The Molten Salt Reactor (MSR) in Generation IV: Overview and perspectives

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I. INTRODUCTION

In a Molten Salt Reactor (MSR), the fuel is dissolved in a fluoride salt coolant. The technology was partly developed, including two demonstration reactors, in the 1950's and 1960's in USA (ORNL). Compared with solid-fuelled reactors, MSR systems have lower fissile inventories, are insensitive to fuel radiation damage that can limit fissile and fertile material utilization, provide the possibility of continuous fission-product removal, avoid the expense of fabricating fuel elements, give the possibility of adding makeup fuel as needed, which precludes the need for providing excess reactivity, and employ a homogeneous isotopic composition of fuel in the reactor. These and other characteristics may enable MSRs to have potentially unique capabilities and competitive economics for actinide burning and extending fuel resources.

Prior MSRs were mainly considered as thermal-neutron-spectrum graphite-moderated concepts. Since 2005, R&D has focused on the development of fast-spectrum MSR concepts (MSFR) combining the generic assets of fast neutron reactors (extended resource utilization, waste minimization) to those relating to molten salt fluorides as fluid fuel and coolant (favourable thermal-hydraulic properties, high boiling temperature, optical transparency). In addition, MSFR exhibit large negative temperature and void reactivity coefficients, a unique safety characteristic not found in solid-fuel fast reactors. [4-8] MSFR has been recognized as a long term alternative to solid-fuelled fast neutron systems with unique potential (negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle…).

Apart from MSR systems, other advanced reactor concepts are being studied, which use the liquid salt technology, as a primary coolant for the Advanced High-Temperature Reactor (AHTR)[11] or intermediate coolant, as an alternative to secondary sodium, for Sodium Fast Reactors (SFR) and to intermediate helium for Very High Temperature Reactors (VHTR).

More generally, the development of higher temperature salts as coolants would open new nuclear and non-nuclear applications. These salts could also facilitate heat transfer for nuclear hydrogen production concepts, concentrated solar electricity generation, oil refineries, and shale oil processing facilities amongst other applications. [3]

In brief, there has been a significant renewal of interests for liquid salt applications.

The paper shows the main technical progress achieved in the countries participating to the R&D effort on the MSR in GIF and remaining issues to be addressed.

II. MSR IN GENERATION IV

A decision to establish a Provisional System Steering Committee (PSSC) for the MSR was taken by the GIF Policy Group in May 2004. The participating members are EURATOM, France and the United States. Other countries have been represented systematically (the Russian Federation) or occasionally (Japan) as observers in the meetings of the PSSC. Russia has played an important role in identifying R&D issues basing on long-lasting R&D programs initiated the 1970s.

The renewal and diversification of interests in molten salts have led the MSR PSSC to a shift of the R&D orientations and objectives initially promoted in the original Generation IV Roadmap issued in 2002, [1] in order to encompass in a consistent body the different applications envisioned today for fuel and coolant salts. [2]

Two baseline concepts are considered which have large commonalities in basic R&D areas, particularly for liquid salt technology and materials behavior (mechanical integrity, corrosion):

- The Molten Salt Fast neutron Reactor (MSFR) is a long-term alternative to solid-fuelled fast neutron reactors offering very negative feedback coefficients and simplified fuel cycle. The potential of MSFR has been assessed but specific technological challenges must be addressed and the safety approach has to be established.
- The Advanced High Temperature Reactor (AHTR) is a high temperature reactor with higher power density than the VHTR and passive safety potential from small to very high unit power (> 2 400 MWt).

In Russia, the efficiency of MSR for actinide burning has been investigated. This resulted into the single stream Li, Na,Be/F MOlten Salt Actinide Recycler & Transmuter (MOSART) fast spectrum system fuelled with compositions of plutonium plus minor actinide trifluorides (AnF_3) from UOX and MOX LWR spent fuel without U-Th support. [13]

In addition, the opportunities offered by liquid salts for intermediate heat transport in other systems (SFR, LFR, VHTR) are being investigated. Liquid salts offer two potential advantages: smaller equipment size because of the higher volumetric heat capacity of the salts; and no gross chemical exothermal reactions between the reactor, intermediate loop, and power cycle coolants.

