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Fast Spectrum Molten Salt Reactor Options

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ACRONYMS

AFCI	Advanced Fuel Cycle Initiative
ARC	Advanced Reactor Concepts
dpa	displacements per atom
DU	depleted uranium
EBR-II	Experimental Breeder Reactor
FS-MSR	fast-spectrum molten-salt reactor
HX	heat exchanger
IFR	Integral Fast Reactor
LMR	liquid-metal reactor
LWR	light-water reactor
MOSART	MOlten Salt Advanced Reactor Transmuter
MOX	mixed oxide
MSBR	Molten Salt Breeder Reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
MSR	Molten Salt Reactor
NRC	Nuclear Regulatory Commission
O&M	operations and maintenance
ORNL	Oak Ridge National Laboratory
SPHINX	SPent Hot fuel Incinerator by Neutron fluX
TRU	transuranics
NatU	natural uranium
RepU	reprocessed uranium
UNF	used nuclear fuel

EXECUTIVE SUMMARY

Fast-spectrum molten-salt reactors (FS-MSRs) have been the subject of periodic investigations since the early 1960s. However, none has ever been built; and the investigations have not proceeded beyond high-level material balance, heat transfer, and chemistry exploration. While the principal concepts underlying FS-MSRs have not changed over the intervening decades, much of the underlying technology base has evolved. The principal purpose of the current report is to provide an integrated overview of FS-MSR design options reflecting the current technology status.

FS-MSRs can be employed to consume actinides from light-water reactor (LWR) fuel or, alternatively, to extend fissile resource availability through uranium-to-plutonium breeding. FS-MSR reactors are highly flexible and can be configured into modified open or full-recycle configuration. The modified open FS-MSR fuel cycle options do not include chemical processing of the fuel salt. A traditional fully open fuel cycle is not an option with a liquid fuel reactor in that the gaseous fission products inherently separate from the liquid fuel. The conversion ratio of an FS-MSR is largely determined by the fissile-to-fertile-material ratio in its fuel salt. Thus, a single reactor core design may be capable of performing both fissile resource extension and waste disposition missions.

Molten-salt reactors can operate with the salt processing and fuel addition being performed in either continuous or batch operations. For thermal-spectrum systems, it is important to remove the fission products from the salt to minimize the parasitic neutron capture that results from fission products with large capture cross sections. In a fast-spectrum system, these parasitic losses are lower since the fission product capture cross sections are lower in fast-spectrum energy range. Hence, for an actinide burner reactor to maintain criticality, only sufficient additional fuel need be added to primarily compensate for burnup. Therefore, a fast-spectrum system may result in a simpler salt-processing approach or in a batch processing approach with relatively long salt-processing intervals. Similarly, a slightly positive breeding gain converter-type reactor may be able to operate for an extended period without any material additions or removal. In comparison, a breeder reactor will require fissile material extraction once the reactor control system cannot compensate for additional reactivity.

FS-MSRs have the potential for incorporating excellent passive safety characteristics. They have a negative salt void coefficient (expanded fuel is pushed out of the core) and a negative thermal reactivity feedback that avoids a set of major design constraints in solid-fuel fast reactors. Thus, an FS-MSR can provide a high power density while maintaining passive safety. The liquid state of the core also enables a passive, thermally triggered (melt plug) core draining into geometrically subcritical tanks that are passively thermally coupled to the environment. FS-MSRs have a low operating pressure even at high temperatures, and FS-MSR salts are chemically inert, thermodynamically lacking the energetic reactions with environmental materials seen in other reactor types (e.g., hot zirconium or sodium with water).

In the context of proliferation resistance, FS-MSR fuel has a uniform isotopic concentration of actinides, including highly burnt plutonium or uranium isotopes along with other minor actinides and fission products. The local fuel processing of the breeder and burner configurations eliminates the possibility of diversion during transport. The fission-product–saturated fuel salt of the minimal fuel processing converter reactor is highly self-guarding during transportation. Further, the transport casks are massive because of the required amounts of shielding. In general, diversion of molten salt materials is difficult. The reactor operates as a sealed system with an integrated salt processing system that is technically difficult to modify once contaminated. The hot salt freezes at relatively high temperatures (450–500°C), so it requires heated removal systems. FS-MSRs operate with very low excess reactivity. Loss of a significant amount of fuel salt would change the core reactivity, which could be measured by a well-instrumented reactivity monitoring system. During operation (with the exception of deliberate fissile material removal for a breeder or addition for waste burner), the fissile materials always remain in the hot, radioactive salt. However, FS-MSRs, with integrated fuel separation, may be unsuitable for deployment

in nonfuel-cycle states to minimize dispersal of separation technologies. Also, methods of inspection and materials accountability for liquid cores have not yet been fully developed.

All of the reactor-significant transuranic elements can form chemically and radiolytically stable salts with halide elements. Use of heavier halides results in a harder neutron spectrum. Harder neutron spectra improve the reactor burning and/or breeding. However, little information is available about the chemical and material properties of the actinide bromides or iodides. Hence, the FS-MSR investigation was limited to the actinide chlorides and fluorides. In addition to providing a harder neutron spectrum, chloride salts (compared with their fluoride counterparts) have higher solubility for the actinides, increasing the capability of the reactor to accommodate higher fuel loading and thus maintain criticality as fission products build up.

The two thermal-spectrum MSRs operated previously both employed a fluoride-based carrier salt. Much of the structural material information developed for the prior MSR program can be applied to fluoride salt FS-MSRs. The harder neutron spectrum of an FS-MSR, however, can cause additional atomic displacements within the nearby solid materials. Hence the lifetime of the neutron shielding material proximate to the core will be less for an FS-MSR. Additionally, nickel-based alloys embrittle when exposed to core levels of neutron flux ($>10^{20}$ neutrons/cm^2) at high temperatures ($>500°C$). Thus shielding of the primary vessel from the neutron flux is imperative.

Chloride-based salts have been employed in the fuel-reprocessing scheme developed for the integral fast reactor. However, the corrosion processes for chlorine are more complex than those for fluorine. Consequently the knowledge base for structural materials tolerant of chloride-based salts is not as mature as that for fluoride-based salts. A confident structural material selection cannot yet be performed for a chloride salt-based FS-MSR.

A light-water reactor (LWR)–transuranic burner can either make use of centralized fuel reprocessing or use much of the infrastructure of its fuel processing system to directly accept used LWR fuel, avoiding the need for a separate reprocessing plant. In addition to helium sparging to extract the gaseous fission products and mechanical filtering to remove the noble metal fission product particles, a fluoride salt–based FS-MSR would employ fluoride volatility and reductive extraction processes to separate the fission products from the fuel salt. Chloride salt–based reactors would employ electrochemical separation, zeolite ion-exchange capture, and chloride volatility processing. In either case, longer-lived fission products could be returned to the salt for fast neutron destruction, albeit with relatively low efficiency because of their primarily thermal absorption cross sections. As the separated fission products have relatively small volume, they can be left in salt form and allowed to solidify and decay in short-term storage.

A uranium–plutonium breeder would require an initial fissile material charge to initiate the breeding cycle. A liquid-fueled reactor is neutronically efficient compared with solid-fuel reactors, because it lacks in-core parasitic neutron absorbing structures. Therefore, a smaller amount of initial fissile material in the core is required to start up an FS-MSR compared with a sodium fast reactor. Note, however, that the FS-MSR will require additional fissile mass as a result of the fuel salt outside the core. The separations processes for a breeder would be nearly the same as for a burner except that the excess fissile material would be electrowon from the salt. Processing can be done in either batch or continuous form.

