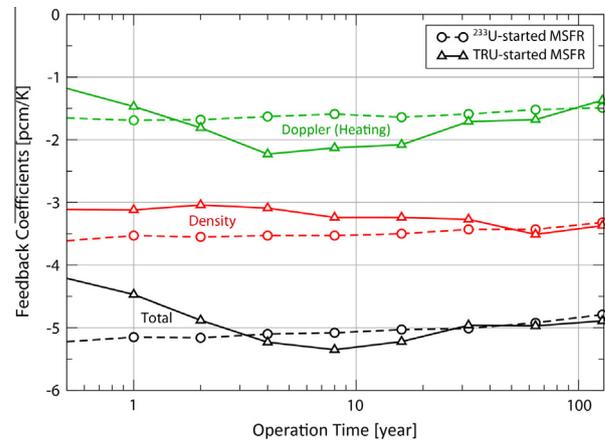


Fig. 5. Evolution of the feedback coefficients during reactor operation for the ^{233}U and TRU-started MSFR.

to the accumulation of lanthanides in the MSFR for the chemical reprocessing rate of 40 l per day chosen here. New measurements made by ROSATOM (Ignatiev et al., 2012) confirm the previous partial solubility data available from experimental measurements performed at BARC in the 1970s (Sood et al., 1975) and listed in

Table 1
Characteristics of the reference ^{233}U -started MSFR.

Thermal/electric power	3000 MW _{th} /1300 MW _e
Fuel salt temperature rise in the core (°C)	100
Fuel molten salt-Initial composition (mol%)	LiF (77.5%)-ThF ₄ (20%)- ^{233}U F ₄ (2.5%)
Initial heavy nuclei inventory per GWe	Th: 25.6 tons ^{233}U : 3.26 tons
Fuel salt melting point (°C)	565
Mean fuel salt temperature (°C)	750
Fuel salt density at 750 °C (g/cm ³)	4.1
Fuel salt thermal expansion coefficient (g cm ⁻³ /°C) (Ignatiev et al., 2012)	8.82×10^{-4}
Fertile blanket salt-Initial composition (mol%)	LiF-ThF ₄ (77.5%-22.5%)
Breeding ratio (steady-state)	1.1
Total feedback coefficient (pcm/K)	-5
Core dimensions (m)	Radius: 1.1275 Height: 2.255
Fuel salt volume (m ³)	18 9 in the core 9 in the external circuits
Blanket salt volume (m ³)	7.3
Total fuel salt cycle in the fuel circuit	3.9 s

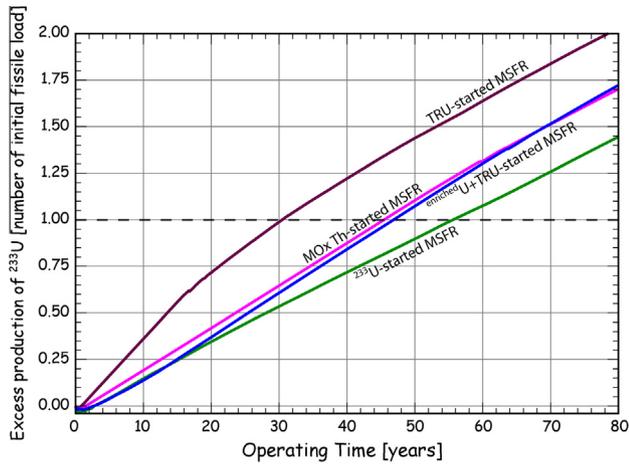


Fig. 6. Excess production of ^{233}U for the different starting modes of the MSFR during reactor operation in number of initial fissile loads produced.

