MOLTEN SALT REACTOR TECHNOLOGY FOR PARTITIONING & TRANSMUTATION AND HARMONISATION OF THE FUTURE NUCLEAR FUEL CYCLE

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Abstract

The most important developments regarding the available options to reduce actinide inventory will be discussed.

The relation between necessary single cycle transmuted fraction, fuel cycle processing losses and total losses to waste will be defined. An attempt is made to arrange the possible systems on order performance with regard to their potential to reduce the actinide inventory and actinide losses to wastes.

The objective of this paper is to illustrate clearly the potential advantages of molten salt fluoride based reactor technology integrated with innovative processing methods including reductive extraction, electro-refining and electro-deposition relative to traditional ones.

Also, this contribution aims to review the status of molten salt reactor technology, (including Russian experience), to evaluate the importance of remaining uncertainties and to identify the additional work needed.

Introduction

It is evident that the successful solution of the separation & transmutation (S&T) problems may be reached in the most natural way if the fuel is in the molten or gaseous phases for the on-line combination of the reactor and fuel processing plants. These systems could be used to consume TRU with high burnup and minimum handling, without need to repeatedly separate, purify and prefabricate TRU. Operating only at safe location with minimum TRU shipments they would function as near ideal sink for TRU which, once added, could not be easily, removed from the system. In other words it is the entrance door for solving difficult problems of spent fuel and weapon grade Pu.

As a foundation for closing of fuel cycle for all actinides we propose to use molten salt - burner reactors (MS-BR) suitable for integration with waste treatment processes in order to provide a low losses to wastes and extremely high burnup. Same time, differ from other fluid fuel reactor concepts, MS-BR is based on proven technology and has good potential to satisfy future safety requirements, operating as a burner.

These technologies allow to make utilisation of all actinides economic and energy efficient which does not leave them unrequired by power industry, but makes them practically inaccessible for terrorists and criminals.

Fuel cycles , losses to wastes and possible waste treatment

Figure 1 simply demonstrates recycling options for multicomponent Nuclear Power System (NPS) [1-2].The right upper part illustrates the activity for fast reactor cycle and the right lower part indicate the operations needed in a case a MS-BR option. In this context the following facts are important:

- − The primary output of the reprocessing/separation process is material for new fuel.

- − Reprocessing/separation process always have some losses of actinides to waste stream as a secondary output for direct disposal or further input to a separate treatment process.

- − After number of cycles primary output could be moved from one separate S&T cycle to another due to technical limitations or economical reasons.
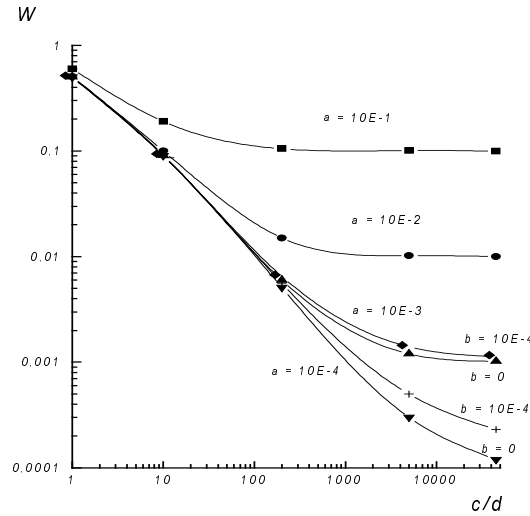
The total fraction of any nuclide which lost to waste stream -W, could be given by:

$$W = a + (1-a)\left[b\sum_{n=0}^{\infty}(1-b)^n(1-d)^n\left\{\prod_{m=0}^{n}\left(1-c_m\right)\right\} + d\sum_{n=1}^{\infty}(1-b)^n(1-d)^n\left\{\prod_{m=0}^{n}\left(1-c_m\right)\right\}\right]$$

where a,b and d are the single cycle fractions of element lost to wastes in normal reprocessing, fabrication and separate processing steps, c_m is the fraction transmuted in recycle pass m ($c_o = 0$).

Using this equation it is possible to analyse various S&T cycles with regard to their ability to reach actinide reduction goals with reasonable losses. It is important that losses in normal reprocessing occur only once in the equation. This term in optimal case should be balanced with other part of the equation. Simple demonstration of the fact that extremely high ratios of c/d in each recycle where ($c = c_m$ = const.) could provide reasonable total losses is given on Figure 1.