A minimal separation, modified open cycle, converter reactor would allow fission products to accumulate within the fuel salt. The lower melting points and much higher elemental solubilities afforded by chloride salts, as well as the resultant harder spectrum, make the chloride salts preferable for a limited-separation converter reactor. Sufficient natural (or depleted) uranium chloride or fluoride could be added to the reactor to compensate for any mismatch between fissile breeding and burnup, as well as the small amount of fission product absorption. Alternatively, the breeding ratio could be set to slightly positive to automatically compensate for the buildup of fission products. Helium sparging and mechanical filtering would be employed to separate gaseous and solid fission products. The process would continue until either the reactor vessel (or other major component) needed to be replaced, a fissile material solubility

limit was reached, or the overall salt melt temperature had been so shifted by fission product dissolution that it exceeded 550°C. At this point, the fuel salt would be pumped out, poisoned, and allowed to solidify into mechanically robust rock salt. The salt containers would then be sent for disposal or centralized reprocessing. Alternatively, the salt could be allowed to solidify in the reactor vessel and the vessel as a whole transported for reprocessing and or disposal.

1. INTRODUCTION

During 2010, fast-spectrum molten-salt reactors (FS-MSRs) were selected as a transformational reactor concept for light-water reactor (LWR)–derived heavy actinide disposition by the Department of Energy–Nuclear Energy Advanced Reactor Concepts (ARC) program and were the subject of a preliminary scoping investigation. Much of the reactor description information presented in this report derives from the preliminary studies performed for the ARC project. This report, however, has a somewhat broader scope—providing a conceptual overview of the characteristics and design options for FS-MSRs. It does not present in-depth evaluation of any FS-MSR particular characteristic, but instead provides an overview of all of the major reactor system technologies and characteristics, including the technology developments since the end of major molten salt reactor (MSR) development efforts in the 1970s.

This report first presents a historical overview of the FS-MSR technology and describes the innovative characteristics of an FS-MSR. Next, it provides an overview of possible reactor configurations. The following design features/options and performance considerations are described including:

- reactor salt options—both chloride and fluoride salts;
- the impact of changing the carrier salt and actinide concentration on conversion ratio;
- the conversion ratio;
- an overview of the fuel salt chemical processing;
- potential power cycles and hydrogen production options; and
- overview of the performance characteristics of FS-MSRs, including general comparative metrics with LWRs.

The conceptual-level evaluation includes resource sustainability, proliferation resistance, economics, and safety. The report concludes with a description of the work necessary to begin more detailed evaluation of FS-MSRs as a realistic reactor and fuel cycle option.

2. BACKGROUND AND PRIOR WORK

2.1 Technology Basis

MSRs were first proposed at Oak Ridge National Laboratory (ORNL) shortly after World War II as a means to power military aircraft as part of the Aircraft Nuclear Propulsion program and for civilian nuclear power. Two thermal-spectrum reactors were developed and operated at ORNL, the 2.5 MW(t) Aircraft Reactor Experiment in 1954 and the 8 MW(t) Molten Salt Reactor Experiment (MSRE), which operated from 1965 to1969 with over 13,000 full-power hours of operation. A significant amount of research and development was performed on MSRs over three decades, with progressive development of more advanced reactor concepts enabled through fundamental research on salt chemistry and high-temperature materials. The work was focused on development of a thermal-spectrum breeder based on the ^{232}Th/^{233}U fuel cycle for nuclear power sustainability.

An early version of an FS-MSR breeder system using NaCl/KCl/PuCl$_3$ salt was proposed by Alexander[1] in the early 1960s. In the 1960s–1970s, Taube,[2] from the Swiss Institute of Energy Research, explored a number of FS-MSRs, including a PuCl$_3$/NaCl salt. More recently, several fluoride fast-spectrum reactors have been proposed for use as actinide burners as part of the Gen IV program. These include the Czech

Republic's SPent Hot fuel Incinerator by Neutron fluX (SPHINX) reactor,[3] the European MOSART reactor[4] using $LiF/BeF_2/(TRU)F_3$ as a salt, and a more recent European MSFR[5] concept using $^7LiF/NaF/(TRU)F_3$.

The ARC program has recently sponsored a small, ongoing effort to evaluate the neutronic feasibility of transuranics (TRU) burning with an FS-MSR. Much of the information presented here derives from that evaluation.

FS-MSRs have highly flexible fuel cycle and energy production missions. Breeder, burner, and converter forms of FS-MSRs are all possible. Both chloride- and fluoride-based fuel salts are possible for all of the missions; there are significant remaining unknowns as to the advisability of specific implementations.

2.2 Innovative Characteristics

The unique characteristic of MSRs is the use of a liquid fuel rather than the solid fuels used in more conventional designs. Halide salts have been demonstrated to provide a high degree of solubility of actinides in concentrations sufficient to maintain a critical system. The use of liquid fuel enables many design options and fuel cycle opportunities that are not possible with solid fuel. Liquid-fueled reactors eliminate fuel or target fabrication, which presents technical challenges when using actinide and/or TRU fuel and can result in the need for capital-intensive facilities. In the MSR, each batch of fuel that is fed into the reactor is blended into the existing fuel inventory; consequently, the addition of fuel has a limited impact on the isotopic composition of the fuel so that no need exists to control the variability of the isotopic concentration of the feed fuel. The fuel feed can be in solid or liquid form.

An FS-MSR fuel cycle consists of a fast-spectrum molten salt core, a heat removal and power conversion system, and a salt processing and cleanup system. The reactor can be designed to have a range of heavy metal conversion ratios so that it serves waste management functions (with a low conversion ratio) or fuel cycle sustainability functions (with a high conversion ratio, unity or greater). For the waste management function, the system would be configured with a front-end processing system for used fuel, like the ones used in LWRs; whereas for the sustainability mission, after an initial charge of fissile material, the feed material could consist of natural or depleted uranium or thorium. For a waste management reactor, the front-end processing system could be located on site or the used LWR fuel could be processed at a central facility supporting several reactors. For the on-site reprocessing option, once the used LWR fuel is brought into salt form and the excess uranium is removed, the processing steps are identical to those for the FS-MSR's used fuel salt. Hence much of the infrastructure can do double duty, removing fission products from both used LWR fuel and FS-MSR fuel salt.

The safety aspects of FS-MSRs are also innovative. FS-MSRs have a negative salt void coefficient (expanded fuel is pushed out of the core) and negative thermal reactivity feedback that avoids a set of major design constraints in solid-fuel fast reactors. A passive core drain system activated by a melt plug enables draining the radioactive inventory into geometrically subcritical drain tanks that are passively thermally coupled to the environment. FS-MSRs have a low operating pressure even at high temperatures. The fuel/coolant is transparent, allowing visual inspection, and methods of maintenance for the system have been conceptually developed based on the MSRE experience. The high-temperature operation of the reactor is compatible with process heat applications and can be coupled to high-efficiency power conversion systems for electricity production.

Limited studies indicate that the reactor can start up on enriched uranium, actinides, or other mixtures and transition to thorium fuel cycles if desired (Commissariat à l'Énergie Atomique studies)[5] or be used as a minor actinide burner (Russian Research Center-"Kurchatov Institute" studies).[4]

2.3 Technical Maturity

Overall, FS-MSRs are quite immature. However, a somewhat higher level of confidence can be placed in FS-MSRs than in other preconceptual reactor concepts, as they build upon reactor technologies demonstrated in both in MSRE and the integral fast reactor (IFR) program. Liquid-fueled reactors represent an almost complete departure from the current Nuclear Regulatory Commission (NRC) licensing path; the only recent relevant experience is the NRC review of the Babcock and Wilcox aqueous homogeneous reactor for medical isotope production. A risk-informed licensing strategy would need to be developed to suggest licensing changes to be implemented to enable evaluation of FS-MSRs.

Neither the current fuel fabrication infrastructure nor the heavy section structural steel manufacturing infrastructure of LWRs is required for FS-MSRs. As a low-pressure, homogeneous reactor type, FS-MSR cores are structurally simpler than conventional heterogeneous LWR cores. The thinner-walled containers required for low-pressure operation are easier to manufacture than their thick-walled counterparts. Consequently, the manufacturing technology for the FS-MSR major structural components is relatively mature.

