

Concept of the Demonstration Molten Salt Unit for the Transuranium elements Transmutations

P.Alekseev, A.Dudnikov, V.Prusakov, S.Subbotin, R.Zakirov

RRC Kurchatov Institute, Moscow, Russia

V.Lelek, I.Peka

Nuclear Research Institute Rez, Czech Republic



SK00ST124

Abstract

In this report it is considered fluorine reprocessing of spent fuel and fluoride molten salt reactor in critical and subcritical modes for plutonium and minor actinides burning.

International collaboration for creation of such system is proposed.

It is without any doubt that additional neutron source in the core will have positive influence on the transmutation processes in the reactor. On the other side there is lot of problems to realize it technically and to ensure stable work of the whole complex.

Demonstration critical molten salt reactor of small power capacity will permit to decide the most part of problems inherent to large critical reactors and subcritical drivers which could be useful in future for closing of fuel cycle of large scale nuclear energy system.

If we take into account standard reactor safety demands - negative reactivity coefficients it could be expected that fluoride molten salt transmutor can work without accelerator as a critical reactor.

Series of problems (physical, chemical, material) are formulated and partially answered based on the experience in Russia and Czech Republic to be able to start project works of the such special reactor.

1. Introduction

The large-scale and long-term nuclear power can be developed only if a number of conditions are met: economical efficiency, resource provision, safety, non-proliferation of nuclear materials, ecological acceptability. Each of these problems are currently studied in details, but we will consider only the last one.

The problem of "public acceptance" has become especially critical for the last decade. This problem originates from a negative impact of technologies on the environment and mankind. The nuclear power requires an evident argument that possible negative impacts of the nuclear power radwastes can be minimized and practically eliminated. This directly relates to the problem of closing the nuclear fuel cycle.

Concern about the ecology made some countries establish laws prohibiting export of any radwastes. These laws set an additional technical restriction: the problem of spent nuclear fuel must be solved in each country alone for the established capacity of the nuclear power.

We face the problem to propose technical decisions, to show the readiness for demonstration of these technologies, and to formulate and recommend organizational steps for their implementation, taking into account that the real creation of these technologies evidently exceeds the possibilities of separate countries.

2. Structure of the nuclear fuel cycle

The structure of the nuclear fuel cycle was developed historically and currently with rare exceptions it is one-component with light-water thermal reactors. Fast reactors are economically not competitive and their introduction in the nuclear power will take a lot of time. Under these conditions to close the nuclear fuel cycle means, mainly, to minimize the amount of radwastes from the light-water nuclear power. The lack of a practical demonstration of ecologically acceptable way to close the fuel cycle is an obstacle for further development of the nuclear power.

All isotopes of Np, Am, Cm as well as fission products ^{99}Tc , ^{129}I , ^{157}Sm , ^{158}Tb , ^{168}Ho , ^{126}Pb , ^{93}Zr , ^{79}Se are usually assigned to the long-lived radwastes. If the toxic impact of minor alpha-radiator is evident, the danger of long-lived fission products with low β -activity should be further studied. The balanced and comprehensive consideration of the problem of risk connected with the long-lived radwastes and its minimization can cause that their transmutation will be abandoned in favor of their use or effective ways of their removal from biosphere.

Radioactive isotopes ^{137}Cs , ^{90}Sr has a half-life up to 30 years and low neutron capture cross section, therefore, their transmutation is not efficient. These isotopes should be stored and applications should be searched for them (for instance, as radiation sources for sterilization in industrial conditions).

A list of radioactive isotopes to be burned and transmuted can be changed, in particular, this relates to plutonium which is assigned to radwastes in a number of countries.

The search for ways how to close the nuclear fuel cycle leads to technological decisions with a multi-component structure of the nuclear power (Fig. 1). This structure includes a

reactor-burner for burning and transmutation of radwastes [1]. In such a structure the nuclear fuel cycle becomes closed not in far future when fast breeders operates, but “right away”.

The three-component structure of the nuclear power includes:

- thermal reactors in which, as known, balanced amounts of plutonium are minimal and provide economical efficiency of the nuclear power;
- fast reactors provide neutron balance in the system of the nuclear power necessary for closure of the nuclear fuel cycle for all dangerous long-lived radioactive nuclides; these reactors allow to decrease the consumption of uranium-235 in the nuclear power system and minimize the plutonium amount in the system;
- molten-salt reactors-burners (critical and subcritical) as components of the fuel cycle are necessary for closure of the fuel cycle for Np, Am, Cm and allow to operate with variable nuclide composition;
- technologies on fuel reprocessing that allow to decrease the typical times of the nuclear fuel cycle and diminish the amounts of radioactive nuclides that are unrecoverably lost.

Table 1.

Quantities of Different TRU in Equilibrium Closed Fuel Cycles
for Thermal (LWR), Fast (FR) and Molten Salt Reactors (MSR), Ton/GW(e)

System	LWR	75%LWR+ 25%MSR	FR	89%FR+ 11%MSR	51%LWR+38%FR +11%MSR
Cycle Average Neutron Flux Φ , n·cm ⁻² ·sec ⁻¹	10 ¹⁴	10 ¹⁴ (LWR) and 5·10 ¹⁵ (MSR)	10 ¹⁵	10 ¹⁵ (FR) and 5·10 ¹⁵ (MSR)	10 ¹⁴ (LWR) 10 ¹⁵ (FR) and 5·10 ¹⁵ (MSR)
²³⁷ Np	0.72	0.12	0.11	0.02	0.02
Pu (total)	5.8	3.1	21.1	18.0	10.4
²⁴¹ Am + ²⁴³ Am	0.76	0.08	0.77	0.10	0.19
Cm (total)	1.38	0.09	0.19	0.04	0.11
TRU (total)	8.7	3.4	22.2	18.2	10.7
TRU without Pu	2.88	0.3	1.10	0.17	0.3
Heavy Nuclide	265	283	121	117	221

3. Comparison of efficiency of radwaste burning and transmutation in solid-fuel and liquid-fuel reactors.

3.1. Comparison of Actinide Transmutation Efficiency in Solid Fuel Rod and Liquid Fuel Reactors.

When the reactor operates at thermal power of 1000 MW for 7000 hours a year approximately 300 kg of actinides are undergone fission. In fast neutron spectrum the reactor can operate in critical conditions when only minor actinides are loaded (MA - Np, Am, Cm). Therefore, in case of fast neutron spectrum both in solid fuel rod and liquid-fuel reactors for a year 300 kg of MA will be burned at each 1000 MWth.

However, much more fuel circulates in the fuel cycle of a solid-fuel reactor than in the fuel cycle of a liquid fuel reactor. It is connected both to restricted burnup (5-10 times more fresh fuel will be discharged per kilogram of spent fuel) and to longer duration of the out-reactor fuel cycle. Correspondingly, it deteriorates the neutron balance of the solid fuel reactor because of larger amount of natural decays of actinides (especially, Pu-241) and due to increase in loss of actinides in the fuel cycle, which, while all other factors being the same, is proportional to amount of nuclides in the fuel cycle.

Note, however, that due to technical and technological complexities and large amounts of actinides in the fuel cycle under equilibrium operational conditions, typical for fast reactors, their use as burners of minor actinides will prove to be, evidently, not expedient.

Thermal neutron spectrum in reactors-burners allows for significant decrease in equilibrium amounts of transuranic nuclides to be burned. It is explained by a considerably higher probability of interaction of these nuclei with thermal neutrons.