Liquid salt chemistry plays a major role in the viability demonstration of MSR and AHTR concepts with such essential R&D issues as: (a) the physico-chemical behaviour of coolant and fuel salts, including fission products and tritium, (b) the compatibility of salts with structural materials for fuel and coolant circuits, as well as fuel processing materials development, (c) the on-site fuel processing, (d) the maintenance, instrumentation and control of liquid salt chemistry (redox, purification, homogeneity), and (e) safety aspects, including interaction of liquid salts with sodium, water, and air.

The factorization into projects in the SRP emphasizes cross-cutting R&D areas. A major commonality is the understanding and mastering of fuel and coolant salts technologies, including development of structural materials, reliable knowledge on physical properties for fuel and coolant salts, fuel and coolant salts clean-up, chemical and analytical R&D for fuel and coolant behaviour.

III. MSFR REFERENCE OPTIONS

Starting from the ORNL Molten Salt Breeder Reactor project (MSBR), an innovative concept has been proposed [4, 5], resulting from extensive parametric studies in which various core arrangements, reprocessing performances and salt compositions were investigated. The primary feature of the MSFR (Molten Salt Fast Reactor) concept is the removal of the graphite moderator from the core (graphite-free core).

In terms of fuel cycle, two basic options have been investigated, ^{233}U-started MSFR and TRU-started MSFR.

Realistic drawings showing the main MSFR components and their arrangement in the vessel have been elaborated. Figure 1 displays a schematic drawing of a vertical section of the MSFR while Table 1 presents some characteristics of the reactor.

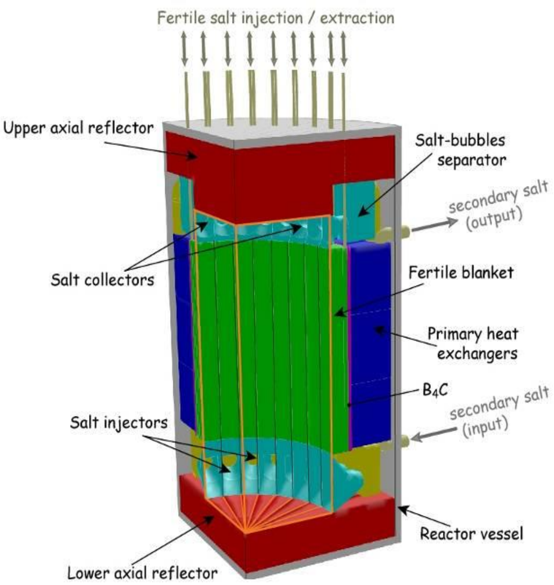

Figure 1: Schematic view of a quarter of the MSFR

The core is a single cylinder (diameter equal to height) where nuclear reactions take place within the flowing fuel salt. It is made of three volumes: the active core, the upper plenum and the lower plenum. The fuel salt is a binary salt, composed of LiF enriched in ^{7}Li (99.999%) and heavy nuclei (HN) amongst which the fissile element, ^{233}U or Pu. The $(HN)F_4$ proportion is set at 22.5 mol% (eutectic point), corresponding to a melting temperature of 550°C. The choice of this fuel salt composition relies on many systematic studies (influence of chemical reprocessing on neutronic behavior, burning capabilities, deterministic safety level, deployment capabilities). [6-10] This salt composition leads to a fast neutron spectrum in the core. The outer core structures and heat exchangers are protected by thick reflectors designed to absorb more than 80% of the escaping neutron flux. These reflectors are themselves surrounded by a 10 cm thick neutronic protection of B_4C absorbing remaining neutrons. Axial reflectors are made of nickel-based alloys. The radial reflector consists of a fertile blanket (50 cm thick) filled with a fertile salt of LiF-ThF_4 with 22.5 mol% ^{232}Th.

The level of deterministic safety reached by the concept is excellent since the feedback coefficients of the MSFR are negative in both ^{233}U and TRU starting modes. [6,8,10] The total feedback coefficient is equal to -6 pcm/°C when the equilibrium state of the reactor has been reached and the density coefficient, which for MSRs can also be viewed as a void coefficient, is also largely negative at about -3 pcm/°C.