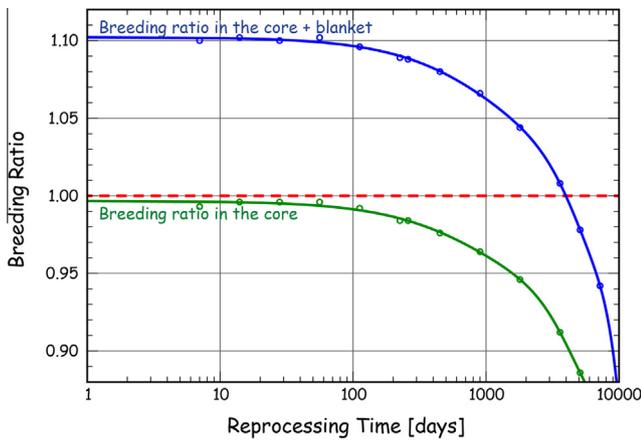


Fig. 7. Influence of the batch reprocessing rate on the breeding ratio in the core and in the whole MSFR system (core + fertile blanket).

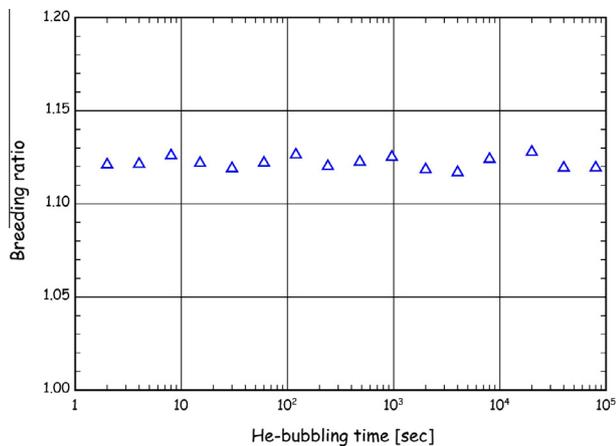


Fig. 8. Influence of the on-line bubbling reprocessing rate on the breeding ratio of the MSFR.

Table 3. According to these data, the initial Pu proportion of the TRU-started MSFR reaches the solubility limit given for a LiF–ThF₄ salt.

Two remedies are available that maintain the solubility within acceptable margins. One consists in increasing the input tempera-

ture of the salt by 50 °C during the first 10 years, but this adds a constraint for the selection of the structural materials. The other solution is to reduce the initial Pu proportion needed to start the MSFR, either by supplementing the initial Pu load with a small amount of ^{233}U , or by starting the MSFR with enriched uranium with a complement of TRU elements. These starting modes are described in the next two sections.

3.3. MSFR started with a mix of ^{233}U and TRU

Here, the initial fissile load is produced by a Th–Pu Mox fuel in an EPR (Heuer et al., 2009). As detailed in Table 3 (fourth column), this results in a mix of different uranium isotopes, mainly ^{233}U , together with TRU elements. The evolution of the heavy nuclei inventories for this MSFR is displayed in Fig. 9 (solid lines). Thanks to the ^{233}U , the molar proportion of Pu in this configuration is down to 4.5%, i.e. below the solubility limit. The maximal amounts of Am, Cm and Cf are higher here compared to the previous TRU-started MSFR, due to the use of Mox fuel. The two configurations are identical after about 20 years, except for Cm and Cf.

The deployment capacities of this MSFR configuration are identical to those of the MSFR started with enriched U and TRU elements, i.e. a reactor doubling time of 45 years for the reprocessing rate considered here.

If UOx fuel from Generation 2 or 3 reactors is still available, another way to start an MSFR is to use this fuel mixed with ^{233}U produced by breeder MSFRs based on another starting mode. This also allows a complete closure of the current fuel cycle. Such MSFRs are quite similar to the MSFRs presented in this section, in terms of heavy nuclei inventories and deployment capacities.

3.4. MSFR started with enriched uranium and TRU

Optimization studies (Guillemin, 2010) bearing on solubility limits, proliferation resistance, initial fissile inventory and breeding capacities of the reactor have singled out an interesting configuration of the MSFR started with TRU elements and enriched uranium. These studies adjust 2 parameters: the uranium enrichment ratio and the proportion of Thorium in the initial fissile load.