However, thermal reactors cannot operate in critical conditions when fed up by only minor actinides. Along with minor actinides U-235, or Pu-239, or U-233 should be loaded as additional neutron source, or they should be fed from external source (subcritical operational conditions).

For case of burning of minor actinides with composition typical for fuel discharged from LWR preliminary estimates of the burning rate in equilibrium reactor operation are presented in Table 2. Uncertainty of these results is about 10%. These estimates show that due to higher neutron density and better neutron balance the MA burning rate in molten-salt reactor higher

than in a solid fuel reactor by 20 kg/GWt-yr if U-235 is used as a neutron source, and by 30 kg/GWt-yr if Pu-239 is used.

Table 2.

Estimate of the burning rate of MA (Np, Am, Cm) discharged from LWR for various reactors-burners (thermal solid-fuel and molten-salt reactors) with the use of different neutron sources (U-235, Pu-239, external neutron source), kg/GWt-yr

Neutron source	Solid-fuel $\Phi^*=10^{14}$ n/cm ² s	Liquid-fuel $\Phi^*=10^{15}$ n/cm ² s
²³⁵ U	70/100**	90/120
²³⁹ Pu	80/110**	110/140

* - average neutron flux density with account for external fuel cycle

** - critical/subcritical reactor $K_{eff}=0.95$

Under critical operational conditions the MA burning rate increases more by about 30 kg/GWt-yr at subcriticality level of 5%. Noteworthy that, if all transuranic nuclides (TRU - Pu, Np, Am, Cm) discharged from LWR after the first irradiation cycle are burned in MSR, the neutron balance in the reactor is such that even after 20-year exposure of irradiated fuel in the external fuel cycle no additional loading of U-235 or Pu-239 is required to sustain criticality under equilibrium operation conditions.

The external neutron source, in this case, can be used only for enhancement of the reactor safety. The TRU burning rate will equal 300 kg/GWt-yr both in critical and subcritical reactor operation.

Molten salt reactor (MSR) with fluoride molten salt is now the only applicant for transmutation both in accelerator-driven and critical options.

Introduction of neptunium, americium, curium, and fission products in solid-fuel thermal and fast reactors can complicate the reactor design, negatively impact their safety, require development of new fuel types. To localize nuclear fuel and fission products as well as to organize heat removal in the core additional structural materials should be introduced, making the neutron balance worse. Deterioration of the neutron balance in the solid-fuel reactors is also connected with neutron capture by fission products generated in the fuel rods for all fuel cycle. Radiation defects in structural materials restrict fuel burnup depth in the solid-fuel

reactors. All this will require multiple fuel reprocessing with on-line refuelling and, therefore, the loss of radioactive toxic nuclides will increase.

3.2. Comparison of critical and subcritical reactors

Advantages and shortcomings of subcritical nuclear reactor scheme are rather evident:

- reactivity-initiated accidents can be excluded;
- reactor control becomes more effective;
- additional neutrons are produced that allow to perform the total number of fissions on Np, Am, Cm without adding in the core ^{235}U , ^{233}U or Pu that decreases required fraction of reactors-burners in the nuclear power sector.

The shortcomings of the subcritical system are:

- decrease in the nuclear energy fraction delivered to consumers;
- complexity of the system design related to the target unit and introduction of neutrons from the external source in the core;
- current lack of neutron sources that meet requirements on reliability typical for power systems (7000 hours of trouble-free operation in a year);
- possible deterioration of the reactor inherent safety in accidents with loss of primary coolant flow when the external neutron flux is wrongly switched off.

The use of the cascade principle of neutron multiplication from the external source allows to decrease by several times its power, but increases spatial power nonuniformity in the reactor [2]. Thus, the subcritical reactor design can be optimized.

One subcritical cascade MSR of 1 GWt can burn about 300 kg Np, Am, Cm annually, while the total power of such reactors in the nuclear power system makes about 3% (Fig.2).

3.3. Molten-salt reactor as an integral element of the fuel cycle

Thus, the solution is in creation (see Fig. 1) of reactors with circulation of molten-salt fuel should operate with thermal and fast reactors to close the fuel cycle. Accordingly, MSR should be considered with fuel compositions, including Np, Am, Cm and various amounts of plutonium.

It is evident that the system with thermal and molten salt reactors, but without fast breeders will be closed, but will have no resource stock on ^{235}U and ^{239}Pu .

Rather large research and technical background has been accumulated for decades of research of MSR and advanced technologies for radwaste regeneration. This background provides the opportunity to make a next step in formation of a concept of the model closed nuclear fuel cycle. The background includes, first of all, the pioneer and now classical researches in Oak-Ridge at the MSRE reactor, as well as:

- RRC “KI” studies at the “Solaris” molten-salt facility, and then at the “Kurs-2” reactor loop with thermal circulation;
- studies on fluoride regeneration of fast reactor fuel at “Fregat” facility (RRC “KI”, NIAR, NRI Rez)
- project of Soviet-French enterprise for regeneration of spent fuel from BN-350 and “Rapsodia” reactors (France, USSR);
- development of structural materials resistant in molten fluorides at high temperatures: Hastelloy-N (USA), XH80MTU (USSR), MONICR (ChR);
- other activities.

All above shows a good background for the next step that can be a creation of a model facility.

However, a number of nuclear-chemical studies should be done to adopt the available background to the MSR-burner features. First of all, there is not enough direct experience on determination of efficiency with which neutrons impact minor actinides. To fill the gaps in technological knowledge the following Programs were developed in Russia and Czech Republic: R&D Program “Development of a concept for molten-salt reactor for the nuclear power fuel cycle” (Russia, April 1999) and Program on burning and transmutation of nuclear fuel (Czech Republic, May 1999).

What are today the problems on the closed nuclear fuel cycle and how should they be solved?

4. Requirements to the closed nuclear fuel cycle

The fate of plutonium in the nuclear industry is determined by the planning police for the nuclear power structure in each country. However, some common requirements to non-proliferation of the nuclear materials should be met. On our opinion, disposition of spent fuel reprocessing facility and MSR-burner on one site correlates with these requirements to the maximum extent.

The countries with not high nuclear power (1-4 GW) do not face the problem of plutonium utilization in light-water or liquid-metal fast reactors. A rather optimal scheme for them is to reprocess spent fuel with uranium extraction and removal of fission products in technological process. Plutonium along with minor actinides and fission products in this process is transferred to MSR for burning and transmutation, correspondingly.

The countries with the developed nuclear power (with established power of 20 GW and higher) plan or already realize MOX-fuel manufacturing (for light-water and fast reactors). The technological process of spent fuel regeneration should, in this case, provide separation of uranium and plutonium with removal of minor actinides and lanthanides. In this scheme fission products are partly left in plutonium to prevent its stealing; separated actinides are transmuted in MSR (critical or subcritical), and lanthanides are removed and transferred for disposal.

For spent fuel from fast reactors separation of uranium, plutonium and minor actinides can be unnecessary.

4.1. Requirements to reactor facility

The molten-salt reactor technology was developed to the enough extent to conduct a test model reactor start. The reactor design is flexible and can be easily adopted to various fuel cycles. It can be used both for decrease of transuranic wastes as well as in Th-U and U-Pu fuel cycles, including for utilization of weapons-grade or civil-grade plutonium in uranium-free composition.

