MOLTEN SALT FAST REACTOR IN GENERATION IV: PROLIFERATION CHALLENGES

V. V. IGNATIEV
NRC "Kurchatov Institute",
Moscow, the Russian Federation
Email: Ignatev_VV@nrcki.ru

O.S. FEYNBERG
NRC "Kurchatov Institute",
Moscow, the Russian Federation

M. ALLIBERT
CNRS-IN2P3-LPSC, UGA, Grenoble INP, France

S.DELPECH
CNRS-IN2P3-IPN Orsay, France

D. GERARDIN
CNRS-IN2P3-LPSC, UGA, Grenoble INP, France

D. HEUER
CNRS-IN2P3-LPSC, UGA, Grenoble INP, France

A.LAUREAU
CNRS-IN2P3-LPSC, UGA, Grenoble INP, France

E. MERLE
CNRS-IN2P3-LPSC, UGA, Grenoble INP, France

Abstract

The Molten Salt Reactor designs, where fissile and fertile materials are dissolved in the liquid salt fluorides /chlorides, under consideration in the frame work of the Generation IV International Forum, are briefly described, including MSR activity in Europe and the Russian Federation focused mainly on liquid fuel fluoride based systems with homogeneous cores. This paper has the main objective of presenting some proliferation challenges for the large power reactor plants: (1) Li,Th,U/F Molten Salt Fast Reactor (MSFR) and fertile free Li,Be/F MOlten Salt Actinide Recycler & Transmuter (MOSART) fueled with different compositions of transuranic elements from spent VVER fuel.

1. INTRODUCTION

Historically molten salt reactor (MSR) concept is based on a philosophy that presuppose the use of fuel in the form that permits continuous management of nuclear-physical, chemical and heat transfer processes in fuel and regulation of its nuclides content. This degree of freedom can be used for optimization of nuclear energy system and gaining maximal profits from physical potential of nuclear phenomenon, but also can advantageously or disadvantageously affect the fuel material attractiveness for nuclear weapon proliferation.

Within the Generation IV International Forum MSR pSSC (provisional system steering committee), research is performed under an MOU signed by Euratom, France, the Russian Federation, Switzerland, the United States and Australia, with Canada, China, Japan, and South Korea as observers. The mission of the MSR pSSC is to support development of new concepts that have the potential to provide significant safety and economic improvements over existing reactors [1].

Recently, a wide variety of MSRs, in which a flowing molten salt mixture based on fluorides or chlorides contains fissile/fertile material serving as fuel and coolant, have begun development around the world [2]. The proposed new MSRs have widely varying fuel cycles to the point that identifying common elements is challenging. Proposed neutron spectra range from very thermal to very fast and also include time varying spectra. Almost every known form of fissile / fertile material or fuel cycle is under consideration as a fuel source [1,2].

The reference US ORNL designs of a breeder (MSBR) and denatured (DMSR) MSRs developed in the 1970s clearly illustrate disadvantage and advantages in terms of fuel material attractiveness for the case of U-Th fuel cycle [3,4]. The low material attractiveness DMSR fuel cycle has been incorporated in some of the new MSR designs [1]. Fuel salt additions in MSRs become part of a homogeneous mixture upon being added to operating fuel, so bred fissile isotopes cannot be readily chemically separated from non-fissile isotopes. Isotopes with higher fission cross sections tend to preferentially burn out lowering the fissile actinide fraction generating deep-burn fuel over time. Development of a lower fissile fraction plutonium isotopic composition tends to happen faster in thermal spectrum reactors because their fissions are dominantly generated from fissile materials whereas the fast spectrum systems consume both fissile and fissionable nuclei.

Several of the prospective vendors [1] have indicated that they intend to employ a Th – ^{233}U breeding equilibrium fuel cycle. Breeding ^{233}U from thorium, however, continues to involve creating a separated stream of ^{233}Pa that is allowed to decay in a lower thermal flux region. Separated ^{233}Pa has high material attractiveness.