Thermal power (MWt)	3000				
Fuel molten salt composition (mol%)	LiF-ThF_4-$^{233}UF_4$ or LiF-ThF_4-(Pu-MA)F_3 with LiF = 77.5 mol%				
Fertile blanket molten salt composition (mol%)	LiF-ThF_4 (77.5-22.5)				
Melting point (°C)	550				
Operating temperature (°C)	700-800				
Initial inventory (kg)	^{233}U-started MSFR		TRU-started MSFR		
	Th	^{233}U	Th	Actinide	
	38300	5060	30600	Pu	11200
				Np	800
				Am	680
				Cm	115
Density (g/cm^3)	4.1				
Dilatation coefficient (/°C)	10^{-3}				
Core dimensions (m)	Radius: 1.15 Height: 2.30				
Fuel salt volume (m^3)	18 9 out of the core 9 in the core				
Blanket salt volume (m^3)	8				
Thorium consumption (ton/year)	1.112				
^{233}U production (kg/year)	93 (^{233}U-started MSFR) 188 during 20 years then 93 (TRU-started MSFR)				
Breeding ratio (^{233}U-started MSFR)	1.085				

Table 1: Reference design characteristics of the MSFR

A good indicator of the deployment capability is the doubling time, defined by the operation time leading to the ^{233}U inventory of a new reactor of the same type through breeding. For a ^{233}U-MSFR, the annual ^{233}U production is 120 kg which corresponds to 50 years doubling time per reactor. [6, 9] Starting a MSFR from Generation II or III reactors spent fuel is more favourable and yields 35 years doubling time. Indeed, the presence of other fissile elements decreases the consumption of ^{233}U and improves the deployment capability of the concept.

IV. AHTR REFERENCE PLANT CONCEPT

The defining aspects of an Advanced High Temperature Reactor (AHTR) are the use of coated particle fuel embedded within a graphitic matrix cooled by liquid fluoride salt. [11] A Pebble Bed Advanced High Temperature Reactor (PB-AHTR) operating at ~900 MWt is the most actively developing commercial scale plant design. [12] The plant design is currently transitioning from a primarily conceptual to an initial engineering scoping phase. A half cross section of the core concept is shown in Figure 2.

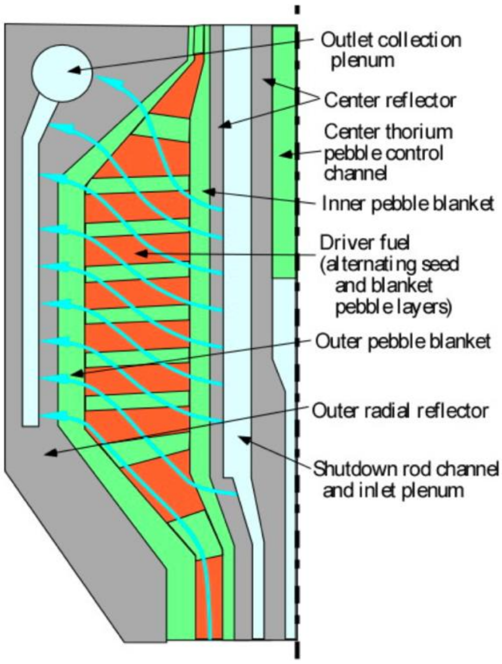

Figure 2: Half Cross Section of PB-AHTR Core

A major design refinement of the current core is the use of inner and outer pebble blankets to reduce the radiation damage to the fixed reflector graphite. The power density of a salt cooled pebble bed is 4-8x greater than that of its gas-cooled cousin. The resultant higher flux level would necessitate more frequent reflector graphite replacement without the use of blanket pebble layer. The controlled motion of a structured pebble assembly has recently been demonstrated using simulant materials at U.C. Berkeley, along with friction coefficient measurements for graphite pebbles verifying that fluoride salts act as effective lubricants and that friction coefficients are very close to those for the simulant materials. Pebble motion demonstration using prototypic materials and temperatures will be a key aspect of future R&D on the PB-AHTR.

V. R&D PROGRESS AND REMAINING ISSUES IN SPECIFIC AREAS

Significant progress has been achieved in 2008 in critical areas of MSR-AHTR R&D. In brief, the essential facts are the following:

1. Salt selection for different applications is stabilized, the needs of complementary data have been clarified. [14, 18]
2. A strongly improved (versus MSBR) fuel salt clean-up scheme has been developed. [8, 15, 16]
3. Criticality tests are being performed for the assessment of MSR and AHTR fuel and core behaviour.