Safety of such a reactor is based on fast negative density reactivity coefficient and passive drainage of molten fuel composition in subcritical tanks when temperature limits are exceeded in the primary circuit.

Creation of the model reactor-burner requires the following sequence of the stages:

- completion of R&D with molten-salt composition, containing Pu, Np, Am, Cm;
- development of MSR conceptual project;
- development of MSR draft project;
- development of technical project and working drawings;
- licensing of the model facility project;
- construction of the model facility project;
- *licensing and test start of the model facility for closure of the nuclear fuel cycle.*

In operation of the model facility the followings will be confirmed or determined:

- parameters of the system;
- technology for the spent nuclear fuel reprocessing;
- technology for sustaining the salt composition and properties;
- technology for separation of fission products;
- resource parameters of equipment;
- behavior of structural materials;
- methods of the model unit monitoring and control;
- choice of the most effective designs of equipment checked in various loops of the model unit.

On the basis of all studies and operation of the model unit a project will be developed for industrial facility of MSR-burner for the nuclear fuel cycle.

As a first step to start the work, experience gained in different countries on molten-salt technology should be generalized. To create the model unit an International Consortium should be organized that will require inter-government Agreements between countries-participants.

In the Agreements division of the activities between countries-participants will be noted, responsibility and types of cooperation will be determined.

An appropriate place for construction the model unit can be found in Russia in frames of the program on conversion of Minatom enterprises.

An important advantage of the molten-salt model reactor is that average neutron flux density can be provided higher than $5 \cdot 10^{15}$ n/s-cm².

The model reactor can operate with a number of molten-salt compositions and with various neutron spectrums (from thermal to fast neutron spectrum).

On-line fuel removal of fuel for regeneration and removal of fission products as well as on-line feeding with actinides can be organized in MSR.

To develop a project of the model unit, safety assurance is required for all system as a whole, including feeding and fuel regenerating circuits with account for decreased number of barriers and new safety requirements and normative requirements.

Technical and economic studies are necessary at all stages of development and operation of the model reactor, including manufacturing of heat-resistant structural materials like Hastelloy, XH80MTU, etc.

MSR chemistry

Compared to the MSRE-ORNL and planned then MSBR (breeder) the molten-salt reactor-burner will undergo practically no changes if the molten-salt coolant $2\text{LiF}\text{-BeF}_2$ remains the same when uranium-thorium fluorides are replaced with fluorides of plutonium and minor actinides. However, solubility of PuF_3 in this matrix is not high from viewpoint of realization of the idea of burner. BeF_2 ties up fluoride-ion due to formation of complex and, hence, excess (or deficiency) of free fluoride-ion in the melt depends on the ratio of initial components of the mixture. This parameter (as well as the redox potential) strongly impacts solubility of various components in the melt. Such a dependency for PuF_3 is shown on Fig. 3 [3]. This Fig. shows that the solubility of PuF_3 has a peak value in pure LiF and in the LiF- BeF_2 melt has a minimum of 0.5-1 mol.% at 600°C for the BeF_2 content equal to about 40 mol.%. The solubility increases when temperature grows and equals 1-2 mol% at 700°C and 2-3 mol.% at 800°C [4]. The solubility of PuF_3 was studied in Oak-Ridge and its studies has recently started in NIAR [5, 6]. All fluorides of fission products can be solved in the LiF- BeF_2 melt. According to [7], the ultimate solubility of fluorides of three-valent elements in fuel mixture of MSR equals about 1 mol.% (at $T = 700^\circ\text{C}$). The solubility of elements of the first (cesium) and second (strontium) groups is higher at least by an order of magnitude.

There is a number of ideas how to increase the solubility of PuF_3 in LiF- BeF_2 . The study of this problem is important, especially, taking into account the dispersion of corresponding data available in literature. It is believed that the updated salt composition LiF- BeF_2 - PuF_3 -MA will have thermal properties close to the salt properties in the ORNL-MSRE test and that the required redox potential will be reached in it.

The chemical reprocessing of irradiated molten salt composition, circulating in MSR, is necessary to remove from the melt neutron poisons which can break the balance of reactor operation. This process can be on-line or periodical in several years. This process will be developed with use of experience gained in classical pioneer studies at MSRE in Oak-Ridge.

The highest neutron capture cross section among well soluble fission products has lanthanides and their removal from the molten salt composition, circulating through the reactor, is the most necessary. The lanthanides in this process should be separated from actinides which should be returned to MSR for burning.

Rare-earth fission products can be removed and separated from actinides by several ways: distillation of fluoride salts of lithium and beryllium [8]; selective reduction, for example, by metal cast introduced in the system through the liquid-metal extractant in the process of extraction [9]; deposition by the method of fluorine-oxygen exchange [10]; and electrochemical methods in combination with extraction by liquid metal.

The process of electrochemical separation of actinides and lanthanides in fluoride medium is rather new and required studies of thermodynamics and kinetics of oxidation and reduction transitions, in particular, reduction of actinides and lanthanides to the metal state.

Fluoride compositions (LiF-KF-NaF, LiF-NaF, LiF-BeF₂) allow to conduct effective separation of actinides and lanthanides. For comparison Table 3 is given below with values of free energy of formation (Gibbs energy) for chlorides and fluorides of the elements most important while reprocessing radioactive elements. Difference between values of Gibbs energy determines a theoretical value of the separation factors for various elements.

Analysis of the data given in Table 3 shows a beneficial opportunity to use electrochemical separation operations in fluoride systems.

Table 3.

Computational values of Gibbs energy for chlorides (800K) and fluorides (1000K) for a number of elements [11].

Element (valence-3)	$-\Delta G_T$, kcal/mol fluoride	$-\Delta G_T$, kcal/mol chloride
U	305	164
Np	310	174
Pu	320	187
Am	325	195-200*
Ce	345	205
La	348	209

* - value defined by comparison of the data on ΔH_T for Pu and Am

To conduct these studies a number of practical problems should be solved. Among them are development of design for special electrochemical cells and ion-conducting membranes,

choice of high-temperature reference electrode, etc. A design should also be developed that combines in one technology unit the extraction process by molten metal with electrochemical reduction with controlled potential.

Main features of a conceptual technological scheme proposed for reprocessing of the fuel composition are presented in Fig. 4.

The basis of the scheme is the use of electrochemical method for separation and purification of actinides and lanthanides. The actinides and lanthanides are separated in electrolytic baths with the use of solid (graphite) and/or liquid-metal (bismuth) electrode-carriers.

At the first stage, electrochemical reduction of plutonium and minor actinides occurs on the electrodes, then deep electrochemical purification of the salt from the rest of minor actinides. At the same stage, besides MA some amount of rare-earth elements can be formed. Significant amount of rare-earth elements along with other fission products is directed with salt flow to operation of deposition by the method of fluorine-oxygen exchange.

Plutonium and minor actinides, deposited on the electrodes of the electrolyzers in form of metal and containing rare-earth elements in amounts allowable for reactor operation, directed to the system of feeding of the molten-salt reactor for burning.

Introduction of iodine and technetium in MSR for transmutation is complex problem that still has not been studied enough. It is connected with the fact that fluoride compounds of these elements are unstable at reactor operating temperature and low-soluble in fuel salt. This problem should be solved. A possible way of the solution is to use hermetic volumes with these elements (or their compounds) for irradiation in the chosen place of the reactor salt core.