Not many of the new reactor developers provides information on where the fuel salt synthesis is intended to occur. Also, no information is currently available on the transport or storage of initial fuel salt loads. This will be especially important for fast spectrum reactors, which will require substantial quantities of fissile materials. Very little plant layout information is available for those reactors that include on-site chemical separations.

The large number of MSR concepts combined with multiple options makes in-depth proliferation resistance evaluation difficult. Most of the designs remain at the concept study or lab scale development phase. Even for the concepts driven by private companies, proprietary restrictions on design information limits the accuracy of any evaluation using only public data. Non-reactor portions of the fuel cycle such as fuel preparation, storage and transport, as well as mid-term waste management, for which almost no public information exists, need to be included for proliferation resistance evaluations to be useful.

This paper discusses proliferation relevant features of new MSR developments in Russia on the 1 GWe molten salt actinide recycler and transmuter (MOSART) [5,6] and in Europe on the 1.4 GWe thorium molten salt fast reactor (MSFR) [7-11] address the concept of large power units with a fast neutron spectrum in the non-moderated core. The main characteristic of both designs is the fuel in the form of a molten salt. This fuel salt plays the role of coolant as well: it circulates in the fuel circuit where it heats up in the core due to nuclear reactions before being cooled down in the heat exchangers.

The MSFR can be started either directly with ^{233}U as fissile matter or with a mix of enriched Uranium (13%), Plutonium and Minor Actinides from PWRs spent fuel [11]. The single fluid MOSART starts with the TRUs from PWR spent fuel without Th-U support.

Some lessons learned from the prior US ORNL MSBR program, are reflected in these advanced designs: (1) strong negative reactivity coefficient is the inherent feature of the non-moderated cores; (2) fuel salt processing time for soluble fission products is extended up to 1-2 years.

Both MSFR and MOSART plants include three main circuits involved in power generation (fuel circuit, intermediate circuit and power conversion circuit). These circuits are associated to other systems composing the whole power plant: an emergency draining system, a routine draining system and storage areas, and bubbling and chemical processing units located on site.

2. PROLIFERATION RESISTANCE ANALYSIS APPLIED TO THE MSFR CONCEPT

2.1. Context of the Proliferation Resistance MSFR studies

The design of the MSFR concept is under development [7-8]. The idea to carry out many partial analyses on topics such as Safety and Proliferation Resistance (PR), to define constrains that should be fulfil in its final design. This is a way of getting Safety-by-design and Proliferation-Resistance-by-design instead of adding relevant features afterward, which is usually more expensive. By doing so the analysis cannot be complete but allows an early detection of potential problems: it is a gradual approach. The first PR case studied for the MSFR and presented here has been focused on the threat “concealed diversion of material by a host state having unlimited means”. This case appeared to be the most dangerous one.

By applying the GIF methodology to this case, we successively identify the Elements of the nuclear power site, we identify the Targets for material diversion and the Pathways to achieve diversion, and we suggest Countermeasures to prevent this. This corresponds to the designer’s work and do not contain risks evaluation.

The data provided here-after correspond to a so called “Reference Reactor” arbitrarily chosen for the studies carried out during the Euratom SAMOFAR (Safety Assessment of the Molten Salt Fast Reactor) project of the Horizon 2020 program.

2.2. Elements of Nuclear Power Plant site

The option chosen for the present PR analysis is to consider a country with a limited number of nuclear sites providing large power. In this case the NPP site could contain several reactors sharing common facilities such as the fuel cleaning unit where small amounts of fuel salt are processed to remove part of the fission products and where bred ^{233}U is extracted from fertile salt to feed the on-site reactors. Due to the penetrant 2.6 MeV gamma radiation any nuclear material transfer has to be done by remote handling. Each of the site elements are connected to each other through monitoring chambers allowing an international control of the transfers. This scheme is presented in Fig. 1.

