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Reactor physic and reprocessing scheme for innovative molten salt reactor system

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ABSTRACT

The molten salt reactor is one of the six concepts retained by the Generation IV forum in 2001. Based on the MSRE and MSBR concepts developed by ORNL in the 60s which involve a liquid fuel constituted of fluorine molten salt at a temperature close to 600 °C, new developments with innovative approach and technology have been realized which contribute to strongly improve the concept. The thorium breeder potentiality is closely related to the use of a liquid fuel which is able to be periodically treated. A reprocessing scheme has been established to treat used fuel by extraction of fission products. According to the Gen IV philosophy for closed cycle nuclear reactor, the actinides are sent back in the reactor core. In this way, the wastes radiotoxicity is strongly decreased and the use of natural resource is optimized. This paper describes an innovative reactor concept, the TMSR-NM (non-moderated thorium molten salt reactor), from the nuclear physic point of view and the different steps involving in the reprocessing scheme from the chemical point of view.

1. Introduction

The concept of molten salt reactor (MSR) has been developed in the 60s by the Oak Ridge National Laboratory (ORNL) of USA. This type of reactor is well adapted to the thorium fuel cycle (^{232}Th–^{233}U). Indeed with an epithermal neutron spectrum, it can achieve significant breeding gain with a lower minor actinides production as compared to the (^{238}U, ^{239}Pu) fuel cycle) [1]. When the MSRE (molten salt reactor experiment) was designed in 1960, a primary objective was a reactor that would be safe, reliable, and maintainable. The level of success that what achieved is described by Haubenreich and Engel [2]. In particular, it is important to keep in mind that MSRE operated for the equivalent of 9000 full-power hours (during which the reactor was critical 80% of the time) when the reactor was fuelled with ^{235}U and 2500 with ^{233}U fissile fuel. In particular, MSRE is the first reactor to have used ^{233}U as fissile material. The successful operation of the MSRE certainly strengthens confidence in the practicality of the MSR concept.

In parallel, the design of an industrial MSR (MSBR for molten salt breeder reactor) was performed at ORNL [3]. Its objective was to investigate the feasibility of low-cost electricity production with a low specific inventory of fissile material, and simultaneously a reasonably high breeding gain in a molten salt reactor. The main features of this reactor concept were: (1) reactor generating heat in a fuel salt circulating through primary heat exchangers, (2) a fuel salt off-gas purging all gaseous fission products and most particles of noble metal and (3) a chemical process designed to continuously removing fission products from the fuel salt while recovering the bred ^{233}U in order to provide necessary amounts of fissile material. The reprocessing time of the complete fuel was close to 10 days in order to reach a breeding ratio of about 1.06.

MSRE and MSBR were designed to operate in a thermal neutron spectrum. The liquid fuel was circulated through a core of graphite bars which acted as moderator. Studies by Scott and Eatherly [4] showed that an average core power density of 22.2 kW/l (operating condition of MSBR design) would result in a graphite lifetime of approximately 4 years. The reactor design must provide for periodic replacement of the core graphite with minimal plant downtime and complexity of maintenance equipment. The molten salt used was ^{7}LiF–BeF$_2$–ZrF$_4$–UF$_4$ (65–29.1–5.0–0.9 mol%) for MSRE and ^{7}LiF–BeF$_2$–ThF$_4$–UF$_4$ (71.6–16–12–0.4 mol%) for MSBR with ^{233}U as fissile material.

Thus, in our opinion, major drawbacks of both designs were: (1) the use of graphite moderator with a rather short lifespan, (2) a

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reactor fuelled with a non-available natural fissile isotope (^{233}U) and (3) the use of Beryllium which is known for its high chemical toxicity. The TMSR design we are advocating, aims at eliminating these three weaknesses.

This paper aims to describe an innovative concept, the TMSR-NM (non-moderated thorium molten salt reactor) covering the nuclear physics properties of the reactor as well as the various stages of the fuel processing. For two of these stages, the process proposed is a reductive extraction using a liquid metal solvent. Some analytical relations will be given to understand the influence of the liquid solvent composition on the extraction efficiency.

2. Reactor physics

2.1. Simulation procedure

The calculations shown below rely on MCNP neutron transport code [5] coupled with a home-made material evolution code REM [6]. While MCNP at a given fixed time intervals evaluates the neutron flux and the reaction rates in all the cells associated with the discretization of the core design, REM solves the Bateman equations and provides the evolution over each time step of all material (fuel and structure) within the MCNP cells. These calculations take into account the reactor parameters (power, criticality level, chemistry, . . .) by a continuous adjustment of the neutron flux and of the composition of the core. The calculation relies on an accurate description of the core geometry and follows several hundreds of nuclei along with their interactions with the neutron flux and their radioactive decays.