4.2. Processes of spent fuel regeneration

Fluoride chemistry of LWR spent fuel regeneration to the most extent corresponds to molten-salt reactor for radwaste burning.

The processes use fluorine and fluorides with methods of gas fluorination, electrochemistry, fluorine-oxygen exchange. The current nuclear industry widely uses fluoride technologies in operating manufacturing of uranium hexafluoride, fluorine, separation of isotopes, metallurgy of uranium, zirconium, etc.

The closed fuel cycle can be implemented in the form of a process in the fluoride environment with high stability against radiation impacts, fire- and explosion-resistant with minimal losses of actinide elements as radwaste (0.1-0.2%).

Preparation of fuel for the molten-salt reactor can not be considered without analysis of technology for LWR spent fuel regeneration as a main source of radwastes. The molten salt reactor for burning of radwastes should play a role of the completing stage of the technology for LWR spent fuel reprocessing.

Conceptual schemes of fluoride LWR spent fuel regeneration are presented in Fig. 5-6. The technological schemes have the following common operations:

- stage of taking off the covers of fuel elements and extraction of fuel composition;
- release and capture of gas components of radwastes;
- separation and purification of the main component of spent fuel – uranium;
- separation of excess plutonium directed to manufacturing of traditional and MOX fuel for thermal and fast reactors;
- separation (if necessary, with deep purification) of the rest of spent fuel components in groups of elements: actinides, lanthanides, alkaline and alkaline-earth metals, for burning in MSR and long-term storage under monitoring;
- utilization (burning and transmutation) of long-lived fission products (minor actinides, iodine, technetium) in MSR and reprocessing of fuel composition in MSR.

They differ, mainly, by the methods of extraction of regenerated uranium from spent fuel in the form of UF_6 by the fluorination reaction.

In fluorination various approaches are used: direct impact of fluorine on radwaste oxides (Fig. 5), fluorination of uranium-containing $LiF-NaF$, anode electrochemical fluorination of soluble uranium fluorides (Fig. 6).

In gas fluorination the initial stage of developed scheme is mechanical cutting off of fuel assembly. Fluorination of fuel rods with zirconium cladding without its previous separation is also possible. Zirconium tetrafluoride is sublimated and can be separated from other more volatile components, as for instance, uranium hexafluoride.

Powder fuel is directed to fluorination in the apparatus for fluorination of a flame type. Such a method of UF_6 generation was used at the "Fregat" test facility and is used in industrial technology for fluorination of uranium dioxide of natural and enriched isotope content. The operation of uranium hexafluoride separation from radwaste at the first stages of the process is especially attractive because the amount of remaining fuel components is reduced to

minimum. The original mass of radwastes after removal of uranium reduces by 80-100 times. The dimension of the equipment for subsequent operations with fuel components becomes laboratory-scale. After distillation and purification from fission products UF_6 is directed to the separation enterprise for isotope correction. The regenerated uranium has high enough residual isotope content of U-235 (about 1%). Recycle of uranium is economically justified and technically feasible on the basis of the centrifugal technology of the isotope separation in spite of the presence of U-232, U-234, U-236 in spent uranium. Assessments that we made show that the cost of the recycled material for the repeated use in LWR is by 30% lower than the natural uranium of the same enrichment.

Formation of the uranium hexafluoride is accompanied by sublimation of those fission products which have volatile fluorides. These are, mainly, elements of 5-7 groups of the periodical system in their highest oxidation levels. The uranium hexafluoride is separated by distillation in packed columns and condensed in transport volumes.

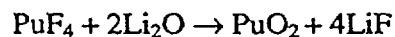
The separated fluorides of fission products (MoF_6 , TcF_6 , PdF_6 , NbF_5) are subjected to pyrohydrolysis directly or previously absorbed on adsorbent of NaF and then subjected to pyrohydrolysis in a combined form, transforming in oxide form with further glass and storage.

NpF_6 accompanies uranium hexafluoride and after separation of fission products sorbited on magnesium fluoride and can be stored in this form.

The vapor pressure of PuF_6 is closed to uranium hexafluoride and the possibility of joint generation of two hexafluorides in fluorination of radwastes cannot be excluded. However, a large excess amount of fluorine is required for it. When fluorine is deficient plutonium tetrafluoride is formed and accumulated in a solid residue of non-volatile products of fluorination along with fluorides of elements of 1-3 groups of the periodical system, including actinides and lanthanides.

The further process of separation of plutonium and other groups of elements in the considered scheme is conducted through molten salts. A mixture of lithium and sodium fluorides ($LiF-NaF$) or other compositions ($LiF-NaF-KF$, $LiF-BeF_2$) with the melting point of $800^\circ C$ to $500^\circ C$ is used as a reaction environment to conduct reaction of fraction separation of the components.

To extract plutonium the process of plutonium deposition in the form of oxide [10] with subsequent its use for MOX-fuel manufacturing is provided in the scheme. The plutonium oxide is deposited from the salt bath with $LiF-NaF$ with addition of lithium oxide (or other oxide):



The use of lithium oxide as a chemical reagent does not cause significant changes in the carrier composition and, hence, its properties. An alternative option for excess plutonium extraction from salt composition can be electrochemical reduction on solid or liquid metal electrode in an elemental form. In such an approach plutonium separation becomes easier (without the use of filtration). However, this option has not been checked experimentally and, according to thermodynamic estimates) in fluoride melts plutonium generation can be accompanied by deposition of minor actinides on the electrodes.

In the schemes we propose actinides and lanthanides are separated in the successive electrochemical reduction and oxidation of the necessary groups of elements in electrolyzers filled with fluoride melts.

At the final stage of the separation of actinides and lanthanides electrochemical reduction is provided with subsequent its electrochemical solution in the LiF-BeF₂ melt and introduction in MSR for burning (Fig. 4). The LiF-NaF melt contains rare-earth elements as well as Cs, Sr, etc and is sent to processing by the fluorine-oxygen method [10]. With this aim Li₂O is introduced in the melt and causes deposition of radioactive elements in the form of oxides. The last ones are separated on the filter and sent to glass and storage.

5. Structure of work on directions.

Physics, hydrodynamics and mathematical modeling

Molten-salt reactor has a number of features that require not standard approaches to modeling:

Neutron-physical processes are calculated with standard models, but the following effects should be taken into account: spatial distribution of temperature and density of molten salt, decrease in fraction of delayed neutrons, isotope kinetics both in the core and out of core, removal of fission gas products and a part of nonsoluble fission products. Adequate estimate of time-spatial distribution of radioactive and heat generation sources is necessary.

Thermal-hydraulics for the MSR core should describe three-dimensional flow and mass transfer with account for volume power generation in all primary volume. Comparison with experimental data is required. Neutron-physical, thermal-hydraulic and chemical processes, determining mass transfer and amount of fission products, should be coupled in a unite model. Uncertainties of parameters will depend on both technological parameters, errors of

comprehensive models and uncertainties of nuclear data for fission products and transplutonic nuclides in the MSR spectrum.

Chemistry and problems of separation

The main problem is without doubt improvement of PuF_3 solubility in the salt by several times (2-3 as a minimum) to close the properties of the molten salt composition to those of MSRE and all experience gained at MSRE and principles of redox potential maintenance for protection against corrosion could be used. It is essential that although new mixtures of fluorides can be advanced in some directions, but can significantly advances readiness of demonstration tests.

