Proceedings of ICAPP '09
Tokyo, Japan, May 10-14, 2009
Paper 9xxx

Innovative nuclear system based on liquid fuel

S. Delpech[*a], S. Jaskierowicz[a], G. Picard[a], E. Merle-Lucotte[b], D. Heuer[b], X. Doligez[b]
a: LECIME/INC/CNRS/ENSCP –
11 rue Pierre et Marie Curie, 75231 Paris Cedex 05 France
b- LPSC/IN2P3/CNRS
53 avenue des Martyrs, F-38026 Grenoble Cedex France
Tel: 33 1 55 42 63 91; email: sylvie-delpech@enscp.fr

Abstract – *The aim of this paper is to present the physical properties and characteristics of the innovative concept of Molten Salt Fast Reactor (MSFR) developed by CNRS (France) and the corresponding fuel salt reprocessing proposed to clean up the fuel salt based on an analytical approach of lanthanides and actinides extraction.*

I. INTRODUCTION

With the particularity of using a liquid fuel, the MSFR (Molten Salt Fast Reactor) concept has been developed. This reactor operates in simplified and safe conditions in the Th/233U fuel cycle with fluoride salts. The major improvement of this concept, compared to the ORNL molten salt reactor developed in the 1960s [1-3], is a fast neutron spectrum obtained by removing graphite and BeF_2.

This paper aims to describe an innovative concept, the MSFR (previously designed as TMSR-NM, Thorium Molten Salt Reactor Non Moderated) and developed by CNRS-Grenoble in France [4-7] 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.

II. REACTOR PHYSICS

The neutronic calculations shown below rely on MCNP neutron transport code [8] coupled with a home-made material evolution code REM [5,9]. The former evaluates the neutron flux and the reaction rates in all the cells while the latter solves the Bateman equation for the evolution of the material composition within the cells.

A positive breeding gain is largely reached with a reprocessing rate of 40 liters of molten salt per day. The reactor is started with ^{233}U or with a Pu and minor actinides (MA) mixture coming from PWR spent fuel. The MA consumption with burn-up demonstrates the burner capability of MSFR. Some characteristics of this concept are summarized in Table 1.

TABLE 1:
Properties and characteristics of MSFR

Thermal power (MWth)	2500		
Fuel Molten salt composition (mol%)	LiF-ThF_4-UF_4 or LiF-ThF_4-$(Pu\text{-}MA)F_3$ with $LiF = 0.77$		
Fertile Blanket Molten salt composition (mol%)	LiF-ThF_4 (72-28)		
Melting/Operating temperature (°C)	550/630		
Initial inventory (kg)	**U-started MSFR**		
	Th	^{233}U	
	46100	5700	
	TRU-started MSFR		
	Th	Actinide	
	37040	Pu	13000
		Np	900
		Am	800
		Cm	130
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 MSFR) 173 during 30 years then 120 (TRU-started MSFR)		
Breeding ratio (^{233}U-started TMSR)	1.12		
Reflector	Radial: fertile blanket Axial: Nickel based alloy		

The external core structures and the heat exchangers are protected by thick reflectors made of nickel-based alloys which have been designed to absorb more than 80% of the escaping neutron flux. These reflectors are themselves surrounded by a 10cm thick neutronic protection of B_4C which absorbs the remaining neutrons. The radial reflector consists in a fertile blanket (50 cm thick) filled with a fertile salt of LiF-ThF_4 with 22.5%-mole ^{232}Th. Thanks to ^{233}U extraction every six months, this fertile blanket improves the total breeding ratio of the reactor.

The evolution of a typical fuel salt composition [5,10] for a ^{233}U-started MSFR and TRU-started MSFR (fuelled with transuranic elements) in operation shows that 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.

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 Fig.3 [5]. For a ^{233}U-MSFR, the annual ^{233}U production is 120kg which corresponds to 48 years doubling time. Starting a MSFR 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.

Reactor deterministic safety is good since the feedback coefficients of the MSFR-NM are negative in both ^{233}U and TRU operating modes [5,11]. For instance, the total feedback coefficient is equal to -6 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 -3 pcm/°C.

III. CHEMISTRY OF THE REPROCESSING

Due to the liquid fuel, the amounts of fissile and fertile elements can be adjusted without unloading the core avoiding any initial reactivity reserve. The gaseous fission products and some noble metal are extracted by helium bubbling in an on-line fuel processing. The other fission products are daily removed and treated with an off-line fuel pyrochemical process. Core calculations presented here have been made considering that 100kg of heavy nuclei are reprocessed per day and sent back into the core. This corresponds to a daily reprocessing volume of 40 liters of fuel salt.