2.2. The non-moderated thorium molten salt reactor

The CNRS-Grenoble in France has proposed an innovative concept called TMSR-NM for non-moderated thorium molten salt reactor [6–9]. This concept is the final outcome of an extensive set of parametric studies of MSR in which various core arrangement and composition, reprocessing performances and salt compositions were investigated. The mean feature of this concept is the removal of the graphite moderator leading to a “close to fast” neutron spectrum. Due to the small moderator effect of fluoride molten salt, the reactor can operate both as a thorium breeder and an actinide burner. This reactor can thus be fuelled either with ^{233}U (U-started TMSR) or with a mix of transuranic (TRU) elements (Pu, Np, Am and Cm) such as the one in spent UOX fuel (one use in a standard light water reactor followed by five years of storage) [10]. This later operation mode will be called TRU-started TMSR. It is then possible to start a first generation of TMSRs from a fuel extracted from the spent fuel of PWRs whose actinides are the needed fissile material. Then, the breeder capacity of TMSR within the same basic design allows the start of a second generation of TMSR fuelled with ^{233}U produced in the Th blankets of the first generation. This scenario removes the concern of the non-availability of ^{233}U while strongly reducing the radiotoxicity of the PWR spent fuels. Moreover, in the TMSR, the molten salt chosen being a mixture of only LiF and ThF_4 removes the undesirable BeF_2 compound.

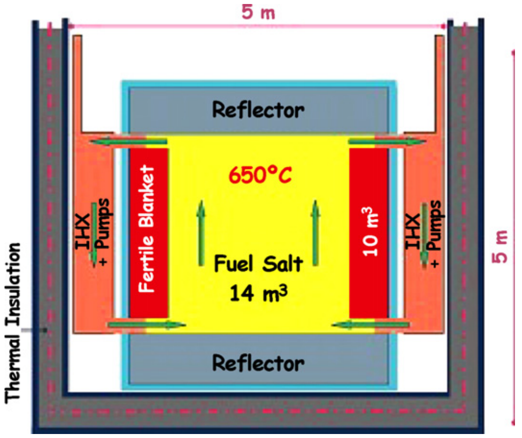

Fig. 1. Schematic vertical section of the TMSR, including pumps and heat exchangers.

2.3. Neutronic core description

Table 1 presents some characteristics of TMSR-NM while Fig. 1 displays a schematic drawing of a vertical section of the reactor.

The core structures are protected by reflectors which ensure that 80% of the neutron flux is either reflected or absorbed. To avoid thermalization of the reflected neutrons, the axial reflectors are

Table 1
Properties and characteristics of TMSR-NM

Thermal power (MWth)	2500				
Fuel molten salt composition (mol%)	LiF–ThF_4–UF_4 or LiF–ThF_4-(Pu–MA)F_3 with LiF = 0.77.5				
Fertile Blanket Molten salt composition (mol%)	LiF–ThF_4 (72–28)				
Melting point (°C)	550				
Operating temperature (°C)	630				
Initial inventory (kg)	U-started TMSR		TRU-started TMSR		
	Th	^{233}U	Th	Actinide	
	46100	5700	37040	Pu	13000
				Np	900
				Am	800
				Cm	130
Density (g/cm^3) [11]	4.3				
Dilatation coefficient (/°C)	10^{-3}				
Core dimensions (m)	Radius: 1.25 Height: 2.60				
Fuel salt volume (m^3)	20.5 6.5 out of the core 14 in the core				
Blanket Salt Volume (m^3)	9				
Thorium consumption (ton/year)	1				
^{233}U production (kg/year)	120 (^{233}U-started TMSR) 173 during 30 years then 120 (TRU-started TMSR)				
Breeding ration (^{233}U-started TMSR)	1.12				