3. FS-MSR DESIGN OPTIONS

3.1 Reactor Configuration

Conceptually, all that is required for a FS-MSR is that fuel salt be brought into a critical geometry and heat be removed from the core. Thus MSRs have highly flexible configurations. FS-MSR configurations are even more flexible in that they do not require moderating media in the core and are more tolerant of fission product buildup, as previously discussed. The high degree of configurational flexibility has resulted in a diverse set of core design concepts over the past 60 years. Both single and dual fuel salt designs, that separate fissile and fertile materials, have been considered.

This report section will briefly sketch the major, single-fluid core and heat transfer component options that have been considered for FS-MSRs and then provide a somewhat more detailed description of the single-fluid loop type and integral reactor configurations. The fuel salt addition and salt cleanup portions of an FS-MSR fuel cycle introduce further variability in the plant configuration. Fuel addition options range from directly adding TRU fluoride or chlorides obtained from a centralized processing facility, to locally processing used LWR fuel into TRU fluorides or chlorides using as much as possible of the reactor's salt cleanup infrastructure.

Four single-fluid-type MSR configurations have been identified. Only loop and integral-type reactors are considered sufficiently practical that further analysis is recommended.

1. *Loop*: The fuel salt is pumped outside of the primary vessel and transfers heat to a secondary coolant in a separate heat exchanger.

2. *Integral*: The fuel salt remains within the reactor vessel. The secondary heat exchanger is located within the reactor vessel. Both the fuel and secondary salts are pumped.

3. *Direct Coupled*: The secondary coolant is directly mixed with the fuel salt within the reactor vessel. The secondary coolant naturally separates from the fuel salt and is pumped outside the reactor vessel to a heat transfer system.

   a. *Boiling Chloride*: Aluminum chloride (with a low boiling point of 180°C) is added to the fuel salt and heated through direct contact. The aluminum chloride vapors are then used as the secondary, heat transfer fluid.

b. *Liquid Lead*: Lead, which has low miscibility with chloride salts, is directly added to the top of the reactor vessel and pools at the bottom of the reactor vessel. Heat is transferred to the lead by direct contact with the fuel salt. The lead is pumped from the bottom pool of the reactor vessel through a heat transfer loop returning to the top of the vessel.

3. **Frozen Wall**: Fuel salt is solidified on the surface of heat exchanger tubing located within the reactor vessel. Secondary coolant is pumped through the heat exchanger tubing. A protective, solid salt layer forms at the surface of the tubing, enabling higher temperatures within the reactor vessel.

In an MSR the reactor core is in direct contact with its inner container layer. Therefore, the inner container layer serves as a fuel cladding layer that must withstand high levels of neutron flux. High-nickel alloys embrittle when subjected to high neutron fluence ($>\sim 10^{20}$ n/cm^2) at high temperatures (>500°C).[6] The embrittlement has both thermal and energetic neutron pathways. Thus the reactor vessel must be shielded from high neutron fluxes, or materials other than high-nickel alloys must be employed.

Some ferritic-martensitic steels have very good neutron flux tolerance.[7] However, the strength of ferritic-martensitic steels decreases greatly at higher temperatures, and even the most temperature-tolerant variants become largely unusable as structural material above 650°C. Note that high-strength, high-temperature ferritic alloy development is a current area of active research, and advanced alloys such as thermomechanically treated NF616 show promise for increasing the use temperature of ferritic-martensitic steels over the next decade.[8] Ferritic-martensitic steels, however, are not chemically compatible with fluoride salts at useful temperatures and are unlikely to be compatible with chloride salts at useful temperatures. One possible approach to enabling the use of ferritic-martensitic steels as MSR vessels is to employ a thick (mm) nickel-based cladding layer on the salt-wetted surfaces. In this way the chemical compatibility of nickel-based alloys can be combined with the strength and radiation tolerance of ferritic-martensitic steels.[9] One caveat to the use of advanced microstructure alloys in nuclear power plants is that, apart from solution-strengthened microstructures, all of the alloy-strengthening mechanisms (e.g., carbide, γ', nitride) dissipate over time at high temperatures. Thus the components will need to be replaced (perhaps more than once) during the lifetime of the power plant.

FS-MSRs can have very high power densities as they have no effective in-core heat transfer limits. Practical FS-MSR power density limits arise from a combination of radiation damage tolerance and heat flux limits on the surrounding shield material, and the heat transfer rate limits of the fuel salt. As the shield steel is massive and becomes highly radioactive during reactor operation, it is expensive to replace. Spreading the fast neutron flux out over a larger area increases the shield replacement interval. Also, the shield material is volumetrically heated by the fission gamma rays, providing an upper limit to the input gamma flux.

The practical limit to fuel salt heat transfer to the intermediate salt loop restricts the power density in the primary salt. Extrapolating from other liquid-to-liquid heat exchangers, roughly 150 MW/m^3 is a practical upper limit to the power density of a primary-to-intermediate salt heat exchanger. For comparison, pressurized-water reactor core power density is ~110 MW/m^3. For a 1000 MW(e) plant with an ~45% thermal efficiency, 2200 MW of thermal energy needs to be transferred. Employing the primary salt on the tube side of a shell-and-tube type of heat exchanger results in roughly 40% of the heat exchanger volume being fuel salt. This corresponds to roughly a 15 m^3 heat exchanger containing 6 m^3 of fuel salt.

Only the delayed neutrons emitted in the reactor core contribute to the controllability of the reactor. Most of the isotopes leading to delayed neutron emission by both ^{235}U and ^{239}Pu have long half-lives relative to the coolant loop cycle time, so the effective delayed neutron fraction, β, can be estimated by the ratio of the core to ex-core volumes. As ^{239}Pu has a small delayed neutron fraction, it is recommended to maintain the core volume at the ex-core volume, at least. Including 2 m^3 for the interconnecting piping, the core volume needs to be at least 8 m^3 with a 275 MW/m^3 power density. Note that these power density and heat transfer values are only best estimates and may be somewhat aggressive. Consequently, a real reactor

may have values only half as high, resulting in a significantly larger reactor core and set of primary-to-intermediate heat exchangers. For comparison, the proposed thermal spectrum molten salt breeder reactor (MSBR)[10] had a core average power density and core salt average power density of 22.2 MW/m^3 and 70 MW/m^3, respectively, because of the radiation damage lifetime limit (4 year replacement interval) on the in-core graphite. The material that determines the radiation damage lifetime limit for an FS-MSR will likely be the steel core shield-reflector piece that shields the reactor vessel from high neutron flux. Ferritic-martensitic steels can have very high radiation damage tolerance (>150 displacements per atom [dpa])[11] compared with graphite (a few dpa),[12] providing a substantial increase in the maintenance-replacement interval.

3.1.1 Pumped-loop

One possible configuration of a pumped-loop MSR is shown as Fig. 1. In this case the reactor vessel is shielded from the core by a thick-shield piece. A heavily nickel-clad ferritic-martensitic steel piece would be suitable to serve as the shield layer. The shield piece would be at the core boundary and thus would experience a comparatively lower fast neutron fluence than in-core structures.

The shield piece also serves to define the vessel downcomer region. The downcomer maintains the reactor vessel at the coldest temperature in the loop. The shield piece would be at least 20-cm thick to both neutronically isolate the core from the downcomer salt and limit incident neutron flux on the reactor vessel. The downcomer region is subcritical as a result of its geometry and the absorption characteristics of the intermediate salt.