The results are presented in Fig. 10 where the configuration at the top of the right axis is equivalent to the configuration started with TRU elements only as described in Section 3.2. The configurations located at abscissa 0 correspond to an initial fuel salt composition without Thorium but with enough TRU elements to reach criticality. The purple line labeled “No TRU” at the bottom left represents different simulations of the MSFR started with enriched uranium only, while the purple line labeled “100%” represents the results for MSFR configurations with pure ^{235}U mixed with Thorium and the amount of TRU elements necessary to reach criticality.

The other solid line curves (labeled 10–30%) show the maximum concentrations in valence-III elements obtained during the reactor operation as a function of the (Th/Th + U) initial ratio for uranium enrichment ratios from 10% to 30%. In the area located on the left of the dotted line labeled “[Pu] stable”, the maximal Pu concentration is reached after a few years of reactor operation, while, for the MSFR configurations located on the right of this line, it decreases during the entire reactor lifetime. These last configurations may raise chemical reprocessing issues that would make their management more complex. The more interesting configurations are placed exactly on the “Pu stable” line, their Pu concentration being stable during the first years of operation to then decrease and reach a Pu concentration equivalent to the ^{233}U -started MSFR at equilibrium (see Fig. 4). Finally, only the configurations located below the line “Solubility limit at 600 °C” (600 °C

Table 2
Initial heavy nuclei inventories per GWe of the different MSFR starting modes.

Starting mode	²³³ U (kg)	TRU (Pu Uox) (kg)	Th Pu–Mox (kg)	enrU + TRU (kg)
Th 232	25,553	20,396	18,301	10,135
Pa 231			20	
U 232			1	
U 233			2,308	
U 234	3,260		317	1,735
U 235			45	
U 236			13	
U 238				
Np 237		531	54	335
Pu 238		229	315	144
Pu 239		3,902	1,390	2,464
Pu 240		1,835	2,643	1,159
Pu 241		917	297	579
Pu 242		577	1,389	364
Am 241		291	1,423	184
Am 243		164	354	104
Cm 244		69	54	44
Cm 245		6		4

Table 3
Plutonium solubility in %PuF₃ for different LiF–ThF₄ compositions (Sood et al., 1975).

LiF–ThF ₄ %	550 °C	650 °C	750 °C
65–35	2.11	4.12	7.08
70–30	2.69	4.91	7.61
75–25	2.75	4.79	7.31
80–20	2.85	4.97	7.71

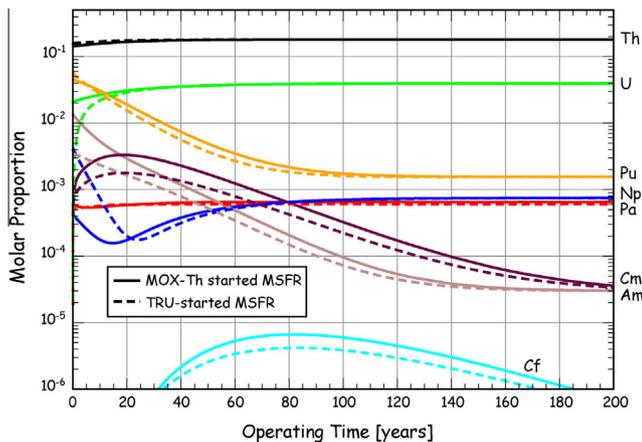


Fig. 9. Time evolution up to equilibrium of the heavy nuclei inventory for the MSFR started with ²³³U and TRU (solid lines) compared to the TRU-started MSFR (dashed lines).

being the coldest point of the salt in the fuel circuit (Merle-Lucotte et al., 2012)) can be considered.

A typical configuration has been chosen to illustrate this MSFR starting mode: the plutonium concentration has been fixed to the estimated solubility limit at 600 °C. The initial fuel salt of this reference configuration also contains 35 mol% of ThF₄ and uranium enriched to 13%.

The calculated evolution of the actinide composition of this fuel salt during reactor operation is displayed in Fig. 11. In this MSFR configuration, the initial Pu concentration is equal to 3.2%, comfortably below the solubility limit. It remains stable during 20 years before decreasing slowly: some Pu is produced from the ²³⁸U initially present in the core.