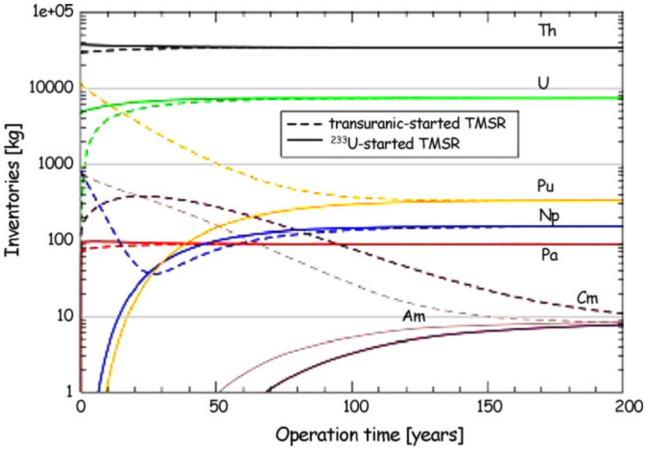

Fig. 2. Heavy nuclei inventory for the ^{233}U-started TMSR (solid lines) and for the TRU-started TMSR (dashed lines).

made of ZrC. In the radial reflector, tubular structures contain binary fluoride salt with thorium (Table 1). Thus, this reflector acts as a fertile blanket to increase the breeding ratio.

These calculations assume a reprocessing efficiency of the fuel salt done in two steps. First, an on-line gaseous extraction with helium bubbling removes all gaseous fission products and noble metals. In our simulations, the extraction time of these elements is assumed to be 30 s. As a matter of fact, lower extraction efficiency would not significantly affect the neutronic of the core. Indeed, the breeding ratio is almost unaffected if these 30 s become a few days. For the removal of most of the fission products, an off-line reprocessing is required. Thus, periodically, batches of salt have to be removed and a succession of chemical treatments effected. At the end, all the actinides (U, Pa, Np, Pu, Am, . . .) are extracted and sent back to the reactor core to be consumed, while lanthanides are sent to the waste.

Core calculations presented here have been made considering that 100 kg of heavy nuclei are reprocessed per day and sent back into the core. This corresponds to a daily reprocessing volume of 40 l of fuel salt.

Reactor deterministic safety is good since the feedback coefficients of the TMSR-NM are negative in both ^{233}U and TRU operating modes [12]. For instance, the total feedback coefficient is equal to -5.7 pcm/°C when the equilibrium state of the ^{233}U mode has been reached. The density coefficient, which for MSRs can be viewed as a void coefficient, is also negative at about -2.7 pcm/°C.

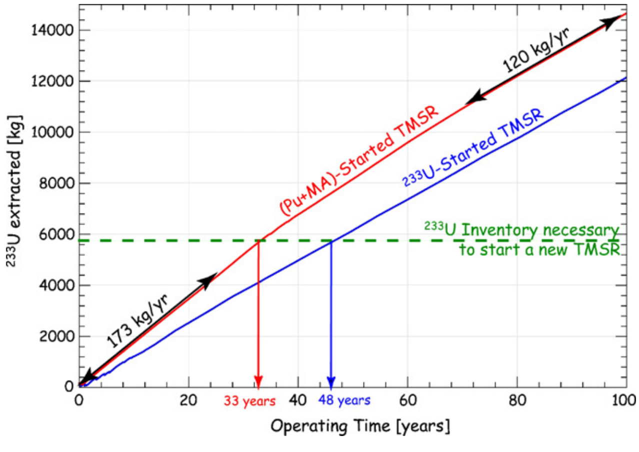

Fig. 3. ^{233}U extracted during TMSR-NM operation time for the two configurations: U-started TMSR and TRU-started.

2.4. TMSR-NM as an actinide burner

The evolution of a typical fuel salt composition for a ^{233}U-started TMSR and TRU-started TMSR (fuelled with transuranic elements) in operation is shown in Fig. 2. The inventories of U and Th in the two reactors becomes equivalent after about forty years in operation. At this time, more than 85% of the initial TRU inventory is burned.

2.5. TMSR-NM as a thorium breeder

The deployment capacity of a given reactor depends on its capacity to generate the amount of fissile fuel (here ^{233}U) necessary to start another reactor of the same type. A good indicator is the doubling time defined by the operation time which through breeding leads to the ^{233}U inventory of a new reactor. The amount of ^{233}U produced and extracted as a function of operation time is given in Fig. 3. For a ^{233}U-TMSR, the annual ^{233}U production is 120 kg which corresponds to 48 years doubling time. Starting a TMSR from Generation II or III reactors spent fuel is more favourable and yields 33 years doubling time. Indeed, the presence of other fissile elements decreases the consumption of ^{233}U.