Heavily nickel-clad ferritic-martensitic steel probably can also serve as the vessel material and the large piping material. As joining clad tubes is technically challenging, and the heat exchanger likely will have a large number of smaller tubes, a monolithic alloy exhibiting both strength and corrosion resistance is recommended. For a fluoride salt reactor, Alloy-N has been demonstrated to perform well at temperatures up to 700°C. No proven choice exists for chloride salt systems. However, alumina-forming alloys show promise for chloride salt compatibility.[13]
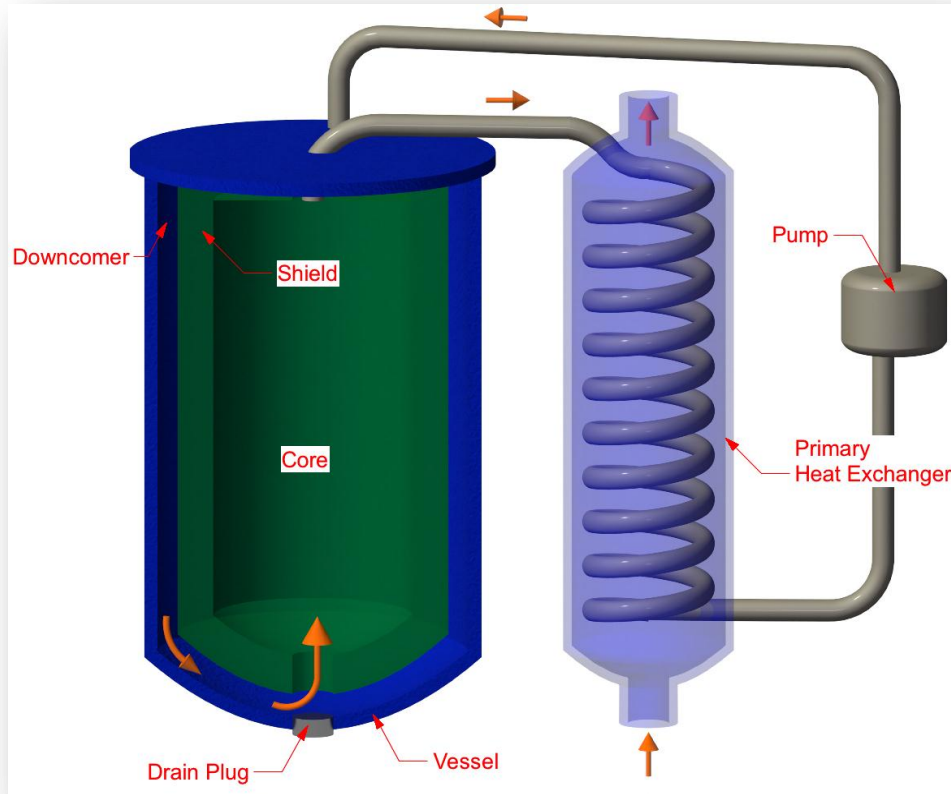
Fig. 1. Pumped-loop MSR major heat transport loop structures.

3.1.2 Integral

For an integral configuration, the fuel-to-secondary-salt heat exchanger is located within the reactor vessel. As complex, joined, thin-section structures, such as heat exchanger tubes and tube sheets, are typically more fast-neutron sensitive than bulk alloys, the heat exchangers are located in a subcritical section of the core. A conceptual diagram showing the major elements of an integral FS-MSR is shown as Fig. 2. In this concept, fuel salt pumping is performed using long-shafted pumps located well above the core.
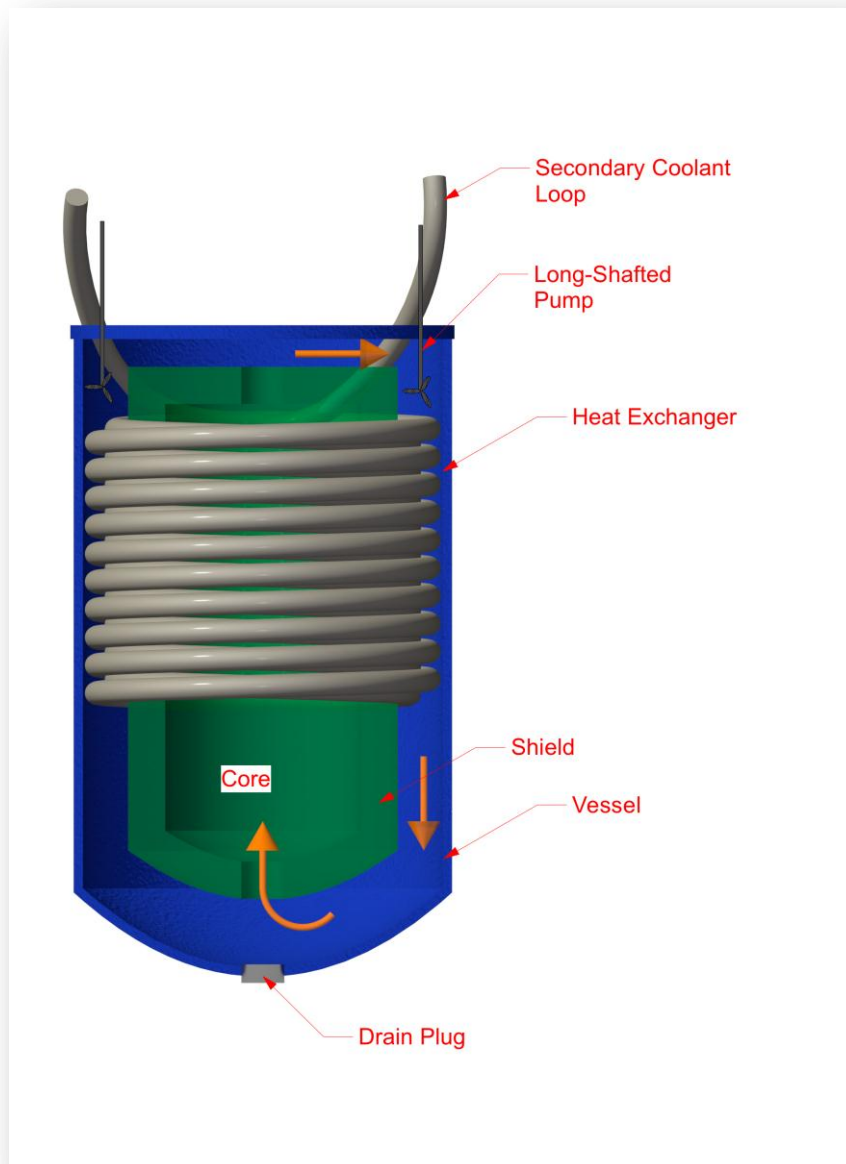
Fig. 2. Conceptual integral core configuration.

3.2 Salt Selection

In an FS-MSR, the fuel salt serves multiple functions and thus must meet multiple constraints. It serves as fuel, moderator, negative thermal reactivity feedback mechanism, heat transfer medium, and natural circulation drive mechanism for decay heat removal. The fuel salt functional requirements are similar for any MSR:

- reasonably low melting point (restricted to less than 550°C for this analysis);

- no isotopes/elements with high-parasitic absorption in a neutron spectral region with high neutron flux (primarily applicable to thermal-spectrum MSRs; a fast-spectrum reactor can tolerate materials with large thermal neutron absorption cross sections);

- large coefficient of thermal expansion to provide strong negative reactivity feedback and efficient natural circulation;

- sufficient dissolution of fissile material to create concentrations to support criticality;

- thermal and radiolytic stability;

- low vapor pressure at operating temperature;

- reasonable hydrodynamic and heat transfer properties; and

- compatibility with structural materials and other materials of core components.

All of the TRU elements contained in used LWR fuel can form chemically and radiolytically stable salts with halide atoms. The relative chemical inertness, acceptably small parasitic neutron absorption, and radiolytic stability of the TRU halide salts enables them to be employed as engineering working fluids near nuclear reactor cores. While any of the halides could serve as a salt anion, the analysis presented here is limited to fluoride and chloride salts primarily because of their larger knowledge base and the generally acceptable performance of the lighter halides.

Only a few mole percent of fissile material is generally required for maintaining criticality in an FS-MSR. Including larger amounts of fissile material necessitates including compensating amounts of neutron-absorbing fertile material or use of a high leakage geometry. A dilutant or carrier salt will, therefore, make up a large part of the fuel salt. The heat removal capacity of the heat transport system sets an effective upper limit to core power density.