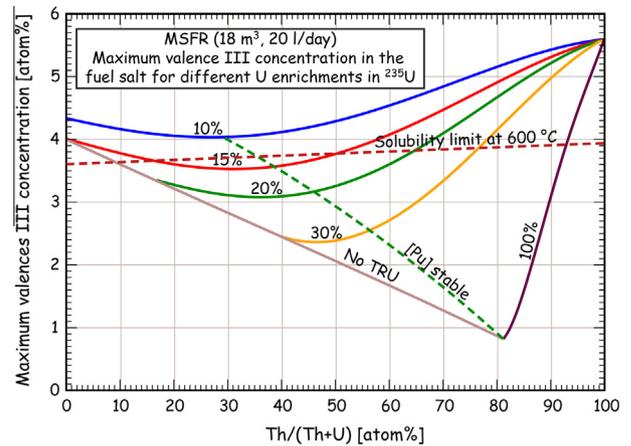


Fig. 10. Maximal concentration in valence-III elements in the initial fuel salt when starting with enriched uranium and TRU elements, as a function of the uranium enrichment and of the initial Th/(Th + U) ratio.

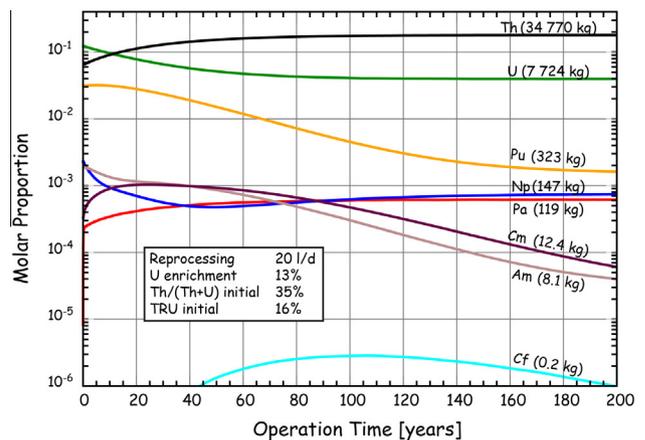


Fig. 11. Time evolution up to equilibrium of the heavy nuclei inventory for the optimized MSFR configuration started with enriched uranium and TRU elements.

The maximal amount of Cm rises to only 210 kg (with 140 kg of ²⁴⁴Cm), while the amount of Cf remains below 600 g with 5.3 g of ²⁵²Cf. This starting mode thus leads to lower TRU concentrations

but they stay in the fuel salt longer. compared to the ^{233}U started MSFR.

The deployment capacities of this MSFR configuration lie between the ^{233}U -started MSFR and the TRU-started MSFR, with a reactor doubling time of 45 years (see Fig. 6).

Regarding safety issues, the uranium enrichment of the initial fuel salt has only a slight impact on the safety coefficients which remain comfortably negative for all the MSFR configurations started with enriched U and TRU, as illustrated in Fig. 12 where the feedback coefficients are evaluated for the initial fuel salt composition.

3.5. Incinerator version of the MSFR

Ultimately when fission based electricity production will be replaced by a novel technology (fusion for instance) all the actinides inside reactors will become discardable wastes. The possibility of eventually shutting down the running reactor parks has to be studied in so-called end-of-game scenarios, the heavy nuclei management being the key issue. If minor actinide losses during reprocessing are less than 0.1% and if the whole fuel salt volume is reprocessed between 1 and 5 years, then the in-core actinide inventory of a reactor is larger than the losses incurred over at least 1000–5000 years of operation. Concerning long-term radiotoxicity issues, finding ways to further reduce the final HN inventory is thus more important than improving the reprocessing efficiency.

We have studied a molten salt reactor used as incinerator to burn (and thus reduce) the final MSFR HN inventories of nearly ten MSFRs within 60 years. This MSR incinerator is identical to the MSFR in terms of system geometry and power production, and differs in the fuel salt composition and the elimination of the fertile blanket.