3. Chemistry of the reprocessing

3.1. Chemical data

The viability of a reactor concept, in terms of both safety and economy, depends on the lifetime of its components and on constraints imposed on its operation mode. For a MSR, they strongly rely on the quality of the chemical processing of the fluorine molten salt (measured both by its extraction efficiency and the amount of waste generated during the process) and on the compatibility of the salt with the reactor (including the heat exchanger) structural materials. Concerning this last point, we have already noted that in the TMSR-NM concept the removal of the graphite moderator automatically improves the performance of the reactor.

As far as reprocessing is concerned, a prior requirement is an accurate knowledge of the physico-chemistry of the molten salt including the redox and chemical properties of the various solutes (heavy elements, fission products). Then, based on this knowledge, a pyrochemical reprocessing scheme must be designed which allows a separation of actinides and fission products within a reasonably short time.

Pyrochemical separation and extraction of actinides and fission products can be performed in different ways. For instance, some methods (Table 2) are based on modifications of oxidation states of the elements either by the action of selected oxidizing or reducing reagents or by the use of electrical current. In several cases, the oxidation leads to the formation of a gaseous halide compound (e.g. UF_6 or NpF_6 for fluoride media). The reduction reaction generally leads to the formation of a metal or an alloy. When metals are obtained by electrochemical reduction on solid and inert cathodes, they are in a solid state. When the reduction is performed on a liquid metal cathode (e.g. Bi, Ga, Al, Sb, Cd, . . .), either by electrochemical reduction or by chemical reduction with a reducing reagent (such as Li and Th) dissolved into the liquid solvent metal, the extracted metallic element is dissolved within the liquid solvent metal. Other methods use the oxoacido-basic properties (oxide anion exchange power) of the solutes. In this case, the control of the amount of oxide ions in melt leads to a selective precipitation of an oxide which has to be subsequently separated from the liquid media.

Both thermodynamic and kinetic aspects have to be considered to evaluate a reprocessing scheme. From fundamental data (redox potentials, activity coefficients [13–15]) determined either by

Table 2
Possible routes for the selective separation/extraction of solutes from a molten salt

REAGENT STATE	INVOLVED PROPERTIES	METHODS	PRODUCT STATE
SOLUTE (in fluoride or chloride media) (M^{n+})	(A) REDOX (oxidation state modification)	(A1) Action of an oxidizing agent F_2, Cl_2	GAS (MF_x)
		(A2) Action of an oxidizing electrical current	
		(A3) Action of a reducing agent, e.g. Li	SOLID METAL (on solid cathode) OR METALLIC SOLUTE (in liquid metal solvant)
		(A4) Action of a reducing electrical current	
	(B) ACIDO-BASICITY (oxide precipitation solubilization)	(B1) Action of oxide ions donors (e.g. Li_2O)	Solid (MO_x)
		(B2) Action of gaseous mixtures $H_2O/HF/H_2$	

experimental measurements or by molecular dynamic simulations, the efficiencies can be calculated for each reaction in its equilibrium state. Such types of calculations and the analytical equations they rely on have already been described [13–17].

The aim of this paper is to present the first chemical scheme established for the reprocessing of the molten salt of TMSR-NM concept. Some of these steps that it includes have been already proposed within the scheme of the ORNL–MSBR project. However, the nature of the molten salt generates a major difference between the two concepts. Indeed, the salt mixture of MSBR, which has fully been deal with references [18–20], contains BeF_2. This compound strongly affects the chemistry of the salt as well as its redox properties. On the other hand, for the TMSR design, the salt is essentially constituted of LiF and ThF_4. The properties of this salt have not yet been much studied so that publications or data are still scarce. The reprocessing scheme we discuss below is based on the results of a preliminary electrochemical study [21,22].

In addition, a major difference between the US concept and the one discussed in this work is the required reprocessing rate. In the MSBR concept, breeding conditions could only be reached when the reprocessing time of the entire core was less than 10 days. Despite the small size of the MSBR core, this translated into a daily treatment of about 4000 l of molten salt.

For the TMSR-NM concept, the reprocessing rate is of the order of 40 l per day. Obviously, this reduction by two orders of magnitude strongly facilitates the design of a process.

3.2. *The reprocessing scheme of TMSR-NM concept*

The complete reprocessing scheme proposed for the treatment of the liquid fuel of TMSR-NM is depicted in Fig. 4. Note that when operated in the TRU mode, the salt will contain high quantities of plutonium. This scheme should thus be considered as a framework where the extraction efficiencies of each separation step have to be further studied and their integration into a single process optimized.

Moreover, we should not only consider the fuel carrying salt. Indeed, since TMSR is intended to be a breeder, the reprocessing must also take into account the salt in the blanket from which the fissile ^{233}U isotope has to be extracted. For this salt component in which the fertile element Th dominates, the required reprocessing rate is also equal to 40 l per day.