This off-line fuel processing involves several successive chemical steps: fluorination (extraction under gaseous state of uranium, neptunium, 90% of plutonium and some fission products such as I, Tc, Ru, Te, Nb), molten salt/liquid metal extraction, anodic oxidation, oxide precipitation [10]. The main difficulty in the proposed extraction process concerns the actinide/lanthanide separation. To extract actinides (Acs) and lanthanides (Lns) from the spent fuel molten salt, the same technique is proposed which consists in contacting a liquid metal (Bismuth) with the molten salt [12-15]. The liquid bismuth contains thorium in its metallic state which is a reducing reagent. In this way, a selective extraction of Lns and Acs can be performed by the following chemical reaction:

$$4Ln(III)_{\text{fuel salt}} + 3Th_{\text{liquid Bi}} \rightarrow 4Ln_{\text{liquid Bi}} + 3Th(IV)_{\text{fuel salt}}$$

Using this reaction, the analytical expression of extraction efficiency can be written for each element M as follows.

$$Eff(M) = \frac{n(M)}{n(M)+n(MF_z)} = \frac{1}{1+10^{y}} \quad (1)$$

$$with\ y = (E_{ML} - E°_{MFz/M}) * \frac{zF}{2.3RT} - log\left(\frac{n(LM)}{n(MS)}\right) - log\left(\frac{\gamma(MFz)}{\gamma(M)}\right) \quad (2)$$

$$and\ E_{ML} = E°_{ThF4/Th} + \frac{2.3RT}{4F} log\left(\frac{x(ThF4)\gamma(ThF4)}{x(Th)\gamma(Th)}\right) \quad (3)$$

Where M characterizes the element M in its metallic state dissolved in the liquid metal. MF_z characterizes the element M in an oxidized state dissolved in the molten salt.
In these equations, n(i) is the number of moles, x(i) the mole fraction, γ(i) the activity coefficient of the element i.
E_{ML} is the potential of the liquid metal, $E°_{ThF4/Th}$ and $E°_{MFz/M}$ are the standard potentials of the redox systems ThF_4/Th, and MF_z/M.
n(LM) and n(MS) are the total number of moles respectively of the liquid metal and of the molten salt.
R is the ideal gas constant (8.32 J. $mol^{-1}K^{-1}$), T the temperature (K), F the Faraday constant (96500C), z the valency of the element MF_z dissolved in the molten salt.

In these relations, some parameters only depend on thermochemical data and solvation properties such as potentials and activity coefficients. Only two experimental parameters can be optimized to perform the extraction process: the mole ratio n(LM)/n(MS) and the mole fraction of metallic thorium in the liquid metal, x(Th). Calculations were performed to foresee the extraction efficiency and the selectivity of extraction as a function of the experimental parameters. The thermochemical data were calculated using the HSC chemistry ver. 4.1 software (Outokumpu Research; Finland) and the solvation constants (activity coefficients) were determined from several sources [16-19].

Figure 1 presents the extraction efficiency calculated for plutonium and neodymium as a function of the mole ratio of the two phases.

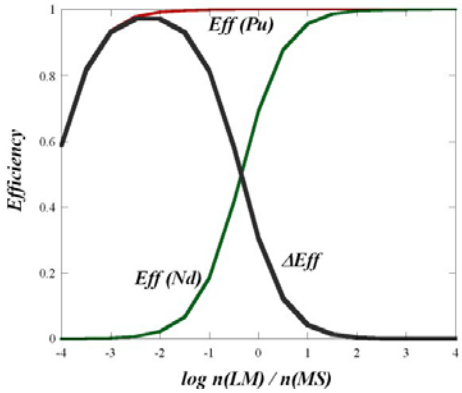

Fig. 1: Extraction efficiency of Pu (in red) and Nd (in green) as a function of the mole ratio n(LM)/n(MS). The black curve gives the efficiency difference between Pu and Nd. Liquid metal is metallic Bi containing metallic Th (x(Th)=10^{-3}mol%=solubility). Molten salt: LiF-ThF_4 (77-23mol%) at 700°C

The calculated curves (Fig.1) indicate that Pu can be firstly selectively extracted for a mole ratio equal to 10^{-2} and in a second step the extraction of Nd can be performed for a mole ratio of 10^{2}.