The goal is to determine which material could be an attractive target for concealed diversion and in which element it is located. For this, an inventory of the most attractive isotopes is presented in Table 1 for two starting conditions [9] (^{233}U started reactor and ^{enr}U+TRU started reactor after one year of operation) and for the common composition at steady state (i.e. 200 years after starting).

The concentration of ^{238}Pu in the (enrU+TRU) started version of the MSFR stays larger than 5% in any case, making this Pu not suitable for weapon material. The PR analysis thus concentrates on the ^{233}U started scenario in the following.

The concentration of ^{232}U in the U that can be isolated from the salts is always high rendering ^{233}U unattractive for nuclear weapon material because of intense gamma radiation. This statement is common place for Th/U based nuclear cycle and diversion of U has not been considered here. This is justified because Pa diversion potentially leads to a much better material from this point of view. In conclusion, the diversion of radioactive material contained in the reactor core seems impossible, so that we have considered only the possibilities for nuclear material diversions within the chemical processing unit located on site.

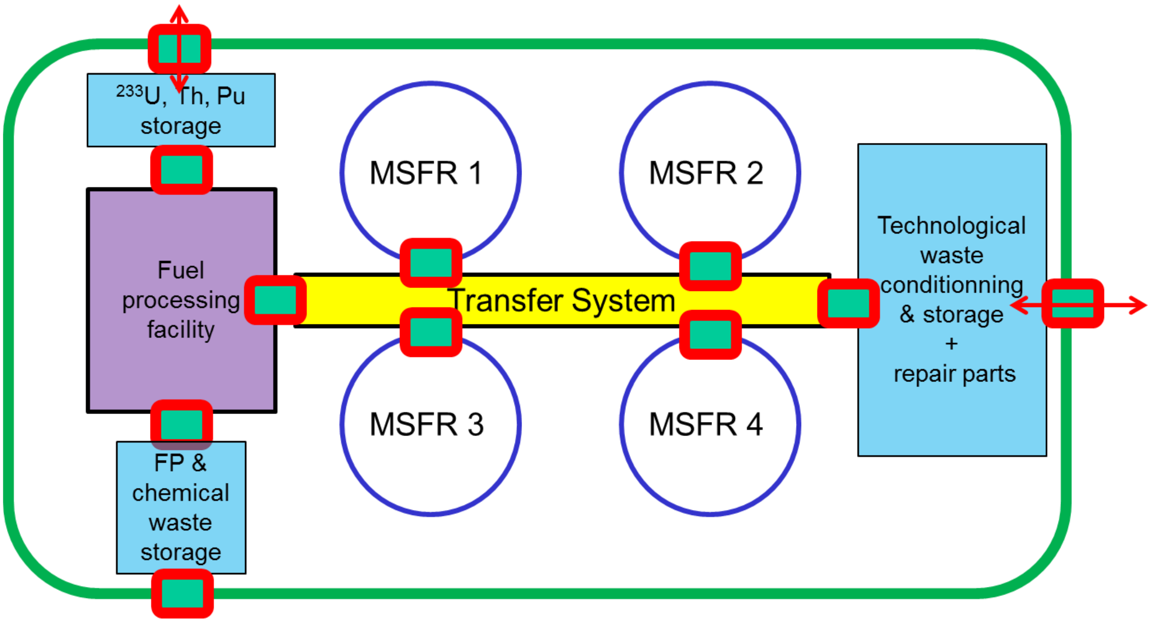

FIG. 1. Schematic representation of a nuclear site with 4 reactors sharing common facilities. Green rectangles with red contours represent monitoring chambers for any transfer in or out the elements. Internal transfers on site are made by remote handling (yellow).