3.2.1. *Helium bubbling*

As mentioned in Section 1, the treatment of the salt is effected in two steps. First, there is an on-line gaseous extraction with helium bubbling to remove gaseous fission products, Xe and Kr, and noble metals by a flotation process. The present knowledge on the bubbling salt cleaning is based on the experimental feedback of the MSRE [20]. There, it was demonstrated that helium bubbling ensured the elimination of the rare gases, while removing also a significant fraction of the noble metals. The remaining fraction accumulated in the reactor depending on the salt circulation mode, the circuit geometry (etc.) is difficult to be predicted. A technical

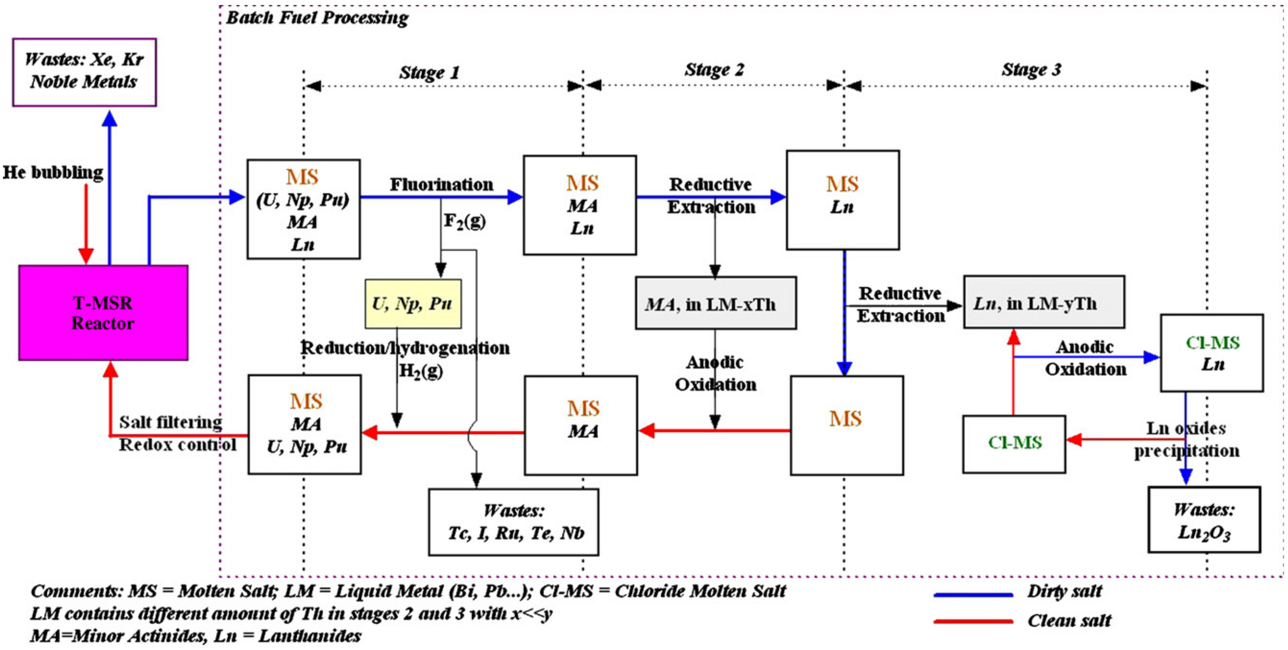

Fig. 4. Reprocessing scheme for TMSR-NM innovative concept.

Table 3
Balance of the first off-line reprocessing stage

Production of gaseous F_2 and H_2 by HF decomposition	$2HF = F_2\ (g) + H_2\ (g)$
Fluorination of element M (M = U, Np, Pu, Te, Tc, Nb, Cr, Mo, Ru, I)	$MF_{(y-x)} + x/2F_2(g) = MF_y(g)$
Hydrogenation	$MF_y(g) + x/2H_2(g) = UF_{(y-x)}$ (in the molten salt) + xHF
Balance	0 = 0

Table 4
Balance of the second off-line reprocessing stage

Reductive extraction of actinides	$4/3AcF_3 + Th(Bi) = ThF_4 + 4/3Ac(Bi)$
Anodic oxidation of metallic actinides cathodic reduction of ThF_4 into Th	$Ac(Bi) + 3F^- = AcF_3 + 3e\ ThF_4 + 4e = Th + 4F^-$
Balance	0 = 0

improvement of the bubbling had been foreseen for the industrial prototype (MSBR). This improvement used efficient technical solutions but was not based on the identification of the physicochemical processes involving in the flotation process.