We have considered a fuel salt made of 46.5 mol% ^7LiF , 11.5% NaF, 41.7% KF and $(\text{HN})\text{F}_4$ whose melting point is sufficiently low even with a small HN proportion (since there is no Th) in the salt and allowing a neutron spectrum that is not too thermalized. The initial heavy nuclei load evaluated to reach criticality is equal to 685 kg of transTh elements (transTh) contained in the final heavy nuclei inventories of the MSFRs presented in the previous paragraphs. The incinerator is also fueled with these transTh final inventories discharged from the MSFRs to maintain reactivity all along the reactor operation, leading to the incineration of 9.4 final MSFR HN inventories as detailed in Table 4. The total burning rate of transTh elements is equal to 9.1, leading to a reduction by one order of magnitude of the long-term radiotoxicity in the period

Table 4
Heavy nuclei inventories before/after incineration.

HN inventory (kg)	Inventories of 9.4 MSFR	Incinerator after 60 years of operation	Burning rate
U	72,751	6 407	11.5
Np	1 381	506	2.8
Pu	2 768	1 530	1.8
Am	72	39	1.8
Cm	33	64	0.5
Total	77,005	8 550	9.1

of 10^3 – 10^6 years (see Fig. 13), mainly thanks to the destruction of the ^{233}U stockpile.

4. Deployment scenarios

Given the absence of naturally available ^{233}U , a standing question is whether a park of MSFRs can be deployed whether at the national, the European or the worldwide scales. In this paper, we illustrate the flexibility of the concept in terms of deployment and end-of-game capacities of the MSFR at the French national scale.

The deployment scenarios of a park of nuclear reactors also led to an estimation of the amount of heavy nuclei produced by such a deployment. We aim at evaluating the complexity of the management of these heavy nuclei stockpiles, as well as their radiotoxicity.

We present here the following French scenario, displayed in Fig. 14: we consider that the natural uranium resources available are large enough to require generation 4 reactors in 2070 only. The deployment scenario starts with the historical French nuclear deployment based on light water reactors (PWRs followed by EPRs) (De Saint Jean et al., 2000; Merle-Lucotte et al., 2006). By 2040, some Generation 3 reactors are fueled with Pu–Uox in a Thorium matrix both to reduce minor actinide production and to prepare the launching of the Thorium fuel cycle in MSFRs. The park of these Generation 3 reactors are then progressively replaced with MSFRs started with this Th–Pu Mox fuel from the last Generation 3 reactors, as described at the beginning of Section 3.4. The deployment is finally completed with MSFRs started with a mix of ^{233}U produced in the existing MSFRs and the remaining stockpiles of Pu–Uox and Pu–Mox irradiated in the light water reactors.

Assuming that, at any time in the future, here in the first half of the XXIInd century, France resolves to dispense from the production of fission based nuclear energy, the scenario ends with the

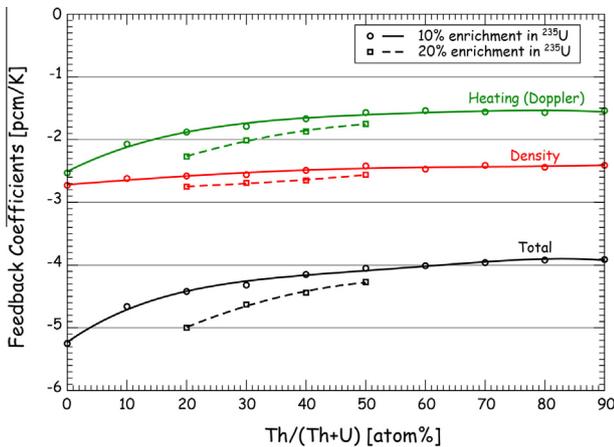


Fig. 12. Feedback coefficients as a function of the U enrichment.