TABLE 1. MAIN ISOTOPIC INVENTORY FOR A 3 GWt MSFR (IN Kg OTHERWISE STATED)

Isotope	^{233}U -started after 1 year	^{enr}U+TRU -started after 1 year	Fuel salt steady state 200 years	Fertile salt
^{232}U	3.5	142 g	13	34 g
^{233}U	4976	514	4658	58.5
^{238}U	0	16300	1	0
^{232}U/U	700 ppm	50 ppm	1700 ppm	600 ppm
^{233}U/U	97%	2.7%	62%	99%
^{238}Pu	0	239	161	0
^{239}Pu	0	3265	66	0
^{240}Pu	0	1617	57	0
^{241}Pu	0	641	48	0
^{242}Pu	0	491	10	0
^{239}Pu/Pu		52 %	19 %	
^{232}Pa	3.9 g	0	15 g	15.4g
^{233}Pa	124	45.6	108	13

2.3. Pathway to Pa diversion

Fuel contains more than 100 kg of ^{233}Pa in 18 m^3 of salt and blanket contains 13kg of this isotope in 7.7 m^3 of fertile salt (“Reference Reactor”). The assumption is that Pa can be isolated by a chemical processing taking place in the cleaning unit. If this possibility exists, one can wait until ^{232}Pa decayed into ^{232}U, separate the remaining ^{233}Pa from the other isotopes to get rid of 2.6 MeV gamma rays in order to get a weapon grade ^{233}U. After 3 weeks of storage the fraction of ^{233}Pa remaining is 58% but the amount of ^{232}Pa has been reduced by a factor 4 10^4. After 4 weeks this factor becomes 1.5 10^6 and the remaining ^{233}Pa is still 49% of the initial amount. In such condition getting one Significant Quantity of ^{233}U (8 kg) would require the processing of about 9 m^3 of fertile salt or 2.7 m^3 of fuel salt.

Eliminating the 2.6 MeV gamma ray emission of ^{208}Pb requires the separation of Pa from all the descendants of half-life larger than the duration of Pa exfiltration from the site. The decay scheme is presented in Fig 2.

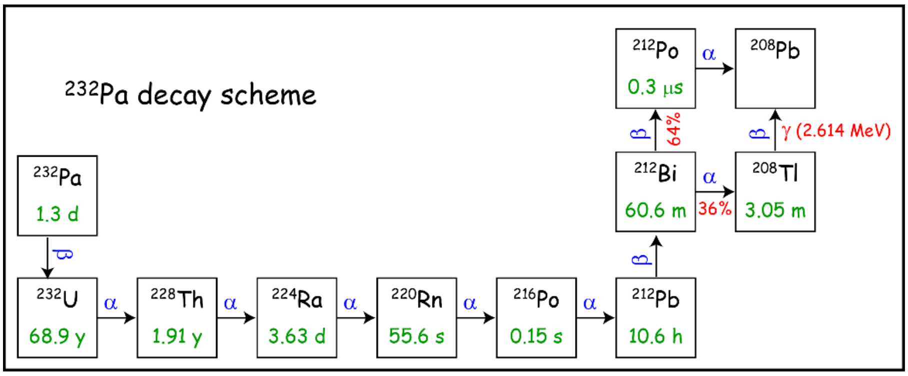

FIG. 2. Decay scheme of ^{223}Pa showing the elements that should be separated from Pa to eliminate ^{208}Pb production and its 2.6 MeV gamma ray emission

The most effective technique to achieve Pa isolation from other elements seems to be fluorination as volatilization of UF_6 and PaF_5 may be obtained under different fluorination conditions or volatilization temperatures.

Reduction of Pa and U into metallic solvent, such as liquid Bi, are much less effective and represent no threat of proliferation.

2.4. Countermeasures

Apart from the detection by isotope balance or abnormal transfer of material, specific attention is to be paid to the cleaning unit. Indeed, Pa diversion requests complex diversion of fluorination equipment use, a storage of ^{232}U containing Pa during 3 to 4 weeks in a concealed space and abnormal transfers from this storage space to the fluorination equipment. This means that the cleaning unit conception has to prevent such misuses.