All fluoride and chloride salts are good heat transfer materials with large specific heats and large coefficients of thermal expansion. The lighter alkali salts transfer heat somewhat more effectively than their heavier counterparts, resulting in lower pumping power requirements. However, the difference in pumping power needed to overcome the heat capacity differences is not large. The higher density of the heavy metal chloride salts impacts pumping power requirements significantly less than do the mass differences of the cation choice for a fluoride salt. The chloride salts, however, tend to have lower viscosity than their fluoride counterparts, somewhat offsetting the impact of their higher density on pumping power. Overall, other than melting point differences, the hydrodynamic and thermal properties of the fluoride and chloride salts are sufficiently similar that the other performance characteristics are dominant in the salt selection.

3.2.1 Fluorides

Fluorine is the most electronegative element and has a single oxidation number (–1). The actinide fluorides have several ionization states; however, little is known about the physical or chemical properties of the higher-ionization-state TRU fluorides. Even the melting points of Am, Np, and Cm tetrafluoride remain unknown, although they are anticipated to be lower than those of the trifluorides.

Plutonium trifluoride has low solubility in fluoride solvents.[14] Plutonium tetrafluoride exhibits a much higher solubility than PuF_3 in fluoride solvents but would be a stronger oxidizing agent necessitating alternative container materials.[15] The other heavy actinides are at such low concentrations as to be well soluble in any fluoride melt. The corrosion reaction between PuF_4 and a container atom would reduce the plutonium into the less soluble PuF_3. Overall, the tetrafluoride TRU salts would provide a higher heavy metal fraction within the core than the equivalent trifluoride salt. However, using TRU tetrafluoride salts would require developing advanced structural materials such as refractory alloys and/or carbon-based

composites. For practical applications, the larger corrosivity of the TRU tetrafluorides is sufficiently deleterious that only the TRU trifluorides are considered further.

The solvent properties of the dilutant salt are a critical parameter in salt selection. All of the TRU trifluorides have melting points near 1400°C. While the phase diagrams of multicomponent fluoride salt mixtures with TRU trifluorides are not known, the available phase information indicates that TRU trifluoride mixtures with alkali fluorides all melt above 700°C and at useful dilution ratios generally above 1000°C. As these temperatures are well above those tolerable for available structural materials, the obtainable TRU fluoride salt composition becomes largely determined by dissolution chemistry.

As a general solubility model for the TRU trifluorides does not exist, conservative assumptions on solubility are required. A recent TRU burner reactor model employing PuF_3-NaF-ZrF_4 fuel salt was not able to achieve criticality with the 1.56 mol % plutonium that was assumed to be soluble in this salt at 650°C (Ref. 16). Lithium and beryllium fluorides are known to exhibit relatively high solubility for the TRU trifluorides, along with low parasitic neutron absorption. Consequently, thermal-spectrum MSR designs have relied upon these salts. The MSRE dilutant salt was 2^7LiF-BeF_2. The MSRE operated with between 4.5 and 4.75 wt % (~0.5 mol %) uranium in the fuel salt.[17] The dilutant salt employed in the MOSART reactor concept, NaF-^7LiF-BeF_2 (58-15-27 mol %), dissolves sufficient TRU fluoride to perform as a useful reactor core, albeit with a neutron spectrum too thermalized to maximize TRU consumption.

Fluoride salt reactors can be designed to have a harder neutron spectrum. Neutron kinetic energy loss in the MOSART salt is dominantly caused by elastic scattering with the lithium and beryllium nuclei. Alternate, higher-mass fluoride solvents, such as lead difluoride, will decrease the neutron lethargy gain per scatter, resulting in a harder neutron spectrum. The heavy metal fluoride glasses (e.g., NaF-PbF_2-ZrF_4-LaF_3) are promising carrier salts with relatively low melting points and capability for high lanthanide and actinide solubility. For example, (assuming LaF_3 has similar solubility characteristics as the TRU fluorides) ZPLN glass [$55ZrF_4$-$20PbF_2$-$5LaF_3$-$20NaF$ (mol %)] includes 5 mol % LaF_3 and has a melting point of 512°C (Ref. 18). While $(TRU)F_3$ would be able to substitute for some of the LaF_3 in the melt because of its similar chemical properties, insufficient information is currently available to be able to confidently state that a sufficient substitution is possible to achieve critically. Additional phase simulation and measurement work would be required to confidently recommend proceeding with reactor design based upon a ZPLN melt.

For the majority of uncooled metallic components in a fluoride salt system at 650°C operating temperature (reactor pressure vessel , pumps, shields, heat exchangers), there are several materials that would provide the needed performance, including Alloy N. Alloy N is a nickel-base alloy (Ni-7Cr-16Mo-1Si) developed at ORNL explicitly for containing high-temperature molten fluoride salts. It is currently codified for use in ASME Code Section VIII under Code Case 1315 for use in pressure vessels at up to 704°C, but it is not explicitly approved for nuclear construction within ASME's Section III. Notwithstanding, Alloy N has adequate strength and excellent salt and air compatibility up to 704°C and has been successfully used in construction of both of the MSRs developed at ORNL.

3.2.2 Chlorides

Chlorine has two stable isotopes ([35]Cl –75.77 atom % and [37]Cl – 24.23 atom %) and several oxidation numbers (+7, +5, +3, +1, and –1). As a result of the more complex bonding configurations available in chlorides, their solution and corrosion chemistry is more complex than that of fluorides.

The actinide trichlorides form much lower melt point solutions with chloride salts and contain significant amounts of TRU. An example salt that has received significant prior analysis is $PuCl_3$-$NaCl$ (Ref. 19), which can contain ~40 mol % $PuCl_3$ while exhibiting a melt point below 500°C. Overall, NaCl exhibits good nuclear, chemical, and physical properties. As the melt point of NaCl containing only 1–2 mol %

PuCl$_3$ is nearly 800°C, the selected salt will need to be a mixture of NaCl and another dilutant salt selected to lower the mixture melting temperature.

Both MgCl$_2$ and CaCl$_2$ are possible additional components to a NaCl fuel salt. However, they are somewhat less chemically stable and more expensive than NaCl. Both have relatively low neutron absorption. Magnesium activation yields the shortest half-life activation product [T$_½$ ^{27}Mg ≈ 9.46 minutes; T$_½$ ^{41}Ca = 103,000 years]. An NaCl-CaCl$_2$ mixture would have a somewhat higher melt point [507°C for a 48NaCl-52CaCl$_2$ (mol %)] than a eutectic mixture of NaCl and MgCl$_2$ [445°C for a 41.5NaCl-58.5MgCl$_2$ (mol %)].

Overall, MgCl$_2$ would be the preferred addition to a (TRU)Cl$_3$–NaCl melt with the proportions of the components near the low-melt eutectic point for NaCl-MgCl$_2$. Adding a few percent of (TRU)Cl$_3$ to the melt would raise the 445°C melting point somewhat. Chloride melts exhibit low viscosity a few tens of degrees above their melt points; therefore, the melt is anticipated to have suitable viscosity by 500°C.

While the neutron spectrum of a chloride salt reactor can be hardened somewhat by adding heavier cation chlorides, the more complex chemistry of chlorine increases the desirability of employing only alkali and alkaline earth elements with their single ionization states in the melt. If, for example, PbCl$_2$ is included in the melt, radiolysis will inevitably produce some PbCl$_4$. Lead tetrachloride has a boiling point of 50°C (in contrast to the 953°C boiling point of PbCl$_2$) and would thus separate from the melt resulting in a high vapor pressure.

A 42.5KCl-30.5SrCl$_2$-27RbCl (mol %) melt would be technically possible with a melting point of 514°C (Ref. 20). However, rubidium is not regularly used in industry; thus a source of supply would need to be developed. Further analysis was limited to commonly available elements.