Design materials

There are enough data and experience with Hastelloy-N, materials of similar properties developed in Russia (XH80MTU) and in Czech Republic (MONICR). The conducted studies showed no problems with welding.

There is no still unique answer to the problem of destruction of a graphite wall under effect of fluorides. Even if the core is designed without the graphite, according to the current view, the graphite should be used for protection of the reactor vessel against fast neutrons. Life time of these components as well as ultimate allowable neutron fluxes should also be found.

Test facilities

Engineering and technological problems for qualification of the Model test (MT) cannot bypass studies at loop and other test facilities. Among them are the followings:

- loop for imitation of the process with secondary coolant was constructed in NRI (Rez); studies have already started;
- loop from the MONICR material for study of engineering operations connected with primary circuit of MSR (helium introduction, corrosion, heat exchanger, diagnostics, technological control, etc.) will be prepared for operation in the end of 1999; studies of salts containing radioactive materials are not planned;
- the "Ampula" facility for nuclear-physical studies of efficiency of minor actinides burning in molten-salt compositions in the IR test reactor in RRC "KF";
- project of a large loop, probably in Snezhinsk (Russia), for study of circulation, hydrodynamics, heat transfer, scram operation for molten-salt fuel;
- facility for measurement of the electrochemical potentials of lanthanides and actinides.

Studies in fluoride melts are conducted in NRI (Rez) and RRC "KF". In the Kurchatov Institute molten-salt fluoride mixtures, including plutonium, minor actinides, lanthanides are studied.

Territorial proposals on organization of the model test

There are proposals from the following organizations:

- Chelyabinsk-70 (Russia);
- Dimitrovgrad (Russia).
- Electrical plant site of Dukovany (CR) well fits for creation of Interregional (Czech, Slovakia and Hungary) system of the closed fuel cycle with reactor on salt melts and fluoride technology of VVER spent fuel regeneration. These countries express their desire to conduct a common technical politics in the field of the closed fuel cycle.

6. Concept of the first model test

A concept of the model test used in this report is adequate to the concept of the closed fuel cycle of the nuclear power. The latter includes regeneration of LWR spent fuel, MSR-burner, radwaste processing before the deposition. The work should be done for all these components of nuclear fuel cycle, however, MSR will have a central place. Preferences of the experts are that the first model MSR with plutonium would be to a significant extent a copy of MSRE. Determination of its power should be linked with ultimate allowable salt flows, salt volumes, possibilities of chemical reprocessing. It is supposed that possible MSR power will be taken 15 MW and increase with accumulation of the experience.

Although the main aim of the project is a check of the closed fuel cycle for LWR, it should be noticed that such a special test can bring much information on such problems as:

- reprocessing of the nuclear fuel reloaded from submarines and research reactors (MSR of about 50 MWth can burn Pu+MA from all European research reactors. High-enriched uranium can be extracted and radwastes are in a negligible amounts);
- plutonium burning with formation of ^{233}U from ^{232}Th ;
- manufacturing of $^{99\text{m}}\text{Tc}$ and other radioactive short-lived isotopes for medical aims;
- the use of circulating molten-salt composition as a source of γ -radiation of high intensity with corresponding applications;
- generation of high-temperature heat by molten-salt reactor systems.

Financing sources

All possible participants of the project (except Russia for the moment) have foundations for the back end of the fuel cycle and, therefore, the model test can be financed from these funds. It should be noted that in majority of countries the main destination of the foundations for the back end of the fuel cycle is to solve the problems of final radwaste deposition. The problem of physicists and engineers is to convince the foundation holders to finance step by step the technical development of the closed fuel cycle, because it supposes the solution of the radwaste problem that is more economic than direct radwaste storage.

Organizational measures and proposals

Three international organizational structures are currently known to experts in frames of which works are conducted on the closed fuel cycle.

- The ISTC is still the main potential source of financing for Russian works. The main aim is a transformation of military activities to peace applications. The possibility of participation of associated members of EU and Slovakia has not still been clarified.
- The fifth European Program (5EP) is on the opinion of the experts the main organizational structure for conducting of the works. There is a section in it – a large molten-salt reactor. The possibility of financing of the Russian activities or combination of 5EP with works in frames of ISTC should be clarified.
- Atom Energy Research (AER) includes, mainly, the countries and their institutions that operate VVER-type reactors. The topical group “Transmutation” was formed. This group is still limited by only scientific problems and devoted to only a preliminary introduction in the problems in fields of the physics.

Finally, we should note that the experts think it would be very desirable if a large experience gained in nuclear and chemical studies of salt melts beginning from ORNL serves for the nuclear power development.

We hope that the organizational measures will be found to help that small groups of experts which work in the direction of preparation of the technology for the closed fuel cycle.

REFERENCES

1. S.Subbotin, P.Alekseev, V.Ignatiev e.a. Harmonization of Fuel Cycles for Long-Range and Widescale Nuclear Energy System. In Proceed of GLOBAL'95

2. Alekseev, V.V. Ignatiev, L.I. Menshikov e.a. «A Concept of Molten-Salt Subcritical Reactor of Enhanced Safety». *Atomnaya Energiya* 79, p. 243, 1995; A Cascade Subcritical Reactor of Enhanced Safety, ad hoc p. 327.
3. J.C.Mailen et al. «Solubility of PuF₃ in molten 2LiF-BeF₂», *J.Chem.Engng.Data*, 16 (1), 68, 1971.
4. C.J.Barton, *J.Phys.Chem.*, 64, 306, 1960.
5. V.S.Naumov, A.V.Bychkov - International Seminar «Molten Salts in Nuclear Technologies». Dimitrovgrad, June 19-22, 1995: Collected Volume of Abstracts, Dimitrovgrad: SRC RF RIAR, p. 24-25, 1995.
6. V.S.Naumov e.a. Interaction of Actinide and Rare-Earth Elements Fluorides with Molten Fluoride Salts and Possibility of Their Separation for ADTT Fuel Reprocessing. In Proc. 1996 International Conference on Accelerator-Driven Transmutation Technologies and Applications. Kalmar, Sweden, June 3-7, 1996.
7. K.Furukawa, A.Lecocq et al. «Thorium fuel-cycle development through plutonium incineration by THORIUM-NES (Thorium molten-salt nuclear energy synergetics)». - In advanced fuels with reduced actinide generation». - Proc. of a Tech. Committee Meeting, Vienna, 21-23 Nov. 1995, p. 115-127, IAEA-TECDOC-916, 1996.
8. W.E.Miller et al. - *Reactor Fuel - Processing Technol.*, v. 11, No.2, p. 96, 1968.
9. M.E.Whatley. - *Nucl. Appl. Technol.*, v. 8, p. 170, 1970.
10. V.F.Gorbunov et al. *Radiokhimiya*, XVIII, 1, p. 109-114, 1976.
11. R.Ya.Zakirov, V.N.Kosjakov, V.N.Prusakov - Electrochemistry in the regeneration of a molten salt nuclear reactor fuel. Preprint RRC KI IAE-6061/13, Moscow, 1998.