Means to avoid such nuclear matter diversion take advantage of the MSFR's flexibility. A proliferation risk analysis can thus lead to recommendations on the design or operating mode of a future reactor. Such recommendations will be used to attribute a proliferation resistance weight to each design option.

3. MOSART REACTOR

Facilities of Experimental Demonstration Centre (EDC) being built in Russia at the site of the Mining and Chemical Combine (MCC) after 2020 will begin reprocessing of spent nuclear fuel (SNF) from VVER-1000 reactors on the basis of innovative technology, providing a recovered nuclear material (refined products) for recycling in thermal and fast solid fuel reactors [12] After adjustment of all technological processes EDC will become the reference basis for a large-scale RT-2 plant, which will provide an environmentally and economically acceptable system of VVER-1000/1200 SNF recycling both in the Russian Federation and abroad. In accordance with the EDC flowsheet, the highly active raffinate, containing long-lived minor actinides, is sent for conditioning. The obtained vitrified HLW belong to the first class of radwaste. Use of dedicated reactor unit as a TRU incinerator, remaining after the main part of uranium and plutonium recycling to solid fuel thermal and fast reactors, may reduce the volume and radiotoxicity of HLW. The principle attraction of the MSR technology in this application is the use of fuel material flexibility (easy of “short cooled” fuel preparation, processing and multiple recycling) for gaining additional profits as compared with solid materials.

It is proposed to use the technical and technological capabilities of the MCC site to place single fluid fertile free MOSART system in the immediate vicinity of SNF aqueous reprocessing plant, linking it to the EDC infrastructure. The main design objective of the single fluid 2.4 GWt MOSART is to close nuclear fuel cycle for all actinides, including Np, Pu, Am and Cm. It is assumed that the fuel cycle of this complex will be organized as follows (see Figure 1): the bulk of the removed uranium and plutonium return to thermal and fast solid fuel reactors, and the remaining TRUs are transferred for utilization in the MOSART system. The co-location of MOSART and SNF reprocessing plant, will provide the MCC site and the surrounding customers by electricity (7.92 TWe·h), facilitates the problems of nuclear materials transport and radwaste management. Remind, that with fluid fluoride based fuel the entire fuel element fabrication process is excluded. The absence of a solid fuel manufacturing phase provides for exceptional flexibility. The fuel can be blended into reactor straightly as needed at any time. Also, there is no need for long cool times and interim storage. This saves for MOSART significant part of the head end effort (including radioactive doses) and cost. All fresh fuel fluorides containing significant quantities of fissile materials for initial loading and make up, will be manufactured onsite at the EDC by hydrofluorination process.