Chlorine-35 has an additional issue for in-core deployment. Its activation product, ^{36}Cl, is a long-lived (301,000 years) energetic (709 keV) beta emitter that is highly soluble in water. As removing radiotoxicity from the biosphere is a primary purpose for a TRU-burning reactor, production of large amounts of ^{36}Cl could be problematic in some disposal scenarios. In particular, ^{36}Cl may make a significant dose contribution in a repository located in a clay environment.[21] However, ^{36}Cl could be effectively disposed of in a salt geologic repository. Chlorine-36 production can be avoided by isotopically separating the chlorine to minimize the ^{35}Cl. Liquid-phase thermal-diffusion-based separation of the chlorine isotopes appears to be a feasible technology (but with as yet unanalyzed economics).[22] Thus the use of isotopically separated chlorine could be a preferred option for chloride-based fuel salts. Potassium chloride should be avoided in an isotopically separated chloride reactor since it can produce ^{36}Cl through the ^{39}K(n,α)^{36}Cl reaction, albeit with relatively small yield.

None of the likely chloride-based salts is aggressively corrosive to stainless steels or nickel-base alloys at likely reactor temperatures (<600°C) without oxygen being present.[23,24] However, long-term operation requires a high degree of material compatibility, and this has not been established for the particular chloride salt selected (NaCl-MgCl-(TRU)Cl$_3$). Also, in-core testing of material compatibility of structural materials exposed to fuel-bearing chloride salts has yet to be performed, so materials compatibility cannot yet inform the salt selection. Redox control within a chloride salt melt would be more complex (and as yet unproven) than in a fluoride salt because of the more complex chemistry arising from the larger number of ionization states, but it may be a useful technique for limiting structural alloy corrosion.

3.3 Conversion Ratio

The liquid fuel of FS-MSRs enables a wide range of design options. FS-MSRs can have conversion ratios ranging from burner to converter to breeder. They can support a waste consumption, fissile resource extension or may serve as modified open-cycle power plants without heavy metal separation from fission products. The fast neutron spectrum enables the no-heavy-metal separation configuration. Fission products have relatively large neutron capture cross sections in the thermal energy range but smaller

capture cross sections at higher energies. Thus much greater fission product buildup is tolerable in an FS-MSR than in a thermal-spectrum MSR. Figure 3 shows the infinite media calculated neutron energy spectrum for a TRU burner FS-MSR with several different carrier salts. As the figure shows, for the hardest-spectrum salts, the neutron flux has fallen by a factor of more than one million for neutron energies of less than 100 eV. The capability to function for an extended period without fuel processing, beyond trapping fission gasses and mechanically filtering noble metal particles, affords the possibility not to have an on-site fissile material separations capability. Not having integral fuel processing removes the principal proliferation impediment to exporting MSRs. Longer-term, the heavily fission-product-loaded fuel salt would most likely be poisoned, pumped into transport containers, allowed to solidify, and then either disposed of or sent to centralized facilities for processing.
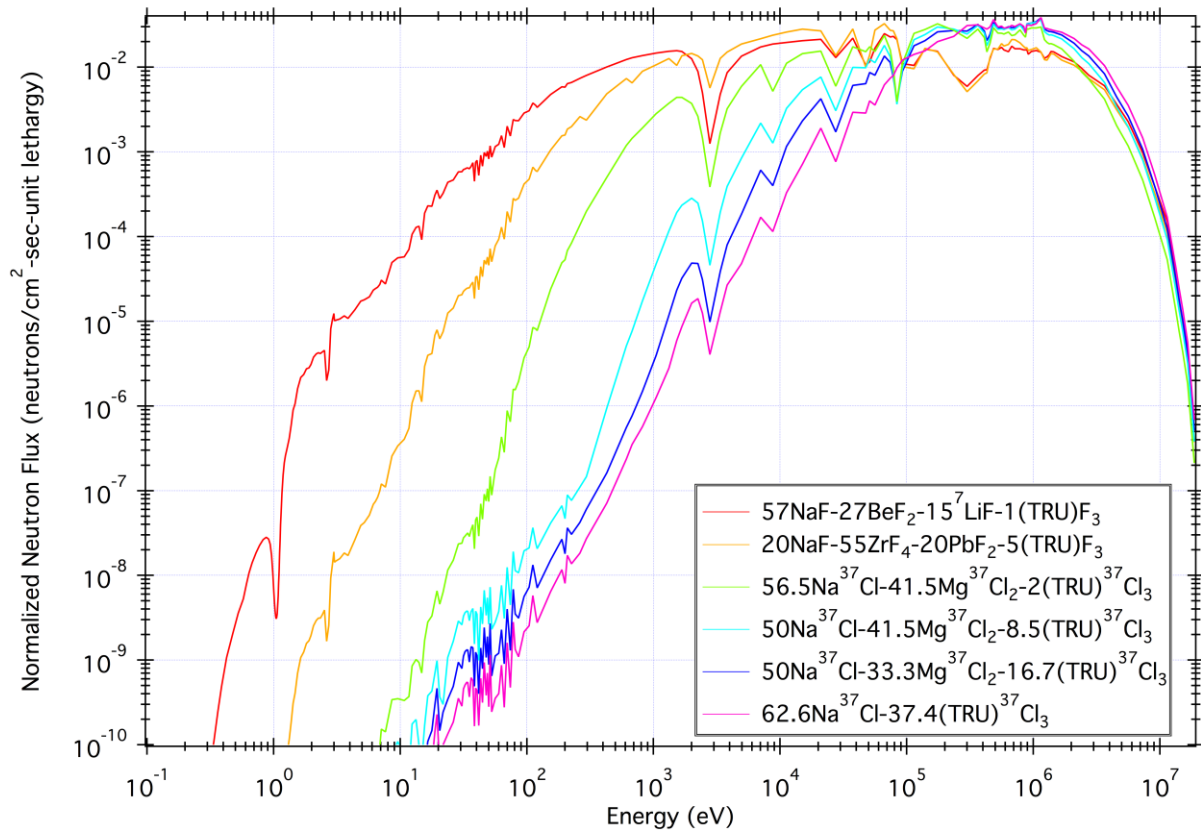
Fig. 3. FS-MSR TRU burner spectra.

An FS-MSR's conversion ratio depends on both the isotopic composition of the fuel salt and the heavy metal loading. The fuel salt heavy metal loading impacts the conversion ratio by changing the neutron spectrum. Increasing the fraction of heavy atomic mass atoms within the core decreases the mean neutron lethargy gain per scattering event, thereby hardening the neutron spectrum and increasing the mean energy of the neutrons causing fission. Since the mean neutron yield per fission ($\bar{\nu}$) increases with neutron energy for all fissile isotopes, more neutrons are available in succeeding neutron generations to convert fertile isotopes to fissile isotopes. Thus the reactor conversion ratio increases.

Figure 4 shows the variation in the average number of neutrons produced per fission for different fuel salts for a TRU burner reactor and the variation in the average energy of neutrons inducing fissions for the same salts.
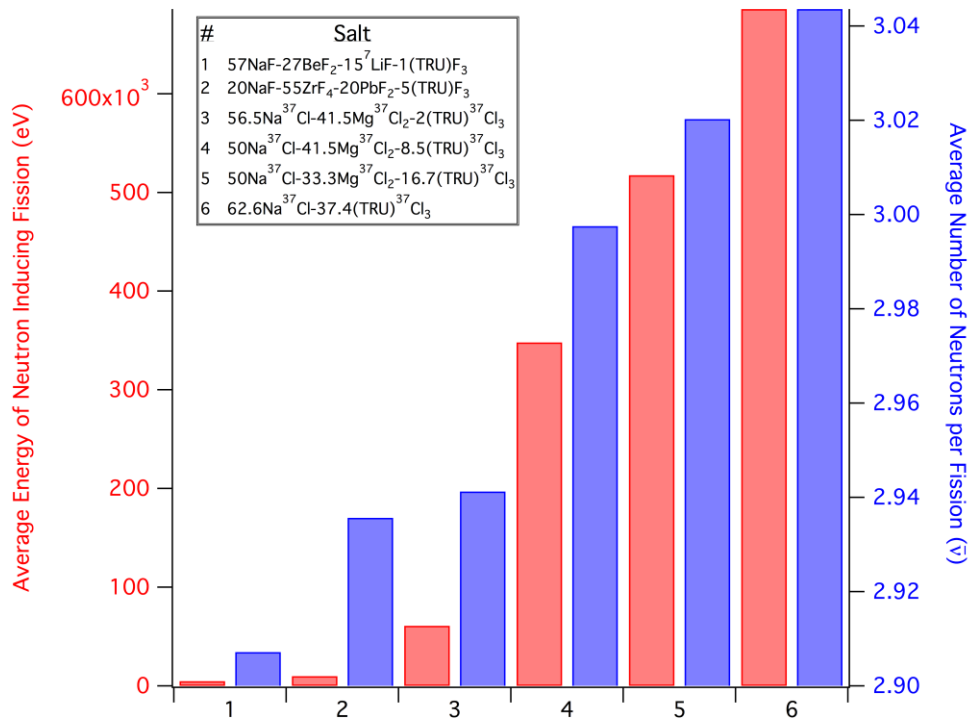
Fig. 4. Average energy of neutrons inducing fission (left axis) and average number of neutrons produced per fission (right axis).