Some examples of extraction efficiencies are calculated and given for different phase's volume ratio (equivalent to the mole ratio) in Table 2.

TABLE 2
Extraction efficiency calculated for two successive molten salt/liquid metal extractions performed by changing the liquid metal volume (v_{MS} is the molten salt volume and v_{LM} the liquid metal volume)

Element	Extraction efficiency v_{MS}/v_{LM}= 100	Extraction efficiency v_{MS}/v_{LM}= 1	Extraction efficiency v_{MS}/v_{LM}= 0.01
Pu	80.01	99.75	99.99
Am	76.38	99.7	99.99
Nd	0.07	6.58	87.57
Pr	0.02	0.19	65.98
Gd	2.10^{-3}	0.019	16.64
Tb	0.013	0.13	56.86

IV. CONCLUSIONS

The innovative MSFR concept appears to be a very promising MSR concept. Due to the fast neutron spectrum, this concept is actinide burner, thorium breeder and the rate processing is drastically reduced compared to the MSBR concept.

A fuel salt processing has been proposed for this concept. A theoretical approach is developed to optimize the experimental parameters of the actinides and lanthanides extraction steps.

The lack of accurate analytical data such as activity coefficients in liquid metal prevents from the calculation of the other lanthanides extraction efficiencies.

This study shows that fundamental data knowledge is required to analyze the process and propose experimental conditions to perform the actinide and lanthanide extraction with optimized efficiency.

ACKNOWLEDGMENTS

The authors wish to thank PACEN (Programme Aval du Cycle Electronucleaire) of the *Centre National de la Recherche Scientifique* (CNRS) for its support.

REFERENCES

1. P.N. Haubenreich, J. R. Engel, Nucl. Appl. Tech., 8, 118, (1970)
2. E.S. Bettis, R.C. Robertson, Nucl. Appl. Tech., 8, 190-207, (1970)
3. D. Scott, W.P. Eatherly, Nucl. Appl. Tech. 8, 179-189, (1970)
4 L. Mathieu, D. Heuer et al, Prog. in Nucl. En., vol 48, 664-679 (2006)
5 E. Merle-Lucotte, D. Heuer et al, Proceeding of the Int. Conf. On the Physics of reactors, PHYSOR 2008, Interlaken, Switzerland (2008)
6 E. Merle-Lucotte, D. Heuer C. Le Brun, L. Mathieu, et al, Proceedings of the International Congress on Advances in Nuclear Power Plants (ICAPP), Reno, USA (2006)
7 E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le Bun, Materials Issues for Generation IV Systems, NATO Science for Peace and Security Series - B, Editions Springer, 501-521 (2008)
8 J.F. Briesmeister, Los Alamos Lab. report LA-12625-M (1997)
9 L. Mathieu, PhD thesis, Institut National Polytechnique de Grenoble, France (2005) (in French)
10 S. Delpech, E. Merle-Lucotte, D. Heuer, M. Allibert, V. Ghetta, C. Le-Brun, X. Doligez, G. Picard, J. of Fluorine Chemistry, 130, 11, (2009)
11 E. Merle-Lucotte, D. Heuer, L. Mathieu, C. Le Brun, Proceedings of the European Nuclear Conference, Versailles, France (2005)
12 S. Delpech, Proceeding of SESTEC2008, March, New Delhi, India, (2008)
13 S. Delpech, M. Salanne, E. Simoni and G. Picard, Atalante 2008, Montpellier, France, (2008)
14 S. Delpech, G. Picard, MS8, kobe, Japon, (2008)
15 S. Delpech, G. Picard, Proceeding of the International Conference GLOBAL 2005, Nuclear Energy Systems for Future Generation and Global Sustainability, Tsukuba, Japon, October (2005)
16 S. Delpech and G. Picard, ACS 233rd meeting, Chicago, USA, (2007)
17 G. Bourges, S. Rochefort, D. Lambertin, S. Delpech, G. Picard, J. Alloys Comp., 444-445, 404-409, (2007)
18 S. Delpech , G. Picard, J. Finne, E. Walle, O. Conocar, A. Laplace and J. Lacquement, Nucl. Tech., 163, 3, 373, (2008)
19 V.A. Lebedev, Metallurgiya, Chelyabinsk, 1993