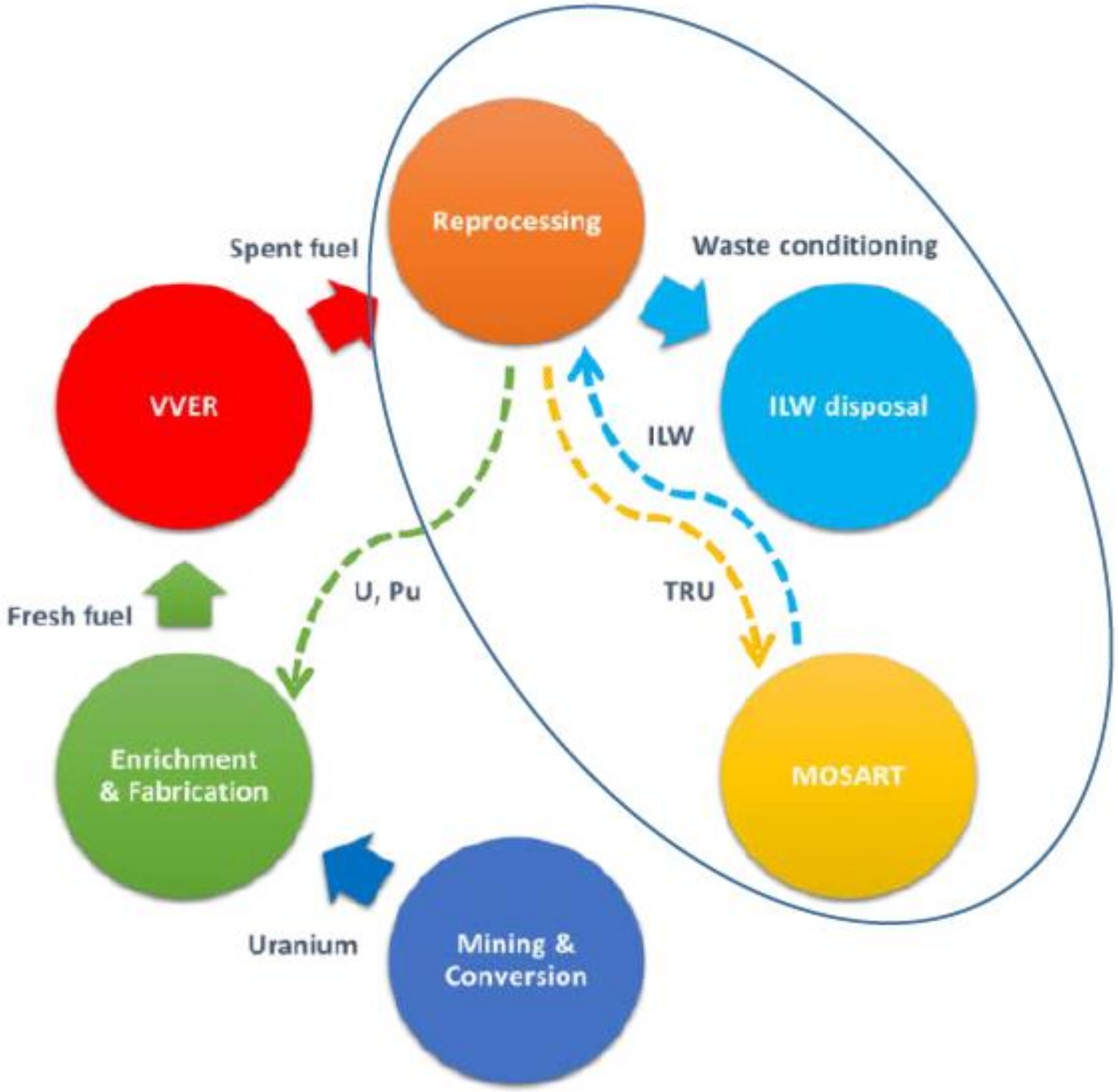

Fig. 3: Nuclear fuel cycle with MOSART at MCC site

The effective neutron flux of 2.4 GWt MOSART is near $1x10^{15}$ n cm^{-2} s^{-1}. The possibility of creating a high neutron flux and the lack of structural materials in the liquid homogeneous core, leads to optimization of the neutron balance, as well as the possibility to change the fuel salt composition without core modification and reactor shutdown, creates favorable conditions for the TRU utilization. The MA burning rate is directly proportional to the core specific power. When choosing this parameter, it is advisable to be within technical limits.

In the MOSART, a well-established molten LiF-BeF_2 salt mixture, is chosen as a solvent for TRU trifluorides fuel addition. The molten fluoride chemistry (solubility, redox chemistry, chemical activity etc) for the LiF–BeF_2 system is well established and can be applied with great confidence, if TRU trifluorides are to be dissolved in the LiF–BeF_2 solvent. The solubility of TRU trifluorides in molten 73LiF-27BeF_2 (in mole %) salt mixture with decreased beryllium difluoride fraction of 0.27 and the minimum temperature in the fuel circuit of 600 ° C is more than 2 mole % [7]. The structural material selected for the MOSART container is the special Ni-Mo alloy of kH80MTY type with a low concentration of Cr alloyed by 1% of Al [13]. The composition of the alloy is optimized for corrosion resistance (both in a low oxygen gas atmosphere and in molten salt fluorides), irradiation resistance and high temperature mechanical properties.