Figure 1. **Relation between total losses to waste and ratio**
c/d of fraction transmuted per cycle to fraction lost to waste per cycle

W

1

0.1
$a = 10E-1$

$a = 10E-2$

0.01

$a = 10E-3$
$b = 10E-4$

0.001
$a = 10E-4$
$b = 0$

$b = 10E-4$
+
$b = 0$

0.0001

1   10   100   1000   10000

c/d

For example, if $a = 10^{-4}$, to reach the total losses to waste W about 10^{-3} will be possible for options, with, $c/d=10^3$ (e.g. MS-BRs with $c = 0.5$, $d = 5 \cdot 10^{-4}$ and $b = 0$).

The following developments will be the most important regarding the available options to reduce actinide inventory:

- High neutron flux and short external recycle time interval to provide required balance of neutrons available for transmutation.

- Fuels suitable for high burnup and for separation processes.

- Reprocessing and separation technology with smaller actinide losses to wastes.

- Fuel cycles with less wastes for discharge and disposal.

Evaluating various advanced waste treatment technologies it has been found that several non-aqueous separation processes could be more suitable for this application than traditional aqueous one (see Table 1).

Table 1. Comparison between different fluid actinide fuelled systems

	Compactness	Radiation stability	Chemical stability	Vapor pressure	Solubility of actinides	Corrosion resistance
Aqueous	-	+/-	+/-	-	+/-	+
Molten chlorides	+	+	+/-	+/-	+	-
Molten fluorides	+	+	+	+	+	+
Liquid metals	+	+	+/-	+/-	+	-

The following features of these processes are important:

− The processing media is extremely radiation resistant.

− The working mechanisms are relatively simple and short cooling time is accepted.

− The actinides (An) and lanthanides (Ln) are separated good between groups but poorly within groups.

− Only a few reagents are needed which are mostly used to substitute carrier components.

− The materials are recycled to avoid waste disposal charges.

Some forms of engineering implementation of these processes are partially successful in Oak-Ridge National Laboratory (ORNL) [3], Argonne National Laboratory (ANL) [4], Los-Alamos National Laboratory (LANL) [5], Russian Research Center - Kurchatov Institute (RRC-KI), Institute of High Temperature Electrochemistry (IVTEX) [6] and Russian Institute of Atomic Reactors (RIAR) [7]. The basic processes include reductive extraction, electro-deposition and electro-refining. The carrier and processing media are liquid metals (e.g. Bi) and molten salts (chlorides, fluorides).

Important parameters for reductive extraction and electro-refining are electrochemical potential/free energy of formation $\Delta G^{o}_{f}(T)$ of chlorides (fluorides) and separation factors of elements between molten salt and liquid metal θ [8]. These separation factors for different elements with the same valency could be given by:

$$\ln\theta = \{\Delta G^{o}_{f}(Ln, T) - \Delta G^{o}_{f}(An,T)\} /(RT),$$

where T - temperature (K), R - molar gas constant (J·mol^{-1}·K^{-1}).

Thermodynamic consideration predicts to need more efforts for separating Am and Cm from Ce and La in chloride system in comparison with fluoride one (see Table 2).The calculated separation factors of various elements in fluoride and chloride systems suggest for fluorides a better separation ability between actinides and lanthanides groups but poorly within groups.

Bi-Li reductive extraction flowsheet developed in ORNL [3] for LiF - BeF$_2$ solvent system could provide negligible losses of TRU ($d \approx 10^{-4}$) by use of several counter current stages. For fluoride based systems alongside with this method other method of TRU/lanthanides separation could be considered. This is electrochemical deposition of elements as metals on liquid or solid electrodes in electrochemical cells, filled with fuel salt and bismuth or lead. It is found that the recovery of 99.9% of TRU could be attained by combination of electro-refining and counter current extraction in the LiCl-KCl/Bi system by using Li as reductunt with satisfying the separation ratio between TRU and rare earths of over 1.0 [9].

The applicability of the given method for fluoride based systems is defined by that the fuel salt at temperature 800-1100K is good electrolyte, the conductivity of which is comparable to conductivity of liquid electrolytes. In fluoride systems based on LiF- BeF$_2$ and LiF-NaF-KF the potentials of decomposition for components are higher than that for An and Ln trifluorides.