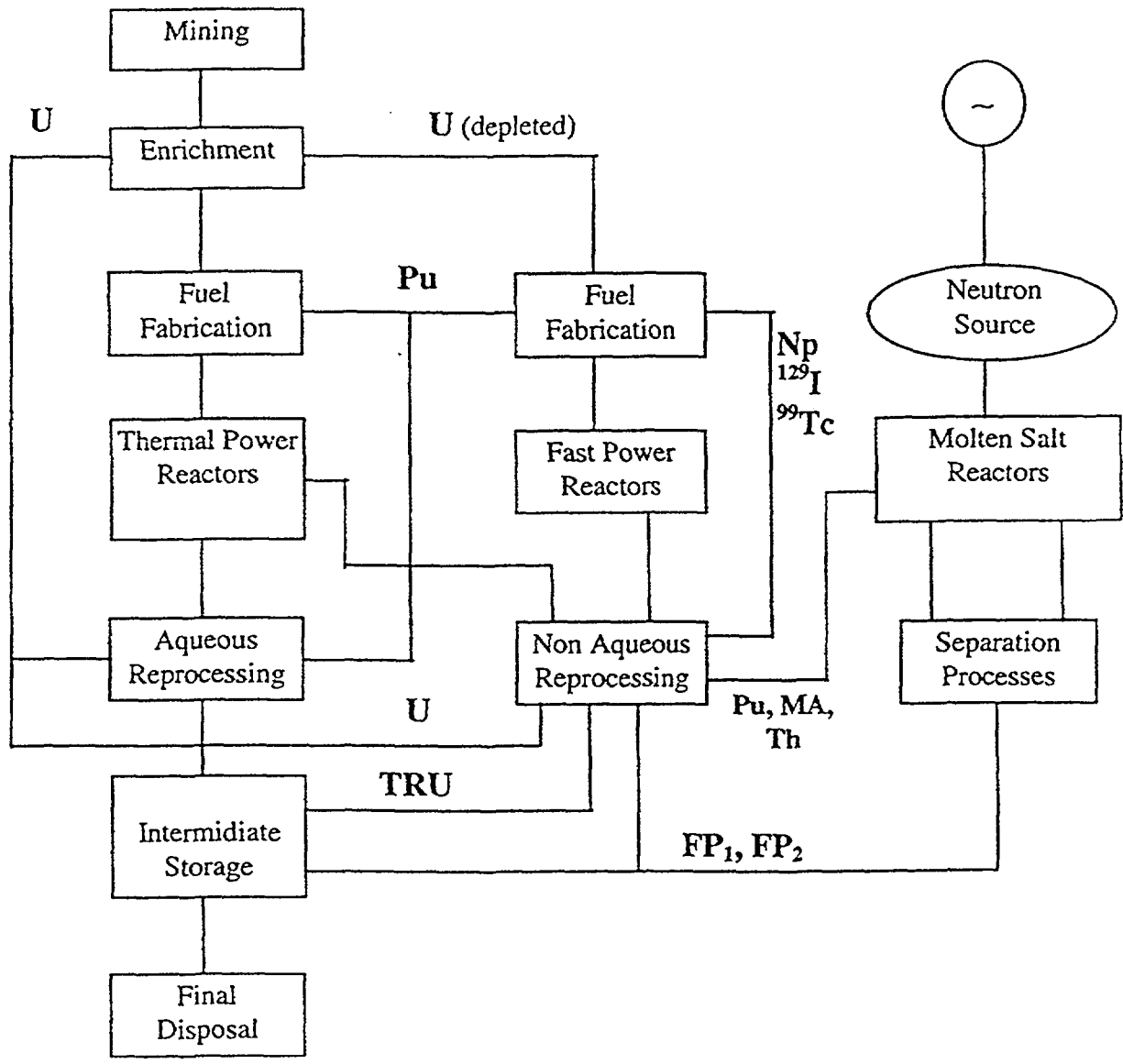


Fig. 1. Multicomponent nuclear energy system with closed fuel cycle for all actinides and dangerous long-lived fission products. FP₁, FP₂ - short-lived and long-lived fission products

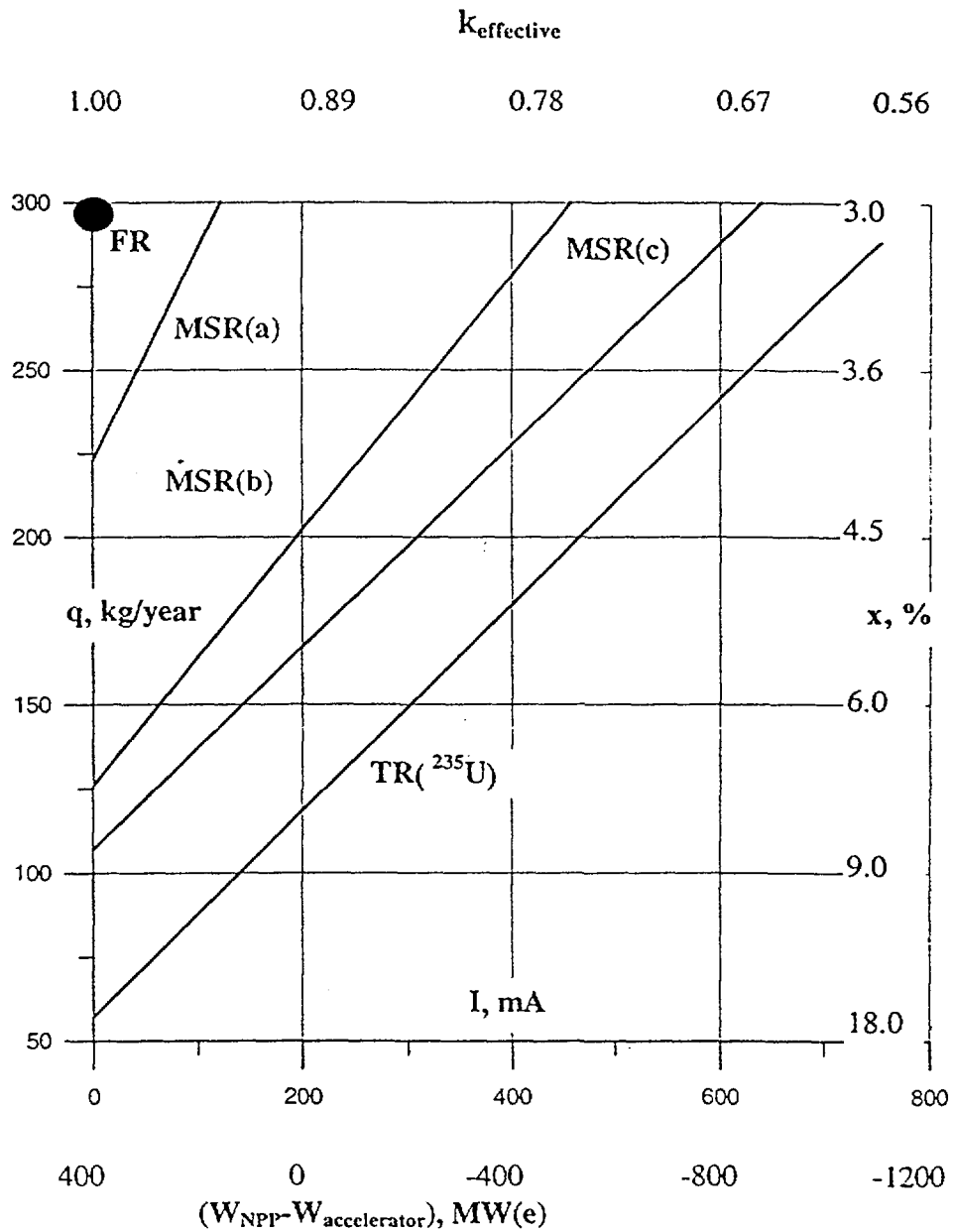


Fig.2. Burner-reactor efficiency for minor actinides incineration

(Thermal power of Burner-reactor - 1000 MW)

q - rate of minor actinides incineration, kg/year;