The performed calculations show that the 2.4 GWt MOSART, starting with TRUs from SNF of VVER-1000 with the ratio of MA to (Pu + MA) equal 0.1 or 0.15 (see Table 2), without core modification and changing temperature in the fuel circuit, can use any TRUs make up with the MA to (Pu + MA) ratio up to 0.33. For scenarios 1 and 2 from Table 2 initial core loading of TRUs required will be 3.5 t and 4.5 t, respectively. Core loading of TRUs at equilibrium will be varied from 3.6 t till to 18.0 t depending from the MA to (Pu + MA) ratio in the core make up. 2.4 GWt MOSART with a fuel salt selected can utilize up to 250 kg of MA and about 500 kg of Pu per year (in the case of make up with 33% of MA) [3]. Note, that after each next recycling fuel is getting less attractive for fissile material diversion. During 50 yrs of operation 2.4 GWt MOSART can utilize more than 12 t of MA. Last TRU loading will be transferred to the next MOSART unit to be constructed at the MCC site.

TABLE 2: ISOTOPIC MASS PROPORTION IN INITIAL MOSART LOADING, IN MASS. %

Isotope	Scenario 1	Scenario 2
Np^{237}	6,42	6,51
Np	6,42	6,51
Pu^{238}	3,18	2,77
Pu^{239}	43,93	48,36
Pu^{240}	21,27	19,97
Pu^{241}	13,52	8,30
Pu^{242}	7,88	6,25
Pu	89,78	85,65
Am^{241}	0,55	5,56
Am^{243}	2,33	1,69
Am	2,88	7,25
Cm	0,92	0,59
Total	100	100

In general, to achieve fuel salt maintenance, (1) the fuel must be delivered to and into the reactor in a proper state of purity and homogeneity, (2) the fuel must be sufficiently protected from extraneous impurities, and (3) sound procedures must exist for addition and recycling of the actinides required and (4) provision of the required redox potential in the system. For MOSART that propose fuel salt processing to remove fission products (see Table 3), the required fuel maintenance operations also include (1) continuous removal (by the sparging and stripping section of the reactor) of fission-product krypton and xenon, (2) addition of TRUs to replace those lost by burnup, (3) recycling of all actinides, (4) removal of soluble fission products (principally rare earths); they probably also include (5) removal of inadvertent oxide contaminants from the fuel; in addition, they may include (6) removal of a portion of the insoluble noble and semi noble fission products.

For multiple TRUs recycling, since plutonium and minor actinides must be removed from the fuel solvent before yttrium and rare earth's fission products, the MOSART must contain a system that provides removal of TRU's from the fuel salt and their reintroduction to the purified solvent. This plutonium reintroduction circuit has the advantage of also returning americium and curium to the reactor fuel (low separation coefficients within TRU group). Since the higher actinides would always accompany the plutonium, this operation would never produce a “clean” material would be attractive for diversion. The molten Li,Be/F salt mixture, due to the high separation coefficients between actinides and lanthanides, make it possible to organize an effective removal of soluble fission products, based on the reductive extraction, to substantially reduce the time of the external fuel cycle for actinides and its losses in waste stream in multiple recycling in comparison with solid fuel reactors.

Interior Ni-base reflectors / shielding are employed to reduce the radiation damage to the reactor vessel and fuel salt chemistry control is employed to substantially limit oxidizing the container alloy constituents. For those systems that include graphite, the salt is maintained in a redox window oxidizing enough to prevent damage to the graphite while reducing enough to prevent alloy oxidative corrosion. Those systems without graphite can maintain the salt in a more reducing condition.