Those topics are the subject of the following sub-sections.

Although progress has been made in the area, the assessment of structural materials remains challenging for MSFR and AHTR as both concepts are supposed to operate at temperatures higher compared to MSBR.

V.A Salt selection for different applications

Potential salt systems have been critically reviewed in the frame of the ALISIA project in the EURATOM 6th FWP. [14] Reference compo–sitions have been proposed or confirmed (Table 2).

Reactor type	Neutron spectrum	Application	Carrier salt	Fuel system
MSR-Breeder	Thermal	Fuel	$^{7}LiF\text{-}BeF_2$	$^{7}LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$
	Non-moderated	Fuel	$^{7}LiF\text{-}ThF_4$	$^{7}LiF\text{-}ThF_4\text{-}UF_4$
				$^{7}LiF\text{-}ThF_4\text{-}PuF_3$
MSR-Breeder	T/NM	Secondary coolant	$NaF\text{-}NaBF_4$	
MSR-Burner	Fast	Fuel	LiF-NaF	$LiF\text{-}(NaF)\text{-}AnF_4\text{-}AnF_3$
			$LiF\text{-}(NaF)\text{-}BeF_2$	$LiF\text{-}(NaF)\text{-}BeF_2\text{-}AnF_4\text{-}AnF_3$
			$LiF\text{-}NaF\text{-}ThF_4$	
AHTR	Thermal	Primary coolant	$^{7}LiF\text{-}BeF_2$	
SFR		Intermediate coolant	$NaNO_3\text{-}KNO_3\text{-}(NaNO_2)$	

Table 2: Fuel and coolant salts for different applications

The $^{7}LiF\text{-}BeF_2$ (66:34 in mol%) salt is the selected fuel carrier for the moderated (thermal) molten salt thorium breeder, giving as fuel salt $^{7}LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$. From neutronic as well as chemical point of view, there are no alternatives for this salt that do not penalise the breeding capacity of the reactor.

$^{7}LiF\text{-}ThF_4$ (78:22 or even 71:29 in mol%) is the reference fuel solvent composition for the fast spectrum molten salt thorium breeder reactor (MSFR). The neutronic analysis of the MSFR concept has demonstrated the feasibility of the concept, but it must still be clarified whether the physico-chemical properties (melting temperatures, solubility for the actinide trifluorides, density, expansivity, viscosity, thermal conductivity, heat capacity) of this salt fuelled by significant amount of UF_4 (2-4% of the total heavy nuclei in the moderated and 12-18% in the fast systems) or AnF_3 (up to 25% of the total heavy nuclei in the fast concept) are consistent with safe operation of the reactor and fuel salt clean-up unit. To tune these properties, addition of other components is possible. The most obvious is BeF_2 but there is an incentive to keep the content of this material low (*e.g.* $71LiF\text{-}2BeF_2\text{-}27ThF_4$ or $75LiF\text{-}5BeF_2\text{-}20ThF_4$ in mol%) or even zero. Alternatives are NaF and possibly CaF_2. Therefore, the $^{7}LiF\text{-}NaF\text{-}ThF_4$ system must be further analysed, whereas scoping studies of the $^{7}LiF\text{-}CaF_2\text{-}ThF_4$ system are required, to assess the pros and cons for both molten salt mixtures, including suitability for fuel salt processing.

The molten salt actinide burner is a fast spectrum concept too. The carrier salt for this application must have good solubility for the actinide trifluorides and this can be achieved using $^{7}LiF\text{-}NaF\text{-}(KF)$ as solvent or $^{7}LiF\text{-}(NaF)\text{-}BeF_2$ melt. Again, the goal is to keep the content of BeF_2 low or even zero. An interesting alternative is the use of plutonium and minor actinides as start-up for the thorium cycle in the MSR, leading to $^{7}LiF\text{-}NaF\text{-}ThF_4$ carrier salt.