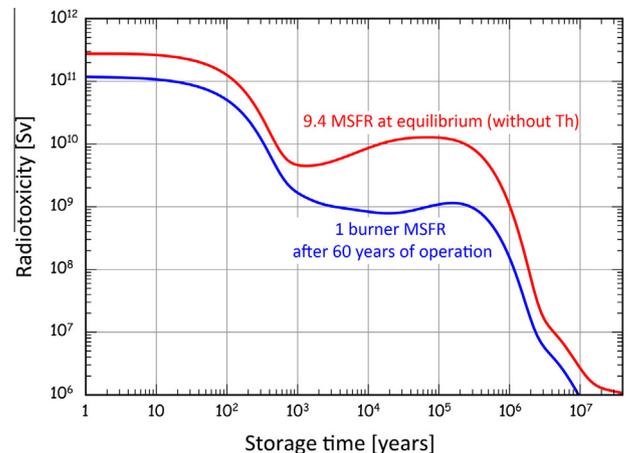


Fig. 13. Time evolution of the radiotoxicity due to final MSFR heavy nuclei inventories with and without final incineration.

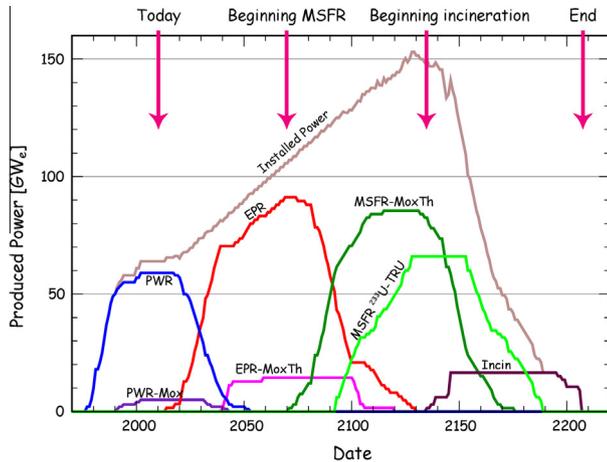


Fig. 14. French nuclear power deployment based on PWRs, EPRs and MSFRs.

introduction of incinerators (as detailed in Section 3.5) with a view to optimizing the end-of-game and further reducing the final TRU inventories after MSFR shutdown. Note that the end-of-game situation would not be different if it occurred after hundreds of years of operation, it depends only on the installed power.

The evolution of the radioactive element stockpiles other than the fission products during the scenario is shown in Fig. 15. The final stockpiles that will have to be managed as the scenario ends are the following:

- Depleted uranium at 0.1%: 803,700 tons.
- Uranium from reprocessing (minimized by the scenario management): 3 250 tons.
- Irradiated Thorium: 5100 tons.
- Irradiated Uox fuel (minimized by the scenario management) represented in Fig. 14 by its Pu content (labeled ‘Pu–Uox’): 5 tons of Pu standing for 450 tons of irradiated Uox.
- Irradiated Mox fuel (minimized by the scenario management) represented in Fig. 14 by its Pu content (labeled ‘Pu + MA Mox’): 0.76 tons standing for 12.4 tons of irradiated Mox.
- Minor actinides separated from the Pu when the latter is used as Mox fuel in light water reactors, and vitrified (labeled ‘MA from Uox’): 612 tons.
- Final incinerator inventories: 106 tons.

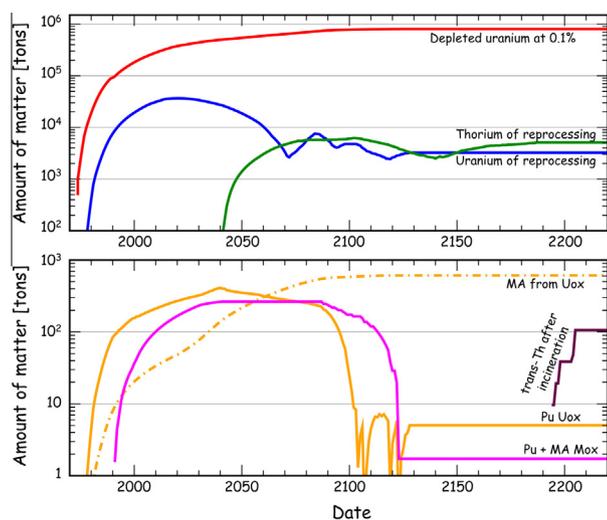


Fig. 15. Evolution of the actinide stockpiles during the scenario considered.