The radiation resistance of reflectors / shielding material determines the upper limit core specific power in the MOSART design [6]. If the damage caused by fast neutrons is critical for graphite, then for high-nickel alloys, the reduction in plasticity at a temperature above 500°C, associated with the formation of helium along the grain boundaries, is the most important process, caused by both fast and thermal neutrons. To obtain an acceptable service life for reflectors (> 5 yrs), the core specific power should not exceed 130-150 W/cm^3. Otherwise, frequent stops to replace the reflector will result in a reduction in the reactor load factor and an unjustified increase in operating costs. In addition, with this limitation for the core specific power, there is no problem of heat removal from the fuel circuit. In this case the service life for the reactor vessel, made of the kHN80MTY alloy, will be about 50 yrs. For 2.4 GWt MOSART, taking into account the adopted limits, the primary circuit will contain 50 m^3 of fuel salt, of which only half is in the core.

The main advantages of MOSART are the ability to vary widely the MA content in fuel salt without losing the inherent safety and the absence of stages related to the fuel fabrication and re-fabrication in multiple actinides recycling. As result there are significant PR and safeguards implications related to the fuel make up and chemical processing in MOSART plant: (1) there will be continuous variation of isotopic concentrations in the fuel salt from both TRU transmutation and chemical processing; (2) refueling scheme include the ability to continuously feed the core with fresh fissile material; (3) plate-out of noble metals in the primary circuit could complicate inventory tracking.

Fuel salt represents a unique combination of high-temperature and high-radiation environments that will be challenging for diversion as well as measurement techniques and instrumentation: (a) temperature in the reactor or fuel processing plant will always be kept in liquid state within 550 -720^{0}C; and (b) fuel salt will be highly radioactive even outside the primary circuit.

TABLE 3: SUMMARY TIMES FOR FISSION PRODUCT REMOVAL AND ACTINIDES RECYCLING [6]

Element	Time
Kr, Xe	50 s
Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, Te	2-4 hrs
Zr	1-3 yrs
Ni, Fe, Cr	1-3 yrs
Pu, Am, Cm, Np, U	1-3 yrs
Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er	1-3 yrs
Sm, Eu	1-3 yrs
Sr, Ba, Rb, Cs	5-10 yrs
Li, Be, Th	30 yrs

In order to avoid nuclear matter diversion MOSART reactor plant is integrated (1) at the front end with VVER SNF aqueous reprocessing plant and (2) at the back end with the high temperature fuel salt clean up facility all located at the MCC site. All fresh fuel fluorides containing significant quantities of fissile materials (Pu+MA) for initial loading and make up, will be manufactured onsite by hydrofluorination process. In molten salt pyroprocessing facility the higher actinides would always accompany the plutonium, this operation would never produce a "clean" material would be attractive for diversion. Last TRU loading will be transferred to the next MOSART reactor plant to be constructed at the MCC site

4. CONCLUSION

Some preliminary PR analyses performed on molten salt reactors have been presented here. The idea is to include the conclusions of such studies at an early stage of the design of such systems to guide the selection of the best design options. MSR designs under considerations, in which a flowing molten salt mixture based on fluorides contains fissile / fertile material serving as fuel and coolant, represent reactor plant integrated with pyro processing unit. Existing IAEA PR and safeguards approaches are mainly prepared for the U-Pu, but not for MA-based and Th-U fuel cycles, item counting for solid fuel reactors, and bulk material accountancy for the front and back end of the nuclear fuel cycle. These techniques and associated instrumentation for bulk accountancy have been developed predominantly for enrichment, fuel fabrication, and aqueous reprocessing. However, none of these bulk accountancy measures can be directly applied to high temperature MSR designs, in general, and for MSFR and MOSART in particular, without evaluation and potential modification. Almost no work has been done to determine how will the fissile material content in the fuel salt mixture be determined, when it is in the primary circuit, in the draining/storage tanks, or in the processing units. This will guide the next steps of the PR studies of such concepts.

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