In summary, it is clear that the $^{7}LiF\text{-}(NaF)\text{-}AnF_4\text{-}AnF_3$ salt (where An represent actinides) is the key system to be further investigated in parallel to the $^{7}LiF\text{-}(NaF)\text{-}BeF_2\text{-}AnF_4\text{-}AnF_3$ system. Optimisation of the fractions of the components is still needed with respect to mentioned-above physico-chemical properties, corrosion behavior in the Ni-Mo alloys and fuel salt processing.

For coolant salts, one has to make a distinction between salt for in-core use (primary coolant) and salts for out-of-core use (secondary or intermediate coolants). For primary coolants in thermal reactors, the requirements are very similar to thermal breeder reactors and $^{7}LiF-BeF_2$ (66-34 with Tm=458°C) is the main candidate, with $^{7}LiF-NaF-KF$ (46-11.5-42.5 with Tm=454°C), LiF-NaF-RbF (46.5-6.5-47 with Tm=426°C) and $^{7}LiF-NaF-BeF_2$ (30.5-31-38.5 with Tm=316°C) as alternatives. Note that the last alternative molten salt mixture has the lowest liquidus temperature.

For secondary coolant applications, neither neutronic considerations nor actinide solubility play a role and a wider choice of materials is possible. For MSRs in which tritium control is the main concern, the $NaF-NaBF_4$ (8:92 with Tm=385°C) system is the prime candidate, mainly because of its satisfactory tritium trapping. A ternary salt $LiF-NaF-BeF_2$ should be considered in future studies as alternative secondary coolant because a freezing temperature range of about 315-335°C would be a practical value for engineering consideration. Because closed gas Brayton cycles can mitigate both the tritium and the melting point concerns, LiF-NaF-KF or $NaCl-KCl-MgCl_2$ may also be considered as a secondary salt.

Finally, heat transfer for lower temperature applications (below 600°C) requires a cheap and stable salt. $NaNO_3-KNO_3$ possibly with addition of $NaNO_2$ is the main candidate identified at this stage.

V.B Fuel salt clean-up scheme

The salt processing scheme relies on both on-line and batch processes to satisfy the constraints for a smooth reactor operation while minimizing losses to waste streams. ORNL experiments have provided some data mainly for the on-line gaseous fission product extraction process.

Acquisition of fundamental data for the separation processes is needed especially for the actinide-lanthanide separation. The extraction of lanthanides has to be done because of the low solubility of these trifluoride elements and neutronic captures that decrease the reactivity balance.

The progress made in core design in the last two years has opened the door for the definition of an improved fuel salt reprocessing scheme with a realistic fuel clean-up rate (40 l/day) and minimized losses to wastes. [6,8,15]

The proposed reference processing scheme is shown in Figure 3. The first step (green box) involves an on-line gaseous extraction with

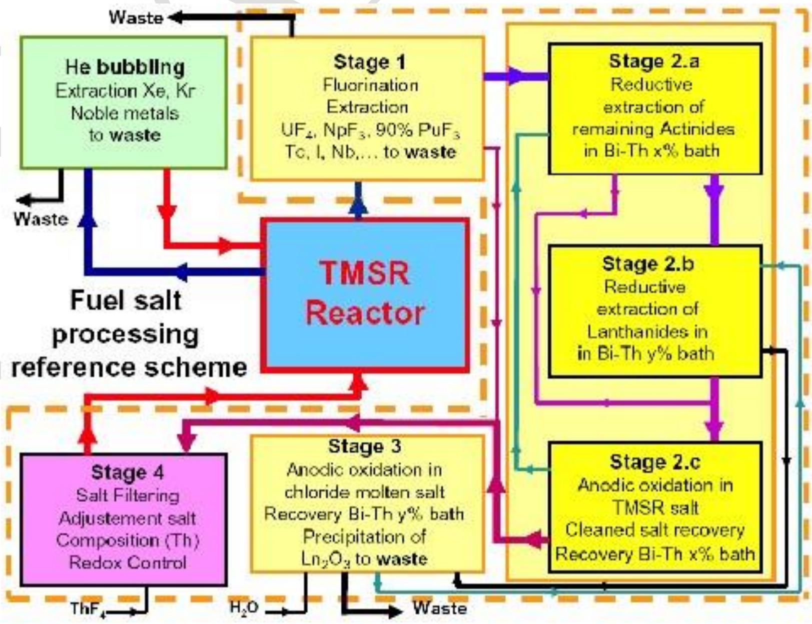

Figure 3: TMSR (MSFR) reference fuel salt processing

helium bubbling to remove gaseous fission products, Xe and Kr, and a part of the noble metals from fuel circuit. On the other hand, a batch fuel process separates the actinides which are returned to the reactor salt from the harmful fission products (mostly lanthanides). The fuel clean-up rate has been set at 40 liters per day, corresponding to the processing of 100 kg heavy nuclei per day. This value is almost two orders of magnitude less than the reference MSBR scheme.