3.2.2. *Off-line reprocessing*

The second phase is an off-line reprocessing line in which 40 l per day of fuel salt have to be treated. This phase can be divided in three stages.

3.2.2.1. Stage 1: Fluorination reaction. This step aims at removing the elements with high gaseous oxidation states, such as U, Np, Pu for the actinides and Nb, Ru, Te, I, Mo, Cr, Tc for the fission products. The global reaction is given in Table 3.

Three successive NaF traps are used at different temperatures to separate and recover the different elements. The rate of recovery of UF_6 is 99.9% and the decontamination factors for Ru, Nb and Te are higher than 10^5 [20].

Concerning Pu, it has been demonstrated that extraction by fluorination is possible when small (100 μm) liquid salt droplets fall in a 50% F_2 atmosphere at 550/650 °C [23]. Up to 90% Pu was removed from the salt falling in a 1.3 m high fluorinator and a better efficiency is expected at higher temperature and smaller drop diameter. In ref. [23], the Pu extraction which requested solid salt crushing was not practical. Nowadays, granulation in the same diameter range has reached industrial stage for high temperature liquids. This technology can be adapted to the formation of molten fluoride droplets falling in an almost stagnant fluorine atmosphere.

The separated elements are next reduced by action of hydrogen. The balance of this first stage is given in Table 3.

3.2.2.2. Stage 2: Actinide extraction. The remaining actinides (Pu (the fraction not removed by fluorination stage), Pa, Am, Cm) are now extracted from this batch by a reductive extraction in a liquid metal solvent constituted of Bi with metallic thorium. The metallic thorium in the bismuth pool is here the reductive reagent. Its concentration is chosen to control the potential of the metallic pool. The potential is given by the following relation:

$$\mathrm{E}_{\mathrm{Bi-Th}} = E^{\circ}{}_{\mathrm{ThF_4/Th}} + 2.3\frac{RT}{4F}\log\frac{a(\mathrm{ThF_4})_{\mathrm{MS}}}{a(\mathrm{Th})_{\mathrm{LM}}} \quad (\mathrm{R1})$$

In this relation, E° is the standard potential of the redox system ThF_4/Th, while the $a(i)$ are the activities in the molten salt (MS) and in the liquid metallic solvent (LM). R is the ideal gas constant ($8.32\ \mathrm{J\ mol^{-1}\ K^{-1}}$), T the temperature (°K) and F the Faraday constant (96500 C). These activities are themselves related to the mole fraction $x(i)$ by $a(i) = x(i) \times \gamma(i)$ in which $\gamma(i)$ is the activity coefficient.

The value of the potential is one key experimental parameter because its control is crucial to optimize the decontamination factor of actinides. Indeed, it must allow a reduction of actinides while leaving lanthanides in the salt.

The elements involved in this extraction are Pu, Pa, Am and Cm. Fission products such as Zr can also be reduced by this step if they are not removed in the on-line reprocessing line of helium bubbling.

The back extraction is realized by anodic oxidation in cleaned LiF–ThF_4 molten salt of the Bi pool which contains the actinides. At the anode, the oxidation of actinides occurs while the reduction of ThF_4 into metallic Th occurs at the cathode, the cathodic limit in LiF–ThF_4 molten salt being the reduction of ThF_4 into Th. The balance of the extraction/back extraction of actinides is given in Table 4. The actinides extracted in the cleaned salt are sent back in the reactor core.

3.2.2.3. Stage 3: Lanthanide extraction. The extraction of lanthanides is also performed by a reductive extraction in liquid Bi pool containing metallic thorium as reductive reagent. The difference with the previous stage is due to a modification of the metallic pool composition by changing the concentration of thorium which modifies the potential value or by increasing the volume of the metallic phase. In this case, the concentration of metallic thorium is kept higher than the concentration used for the actinide extraction in order to sufficiently lower the potential to values which correspond to the reduction of the lanthanides dissolved in the molten salt. The maximum of thorium concentration in bismuth is close to its solubility [20].