The ability of a single FS-MSR reactor configuration to shift from breeding to burning based upon its heavy metal composition is shown in Fig. 5. As progressively more fertile ^{238}U is loaded into the salt, displacing TRU isotopes, progressively larger numbers of neutrons are captured in the ^{238}U, breeding additional fissile material. Thus the TRU to ^{238}U ratio can be used to respond to changes in the reactor mission while maintaining a critical configuration.

Another technique for expressing the LWR actinide burning capabilities of an FS-MSR is the "D-factor," or neutron consumption per ultimate fission destruction of a nuclide or its capture products.[25] A lower D-factor indicates that fewer neutrons are needed to ultimately fission an existing nuclide, and a negative D-factor indicates that there is a net production of neutrons in the fission destruction of the nuclide. A comparison of the D-factors for several different salts is shown in Fig. 6, along with a metal-fueled sodium fast reactor[26] and an LWR.[31] The TRU composition used for Fig. 6 is that for a PWR with 55 GWd/MT followed by 10 years of cooling.[27] The D-factor is only one means of expressing the

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Fig. 5. Variation in fission to absorption probability with shift in fuel heavy metal composition.
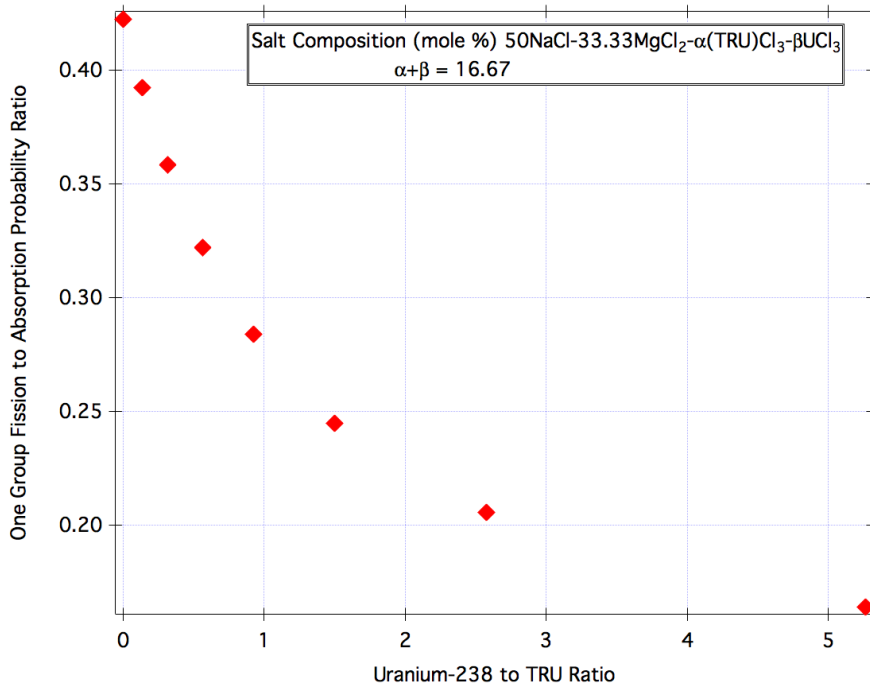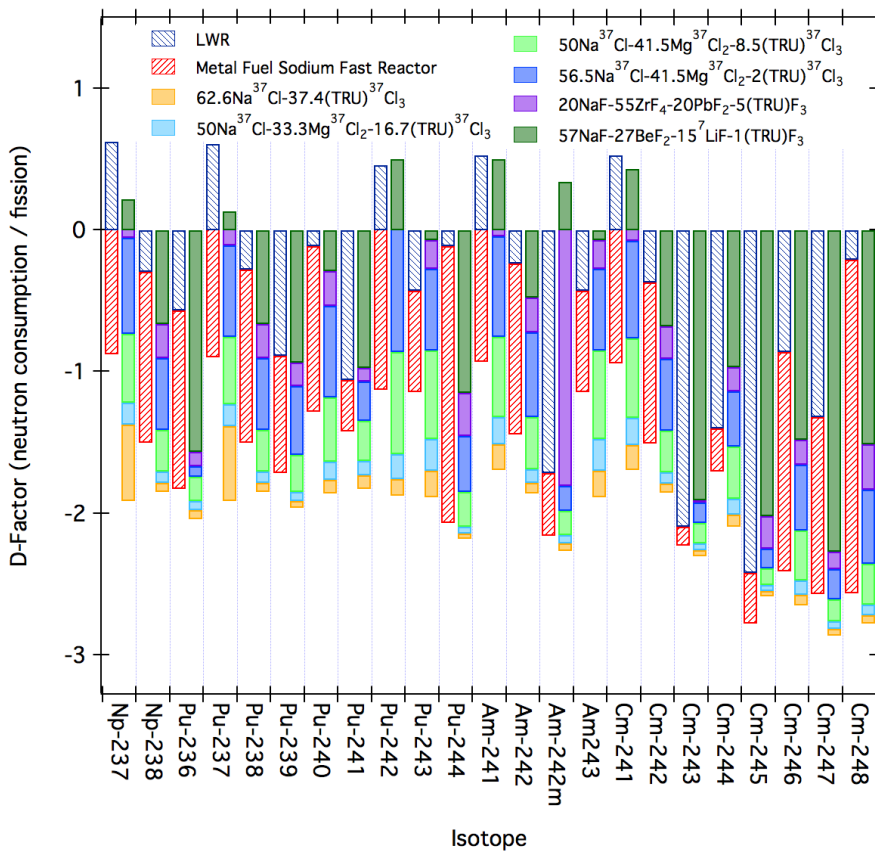

Fig. 6. Neutron consumption per fission (D-factor) for LWR-derived TRU.

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effectiveness of a given reactor system for actinide burning. D-factor analysis does not weight the value of the destruction of any particular isotope either by its abundance or by the eventual repository heat loading. The nuclides heavier than curium, which a more energetic neutron spectrum is especially effective at consuming in fission, have very low abundance in LWR-derived TRU. It is important to note that this D-factor approach represents only the neutron balance between capture and destruction and does not consider other aspects of the system. Additionally, the increase in the neutron yield per fission with the increasing energy of neutrons inducing fission cannot be fully exploited in most core designs because of the requirement to leak or parasitically absorb the neutrons in excess of those required to maintain criticality.

In an FS-MSR, the change in the reactor critical configuration is a balance between the increased neutron yield per fission at higher neutron energies, the increased fission product absorption at lower neutron energies, the harder neutron spectrum produced by increasing the heavy metal loading in the salt, the time-varying isotopic composition within the salt, the fuel salt velocity and consequent in-core delayed neutron fraction, and the reactor core geometric leakage. The large negative thermal reactivity feedback provides inherent stabilization to the reactor. A critical configuration of a highly TRU-loaded salt is inherently small. The reactor critical volume needs to be sufficiently large to avoid rapidly degrading its first wall shield material as well as to enable practical heat transfer to a power cycle. The critical volume for a 2 mol % TRU-loaded chloride salt is roughly 9 m^3, and that for an 8–10 mol % TRU loaded chloride salt is roughly 1 m^3. Substituting ^{238}U for some of the TRU in the fuel salt enables maintaining a harder neutron spectrum in a larger volume core. Alternatively, employing a high neutron leakage core geometry will reduce the neutron multiplication. A significant amount of design work remains to be performed to develop a workable FS-MSR system.