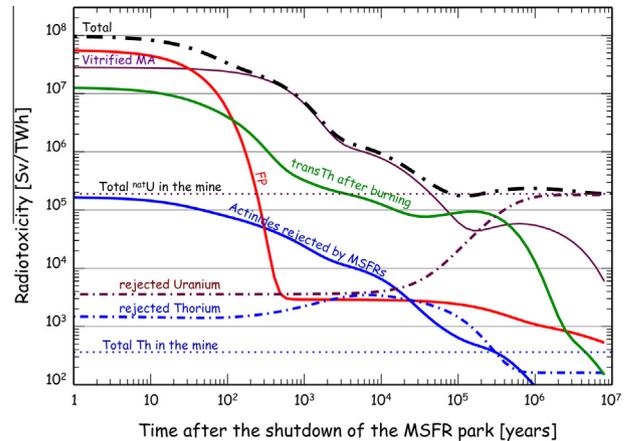


Fig. 16. Time evolution of the various contributions to the radiotoxicity of the final radioactive stockpiles.

The evolution of the radiotoxicity corresponding to the final radioactive stockpiles of this scenario including the fission products is displayed in Fig. 16, where it appears that the short-term radiotoxicity (a few dozen years) is dominated by the fission products (FP) while the long-term radiotoxicity (10^3 – 10^6 years) is mainly due to the vitrified minor actinides produced in light water reactors and not re-used in Mox fuel.

5. Conclusions

In the frame of a major re-evaluation of the molten salt reactor (MSR) concept, and starting from the Oak-Ridge National Laboratory Molten Salt Breeder Reactor project, we have performed parametric studies in terms of safety coefficients, reprocessing requirements and breeding capabilities. Our recent studies have singled out MSR configurations operated with a fast neutron spectrum in the Thorium fuel cycle, the Molten Salt Fast Reactor (MSFR), as robust and very promising. It has been selected for further studies by the MSR steering committee of the Generation IV International Forum.

The standard MSFR is a 3000 MW_{th} reactor with a total fuel salt volume of 18 m³, operated at a fuel temperature of 750 °C. In the MSFR, the liquid fuel processing is part of the reactor where a small side stream of the molten salt is processed for fission product removal and then returned to the reactor. Because of this characteristic, the MSFR can operate with widely varying fuel compositions: its initial fissile load may comprise ²³³U, enriched uranium or also the transuranic elements currently produced by light water reactors.

Our studies show that the MSFR configurations corresponding to various starting modes of the reactor are all characterized by excellent safety coefficients and have the same very good deployment capabilities. Optimizing the specific power in the MSFR configuration started directly with ²³³U as initial fissile material has allowed a reduction of the initial fissile inventory down to 3 metric tons per GWe. The MSFR is characterized by a low proportion of minor actinides in the salt (around one percent at equilibrium) and by its excellent safety coefficients (-5 pcm/°C).

The TRU-started MSFR is able to efficiently convert the plutonium and minor actinides from generation 2–3 reactors into ²³³U while improving the deployment capabilities of the MSFR concept. Its only setback lies in its large initial plutonium concentration, beyond the estimated solubility limit. To overcome this limitation while still using TRU elements in the initial fissile load of the MSFR so as to close the current fuel cycle, we have proposed two

operational alternatives: mixing a lower concentration (around 3–4 mol%) of TRU elements with either ^{233}U produced in other reactors or natural uranium with an enrichment ratio of 13%. Starting from this last composition, initial fuel salt adjustments are being evaluated in the frame of the EVOL project (Euratom/Rosatom collaborative project of FP7) to better take into account the physico-chemical properties of the fuel salt, mainly its melting temperature and solubility limit.

Finally the French nuclear deployment scenario presented here shows that launching the Thorium fuel cycle in Molten Salt Fast Reactors is feasible and efficient while closing the current fuel cycle and optimizing the long-term wastes management by operating the MSFR as an incinerator in the terminal phase.

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