The back extraction of lanthanides from the Bi–Th bath is done by anodic oxidation in LiCl–KCl (58.5–41.2 mol%) chloride medium at 500 °C. In this case, at the cathode, the molten salt is reduced into metallic Li which is separated and recovered. Simultaneously, the lanthanides are oxidized and dissolved in the chloride medium into chloride species. The separation is then realized using their acido-basic properties. H_2O which partial pressure is controlled and introduced through argon flux is used as the oxide donor. The reaction with the chloride species yields HCl(g). The advantage of using a gaseous reagent is that the formation of a gaseous product does not increase the volume of effluents. The chlorydric acid is then used to oxidize the metallic lithium produced from the back extraction of lanthanides, leading to LiCl and H_2(g). The salt LiCl is sent back in the LiCl–KCl molten salt to restore its initial composition while hydrogen, introduced in clean LiF–ThF_4, reduces the excess of ThF_4 resulting from the reductive extraction of lanthanides. All these reactions and the balance of the third stage are listed in Table 5.

3.2.3. *Potential control of the solvent metal*

As mentioned in the last two stages of the salt processing, the potential of the Bi–Th solvent metal is controlled by the amount of Th in the liquid bismuth by the relation (R1). Via the Nernst relation, this concentration determines the equilibrium ratio of

Table 5
Balance of the third off-line reprocessing stage

Reductive extraction by Th	$4/3LnF_3 + Th(Bi) = ThF_4 + 4/3Ln(Bi)$
Anodic oxidation of Ln cathodic reduction of LiCl into Li	$Ln(Bi) + 3Cl^- = LnCl_3 + 3e$ $LiCl + 1e = Li + Cl^-$
Precipitation of oxide by action of H_2O	$2LnCl_3 + 3H_2O(g) = 6HCl(g) + Ln_2O_3$
Oxidation of metallic Li by HCl	$Li + HCl(g) = LiCl + 1/2H_2(g)$
Reduction of ThF_4 by hydrogen	$ThF_4 + 2H_2(g) = Th(Bi) + 4HF$
Balance	$4/3\ LnF_3 + 2H_2O(g) = 2/3\ Ln_2O_3 + 4HF$

Table 6
Data used to calculate the extraction efficiencies of Pu and Nd (Fig. 5)

N_{LM}/n_{MS}	$\log\gamma\ (NdF_3)_{MS}$	$\log\gamma\ (PuF_3)_{MS}$	$\log\gamma\ (ThF_3)_{MS}$	$E^\circ\ (NdF_3/Nd)$ (V)	$E^\circ\ (PuF_3/Pu)$ (V)	$E^\circ\ (ThF_4/Th)$ (V)
1	1	1	−3 [21,24]	−5.02	−4.63	−4.7
	$\log\gamma\ (Nd)_{LM}$	$\log\gamma\ (Pu)_{LM}$	$\log\gamma\ (Th)_{LM}$			
	−10.1 [25]	−7.5 [25]	−6.7 [25]			

concentrations of the element M in the metal and in the molten salt. "MF" designs the element M in the salt phase and "M" the element M in the metallic phase.

$$E_{LM} = E^\circ{}_{MF_x/M} + \frac{2.3RT}{xF}\log\frac{a(MF_x)_{MS}}{a(M)_{LM}} = E^\circ{}_{MF_x/M} + \frac{2.3RT}{xF}\log\frac{x(MF_x)_{MS}\gamma(MF_x)_{MS}}{x(M)_{LM}\gamma(M)_{LM}} \tag{R2}$$

In relation (R2), $a(i)$, $x(i)$ and $\gamma(i)$ are respectively the activity, the mole fraction and the activity coefficient of i in the molten salt (MS) and in the liquid metal (LM). x represents the number of electrons exchanged in the reduction of M. $E^\circ{}_{MF/M}$, R, T and F have already been defined in the previous section.

The extraction efficiency of the element M is given by the following relation:

$$\mathrm{EFF(M)} = \frac{n(M)_{LM}}{n(M)_i} = \frac{n(M)_{LM}}{n(M)_{LM} + n(MF)_{MS}} = \frac{x(M)_{LM} \times n_{LM}}{x(M)_{LM} \times n_{LM} + x(MF)_{MS} \times n_{MS}} \tag{R3}$$

In the relation (R3), n_{MS} and n_{LM} are, respectively, the total number of moles of the salt phase and of the metallic phase. $n(MF)_{MS}$ and $n(M)_{LM}$ are the number of mole of M in the salt phase and in the liquid metal phase at equilibrium. $n(M)_i$ is the total number of mole of the element M initially present in the molten salt before the extraction.