The reference scheme depicted in Figure 3 involves 4 stages for the batch on-site fuel processing. The peculiarity of the concept appears in stages 2 and 3 by combining chemical and electrochemical methods for the extraction and the back extraction of actinides and lanthanides. This choice leads to fuel processing without effluent volume variation and the fuel processing balance is reduced to only one reaction: $2LnF_3 + 3H_2O(g) = Ln_2O_3 + 6HF(g)$.

Critical steps of the new fuel clean-up scheme are addressed and will be experimentally assessed in new facilities. The design and construction of a molten salt loop to study both He bubbling efficiency and material corrosion attack has been initiated. An efficient technique for actinide/lanthanide separation is under qualification. [16]

V.C Criticality tests for the assessment of MSR and AHTR fuel and core behaviour

The SPHINX (SPent Hot fuel Incinerator by Neutron fluX) project was originally defined as a suitable experimental basis at representative scale for the demonstration of MSR-burner feasibility. [17] It relies on the utilization of the zero power experimental reactor LR-0 being operated in the Nuclear Research Institute Řež (NRI), Czech Republic. This full-scale physical model of the PWR cores was modified in order to allow the measurement of all the neutronic characteristics of the MSR burner and/or breeder blanket, at first by room temperature and in future stage by conditions close to operational. (Figure 4).

Figure 4: LR-0 zero power critical test facility

Because two baseline concepts (MSFR, AHTR) are now considered in Generation IV, a corresponding broadening of the SPHINX project was discussed and formally adopted at the end of 2008. The LR-0 will thus be used for the validation of AHTR neutronics models (reactivity coefficients…) in the frame of a collaboration between the Czech Republic (NRI) and USA (University of California, Berkeley).

Two versions of EROS elementary blocks, as simplified models of the AHTR core module, have been designed and manufactured. During December 2008, the critical tests of both those elementary blocks were performed. The simplified models are completely ready for complex testing of experimental and measuring methods for detailed neutron field distribution and principle neutronic characteristics prediction.

VI. CONCLUSION

Europe (Euratom), France and USA participate in the Generation IV MSR Steering Committee. Although the European and USA interests are focused on different baseline concepts (MSFR and AHTR, respectively), large commonalities in basic R&D areas (liquid salt technology, materials) exist and the Generation IV framework is useful to optimize the R&D effort.

In USA, a PB-AHTR (900 MWt) is being developed most actively. A research, development and demonstration roadmap is under study for component testing to support a PB-AHTR prototype scale plant and a development path for the structural materials is being established.

In Europe, since 2005, R&D on MSR has been focused on fast spectrum concepts (MSFR) which have been recognized as long term alternatives to solid-fuelled fast neutron reactors with attractive features (very negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle…). MSFR designs are available for breeding and for minor actinide burning. They are robust reference configurations (with significant improvement compared to MSBR), allowing to concentrate on specific R&D issues [19].

A network on MSR R&D has been active in Europe from 2001 to 2008 with financial support by EURATOM. In parallel, ISTC has provided another efficient way of collaboration between Russian research organizations, European partners and non-European partners (USA, Canada, IAEA).

The GIF plays an important role to enhance and harmonize international collabo–ration on the R&D conducted in the different contexts.

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Nomenclature

AHTR	Advanced High Temperature Reactor
An	Actinide
GIF	Generation IV International Forum
LWR	Light Water Reactor
MSR	Molten Salt Reactor
MSFR	Molten Salt Fast Reactor
PB-AHTR	Pebble Bed Advanced High Temperature Reactor
PSSC	Provisional System Steering Committee
SRP	System Research Plan
UOX	Uranium Oxide

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