I - current of proton beam ($E_p = 1$ GeV), mA;

x - share of Burner-reactor power in Nuclear Power System, %

TR - solid fuel thermal Burner-reactor ($F = 10^{14}$ n/cm²sec);

FR - fast Burner-reactor ($F = 10^{15}$ n/cm²sec);

MSR - molten salt Burner-reactor ($F = 10^{16}$ n/cm²sec);

a) incineration of TRU (Pu, Np, Am, Cm) from PWR;

b) incineration of MA (Np, Am, Cm) from PWR (thermal neutron spectrum);

c) incineration of MA (Np, Am, Cm) from PWR (epithermal neutron spectrum);

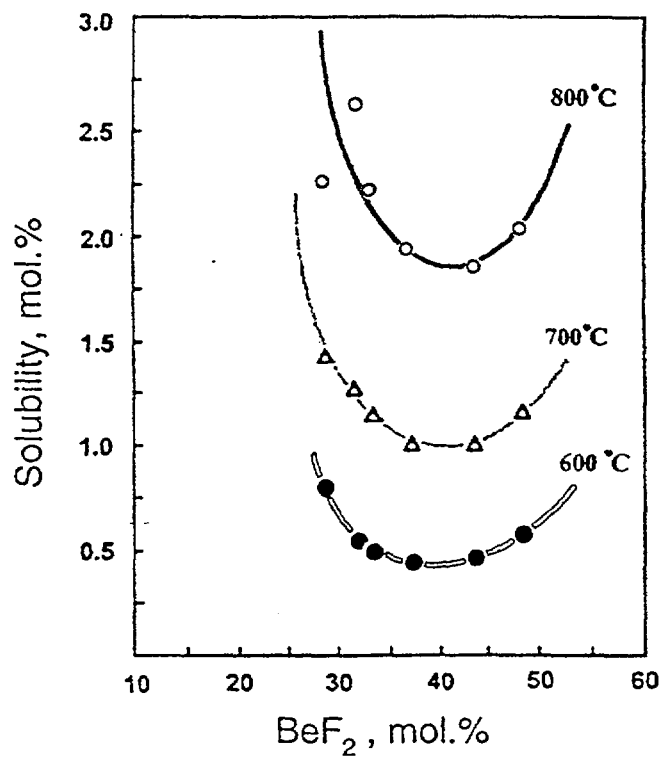


Fig. 3. PuF_3 solubility in LiF-BeF_2

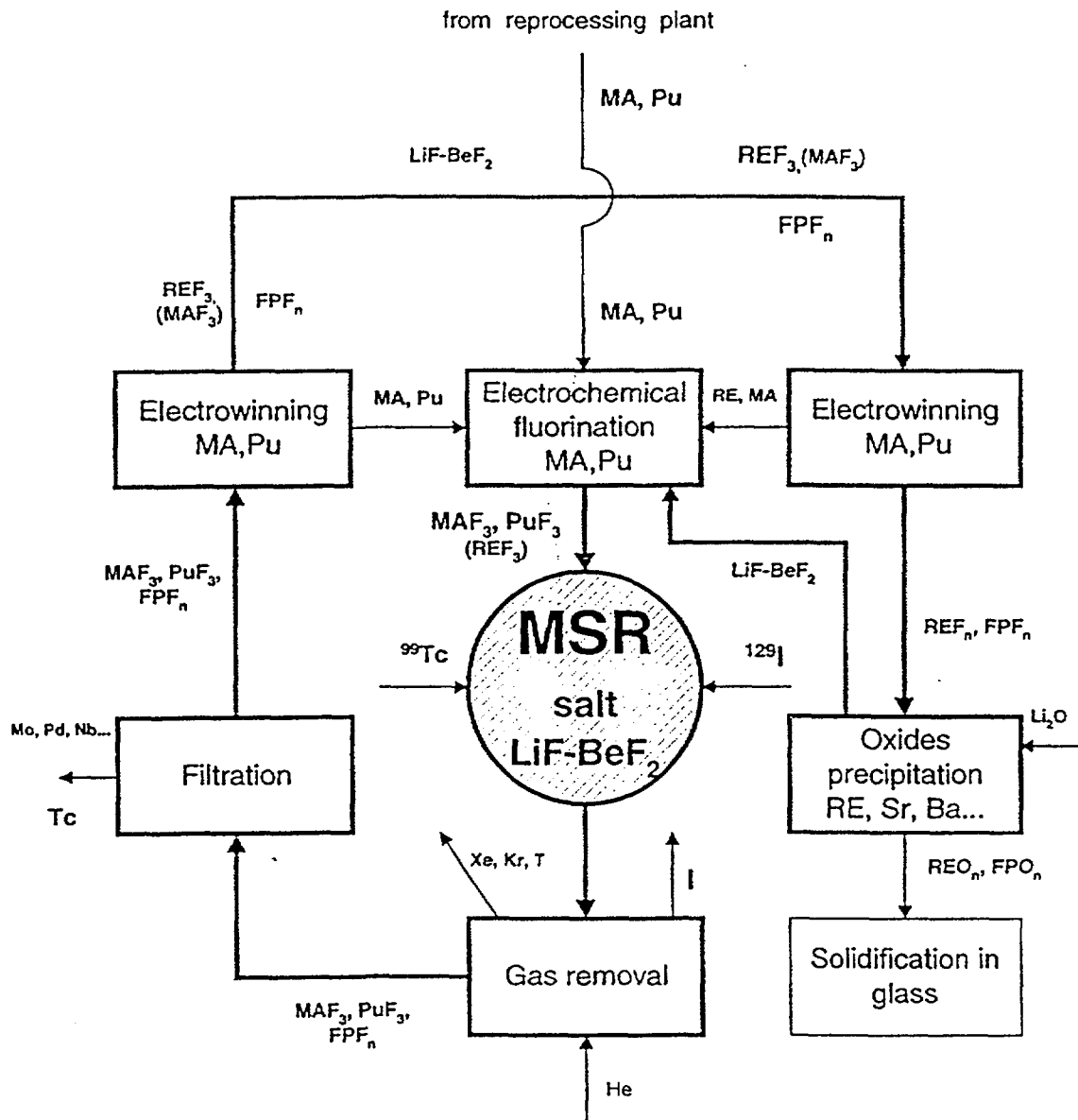


Fig 4. Conceptual flow sheet of MA transmutation and MSR fuel treatment

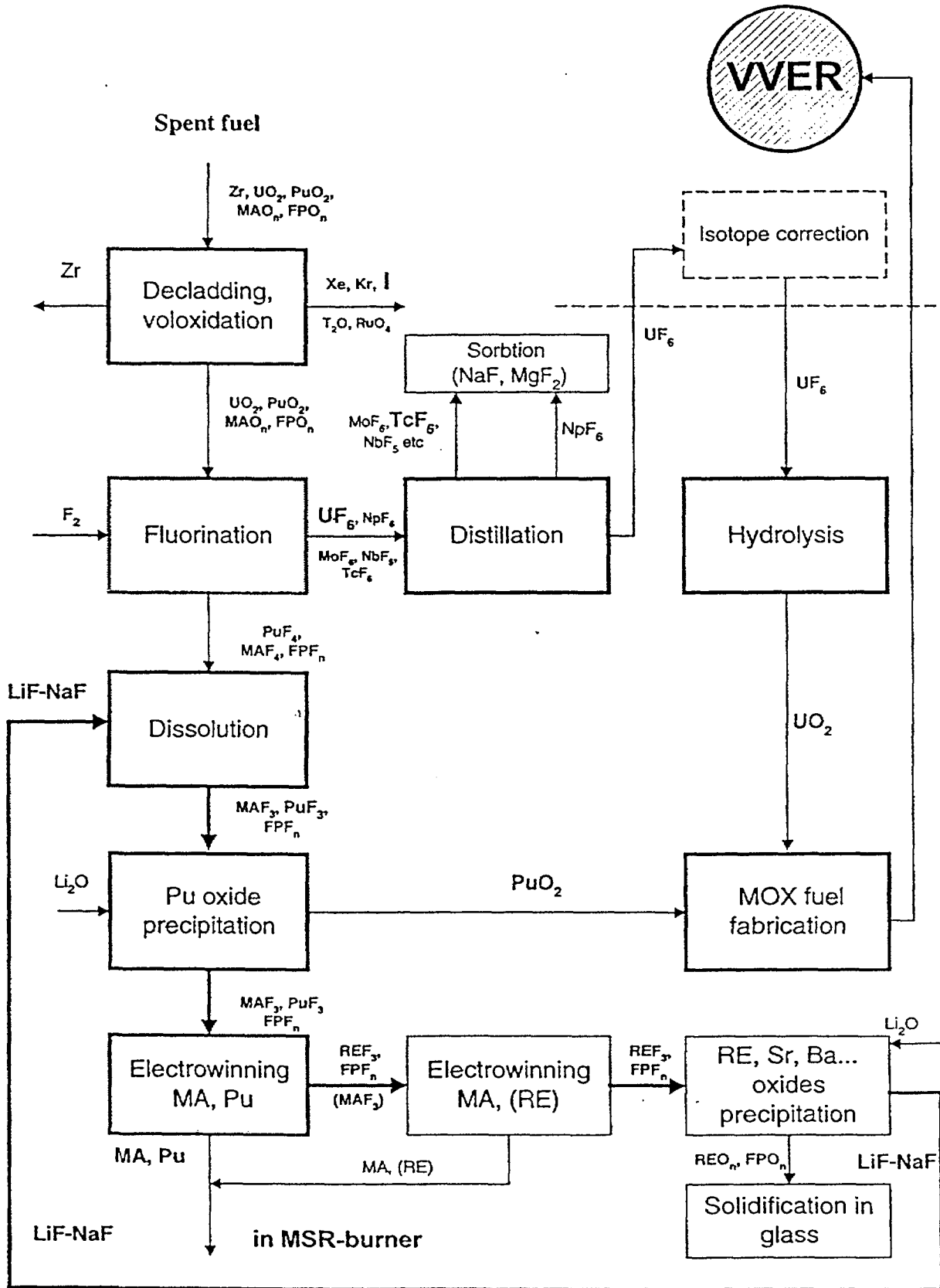


Fig 5. Conceptual flow sheet of VVER spent fuel fluorination and treatment in fluoride melt.

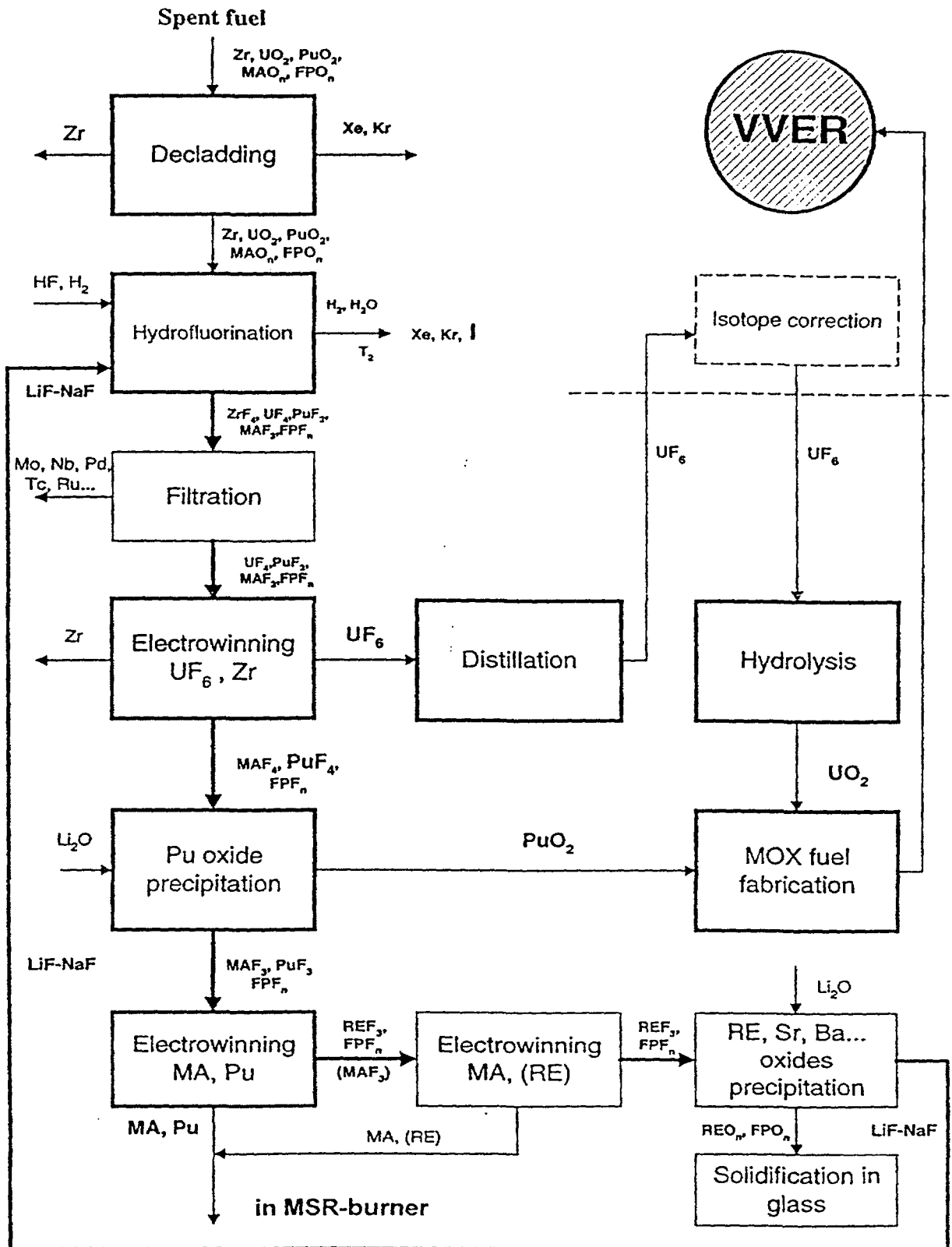


Fig. 6. Conceptual flow sheet of VVER spent fuel electrochemical fluorination and treatment in fluoride melt.