As on account (see Table 2) the difference of potentials of deposition Am and Ce in fluoride systems reaches 0.2V, it is possible to assert, that at electrolysis of fuel salt there will be the selective separation of these elements. The degree of separation will depends on value of an over-voltage on

4

electrodes. As shows experience of industrial production of a number of alkaline and alkaline-earth elements by electrolysis, at a difference of potentials of deposition of elements more than 0.1V, a degree of their separation is more than 100 (at nominal currents about 1A/cm^2). Thus it is possible to expect for a degree of separation of actinides and lanthanides more than 100, and at several stages of electrochemical clean-up the contents of actinides in lanthanides can be reduced up to 10^4 times. Further investi-gations have to be necessitated to make sure the using fluoride system and efficiency of the electro-winning and electro-refining.

Table 2. **Partial molar Gibbs energy of some trivalent halides, $\Delta G^o_f(T)$, kcal/mol**

Nuclide	Chloride[9] (*T*=800 K)	Fluoride[10] (*T*=1000 K)
U	164	305
Np	174	310
Pu	187	320
Am	195-200**	325
Ce	205	345
La	209	348

Outline of the Future Fuel Cycle

Basing on the status of fluid fuel reactor technology more attractive in future multicomponent NPS we consider processes concerned with the final uses of the recycled fuels as fluorides, in the molten salt reactors (MSR):

− Discharged fuel goes through reprocessing plant, actinides are extracted from waste.

− Uranium after isotope correction and plutonium and, possibly part of Np are recycled as a fuel in thermal and fast solid fuel reactors. Purex reprocessing of spent MOX fuel becomes more difficult, as the content in PuO_2 increases.

− Part of the plutonium together with minor actinides (MA) and if need some FPs are incinerated in a molten salt-burner reactor. Stable and short-lived FPs are removed from the burner reactor by the separation systems. These problems can be solved by chemical and physical methods in batch or on-line modes.

− Enhanced Purex, electrochemical and/or gas-fluoride volatility process should be investigated for considered above recycling options.

− The stable and short-lived FPs are directed to an interim storage facility from where they are sent to final disposal.

The advantages of the MSR as a burner reactor follow not only from a possibility for its effective combination with the pyrometallurgical or gas-fluoride technique of fuel reprocessing/separation, which is low-cost and produces a small volumes of wastes, but also from its capability to use fuel of any nuclide composition.

The MSRs have the flexibility to utilise any fissile fuel in continuous operation with no special modification of the core as demonstrated during MSRE operation for [233,235]U and Pu. Also, a good neutron balance allows use MAs largely contaminated by lanthanides. This simplify the lanthanide/MAs separation process and drastically could decrease the MAs losses to wastes [3].

The MS-BRs further require a minimum of special fuel preparation and can tolerate denaturing and dilution of the fuel.

In addition, there is a possibility to eliminate reactivity accidents in the reactor, providing it is operated at subcritical mode with acceptable, economical external neutron source. The subcritical mode could be important for the MS-BR because of the decrease of the delayed neutrons down to 0.1% due to composition of the fuel and its circulation through the core, as well as use of various fuel compositions.

The proposed strategy of Nuclear Power development is not an alternative to fossil-fired power, but represents its essential supplement. Different types of reactors do not compete with, but complement, each other allowing the best manifestation of the advantages of every type of plant designs. In this case accelerators are considered as neutron sources required to ensure the safe conditions for the functioning of Nuclear Power and to arrange more flexible fuel cycles for large scale Nuclear Power as well as to reduce and then to incinerate the equilibrium quantities of TRU and FPs at the stage of termination of Nuclear Power, if required, when more efficient and acceptable sources of energy appear or when the human kind's way of life and demands change.

Conclusion

MSR technology has been proved by multi-years international efforts. The MSR-Program performed in Russia, mainly at RRC - Kurchatov Institute, covered a wide range of theoretical and experimental studies. The main finding of experimental studies is that neither physical nor technical obstacles on a way to implement MSR and related fuel cycle technologies in the future Nuclear Power System were observed.

There is a good basis for the construction of MSR-based DEMO-facility in Russia. As the first step, the ISTC Project #698 devoted to the molten salt test facility construction could provide key data on the way toward the practical implementation of the accelerator-based transmutation and waste management technology.

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