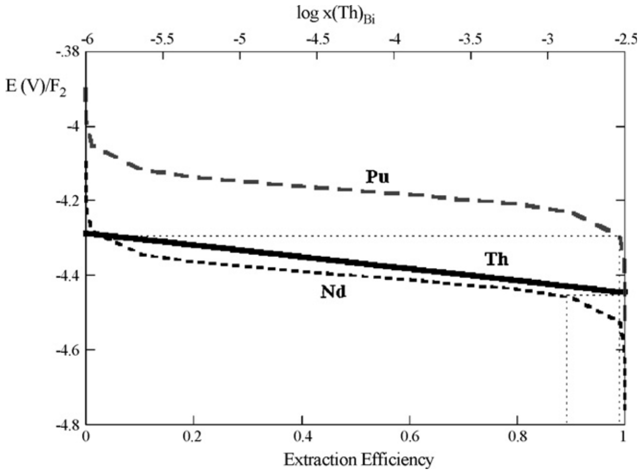

Fig. 5. Variation of extraction efficiency of Pu and Nd as a function of liquid Bi–Th potential and variation of Bi–Th pool as a function of the concentration of Th in Bi.

A relation between the potential and the efficiency can then be deduced from the relations (R2) and (R3):

$$E_{LM} = E^\circ{}_{MF_n/M} + \frac{2.3RT}{xF} \times \left(\log\frac{n_{LM}}{n_{MS}} + \log\frac{(1-\mathrm{EFF(M)})}{\mathrm{EFF(M)}} + \log\frac{\gamma(MF_x)_{MS}}{\gamma(M)_{LM}}\right) \tag{R4}$$

Fig. 5 presents an example of potential variation (given against the usual reference system $F_2(g)(1\ \mathrm{atm})/F^-(a = 1)$) as a function of efficiency calculated for the elements NdF_3 and PuF_3 for the numerical values given in Table 6. In the same figure, the potential variation of the solvent metal as a function of the amount of Th in the metallic phase is calculated using the relation (R1). The solvent metal considered for the calculation is Bi and the molten salt is LiF–ThF_4 (78–22 mol%) at 650 °C. The standard potentials are calculated using the thermochemical software HCS Chemistry version 4.1, Outokumpu Research, Finland. The reference system is the molar fraction scale, with pure solid as reference state.

For the 3.2.2.2 stage, one can work with a Th mole fraction in Bi equal to 10^{-6}. Then, from Fig. 5, the potential of the liquid pool is −4.28 V so that the extraction efficiency of Pu is of 99% and that of Nd only 1%. For lower concentrations of Th (and higher potential values), the extraction efficiency of Pu decreases. In this case, the extraction of large fraction of Pu would require several extraction stages.

The extraction of Nd remaining in the reprocessed salt is then performed via the 3.2.2.3 stage of the scheme. In this case, a lower value of potential is needed. This is achieved by increasing the concentration of Th in the liquid Bi. For a concentration of Th equal to its solubility in Bi, the extraction efficiency of Nd is equal to 87%. To extract a larger quantity of Nd, a multi-stage extraction is necessary. A best efficiency can also be obtained by increasing the mole ratio n_{LM}/n_{MS} [22].

4. Conclusion

The innovative TMSR-NM concept proposed appears to be a very promising MSR concept. It has several specific advantages, mainly due to the liquid fuel and to the thorium cycle. Among the properties not yet mentioned, one can note that the LiF–ThF_4 liquid fuel is very stable under irradiation. As in all MSR, it removes the question associated with the definition of a solid fuel fulfilling all the safety requirements for use in a cladding. On the other hand, the use of a liquid fuel allows an on-line fuel control. The amounts of fissile and fertile elements can be adjusted without unloading the core, eliminating the need for any initial reactivity reserve.

The reprocessing scheme that we propose as reference is both comprehensive and compatible with the volume requirements for the TMSR. It allows a detailed study of all fuel and waste streams.

Obviously, it is still a starting point which needs further exploration and improvements. We should keep in mind that the nature of the liquid metal can still be modified to cope with the low solubility of Th in Bi. The choice of the solvent metal in stages 3.2.2.2 and 3.2.2.3 (here above Bi) has to be optimized considering the several extraction stages. This optimization will take into account the extraction efficiencies for the all elements, particularly those of lanthanides which are neutronic poisons. A more accurate knowledge than presently available of fundamental data, such as activity coefficients, is essential to realize this study successfully. Experimental measurements are currently underway.

All the properties of TMSR-NM combined with an integrated chemical reprocessing scheme put this innovative molten salt reactor concept in a very good position to fulfil the conditions defined by the GEN IV International Forum and to produce the large amount of nuclear energy that the world will need in the future.

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