3.4   Salt Processing Technology

The FS-MSR considered in this study consists of *a front-end processing system for used LWR fuel*, an FS-MSR, and *salt processing system for cleaning the fuel salt* that is cycled through the reactor. The front-end processing system could be either at a centralized fuel recycling facility to produce TRU oxide or co-located with the reactor. If it were co-located with the reactor, it is possible that some of the systems, equipment, and techniques used for fuel salt production could also be used for salt cleaning.

The chloride- and fluoride-based salts processing and cleaning schemes described below build upon previous design and demonstration efforts. Chloride salts have been previously considered for use as part of the IFR fuel process concept, and the electrorefining of spent nuclear fuel in a molten chloride cadmium system was demonstrated using Experimental Breeder Reactor (EBR-II) spent fuel on a pilot scale. Fluoride salts and associated processes were used in the MSRE at ORNL and have recently been considered for use in the MOSART concept.

3.4.1   Front–end processing system options for used LWR fuel

The fuel for a FS-MSR actinide burner is a mixture of fluorides or chlorides of plutonium and minor actinides (Np, Am, Cm) from used LWR fuel. Although many of the fuel fabrication challenges that must be addressed with the fabrication of actinide fuels are avoided in the MSR design and operations, front-end processes are necessary to convert the used LWR fuel into a form suitable for dissolution into a molten salt. Front-end processing will include steps for removing used fuel rods from fuel rod assemblies and chopping or shearing the rods into smaller pieces for further handling. Front-end processing may also include steps such as voloxidation or dry chlorination to remove some fission products for the used fuel prior to separation. Depending on the separations processes selected for treatment of the used fuel prior to dissolution of the actinides into the fluoride or chloride carrier salt, mechanical or chemical decladding of the used fuel may be necessary as well. Initial separations of the actinides may be carried out by traditional hydrometallurgical processes or alternatively by pyroprocesses.

3.4.2 Pyrochemical (electrorefining)

Electrorefining processes have been investigated for use in the IFR at Argonne National Laboratory and could be adapted for use in an FS-MSR actinide burner. Note that this type of process could be used both as part of the front-end LWR fuel processing and as part of the cleanup of fuel salts circulated through the reactor. Electrochemical separations can take place in molten salt electrolytes; for use in the MSR, it makes sense to use the fuel carrier salt as the electrolyte if possible. These techniques take advantage of electrically driven chemical processes instead of chemical equilibrium to achieve separations. The processes include electrolytic reduction and electrorefining as the primary methods to treat used nuclear fuel.

Used oxide fuels must be reduced to metallic form before the electrorefining step; an electrolytic reduction step could be used to remove the oxygen from the melt as a gas so that it need only be treated to remove entrained particles or volatile contaminants.

A specific metal may be recovered by adjusting the electric potential across the separation cell to values opposite in direction to the electromotive force of that metal in the molten salt electrolyte. Successive adjustments in potential permit the various metals to be recovered sequentially, or multiple cathodes could be used to capture the metals of interest.

An example of the use of electrorefining in the FS-MSR is shown in Fig. 7. For an FS-MSR, the chopped spent fuel (or fuel powder if a voloxidation processes is used) is immersed in the NaCl-MgCl$_2$ carrier
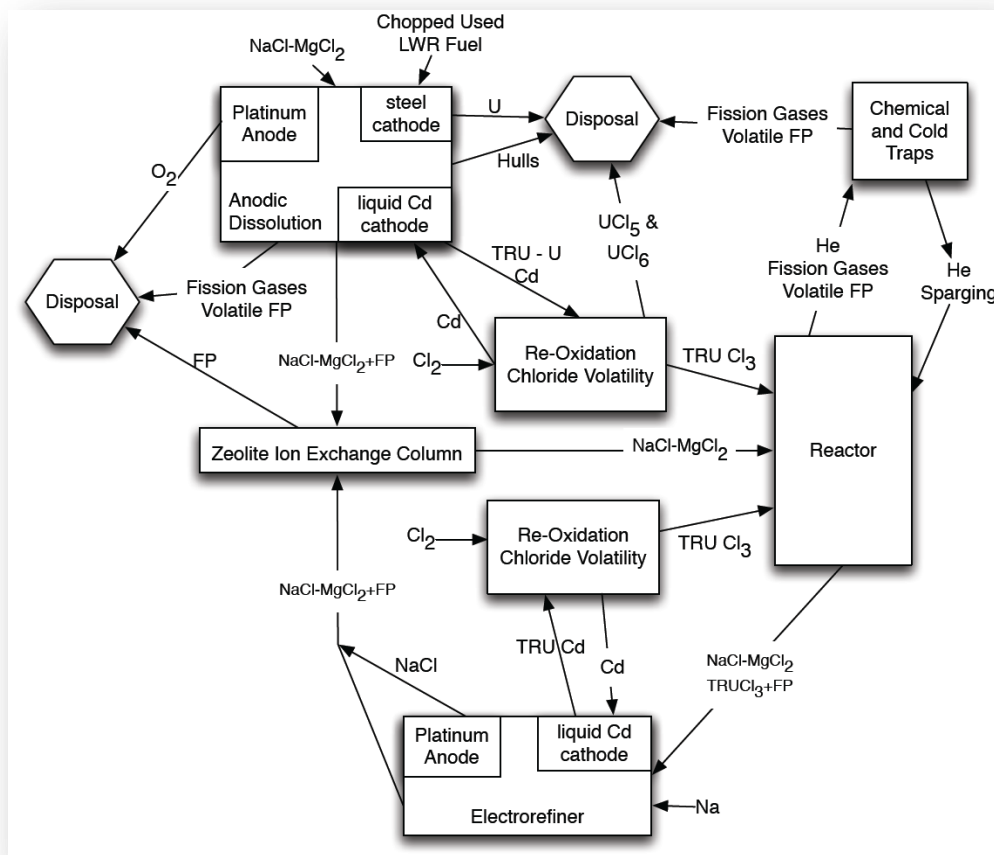

Fig. 7. FS-MSR fuel salt preparation and salt cleanup using an electrorefiner.

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salt used as the electrolyte, where the used fuel is anodically dissolved away from the cladding if cladding is still present. A large fraction of the uranium would be recovered on a steel cathode and further processed for disposal. The transuranics and the remaining uranium would be deposited in liquid cadmium cathodes. The transuranics and remaining uranium could then be sent for reoxidation by a chloride volatility process, since these elements form volatile chlorides. UCl_5 and UCl_6 would be sent for disposal, while the transuranic chlorides could be mixed back in with the $NaCl$-$MgCl_2$ carrier salt and cycled through the reactor. The anode collects material from the cladding hulls, undissolved actinides, noble metal fission products, and entrained salt. Oxygen and water could be removed by a cell gas purification system. Fission product gases (tritium, krypton, and xenon) can be captured and treated in a manner similar to the gas capture system covered in voloxidation process. The salt contaminated with fission product chlorides must be cleaned. (Note that this contamination can be either from the front-end processing of used LWR fuel or from MSR operation.) Cleaning is accomplished by sending the $NaCl$-$MgCl_2$+FP to a zeolite ion exchange system, which could be in the form of a column or bed.[28] The sorbed fission products are then removed for disposal, and the clean $NaCl$-$MgCl_2$ carrier salt is mixed with the recovered actinides and fed to the MSR.

3.4.3 Pyrochemical (fluoride volatility)

Another pyrochemical processing concept that could be developed and might be well suited for the FS-MSR based on a fluoride salt is the fluoride volatility method. This method was investigated as a fuel reprocessing technology for fast breeder reactors and has been proposed for use in the MOSART concept.[4] The use of the fluoride volatility method is illustrated in Fig. 8. In this concept, the chopped used LWR fuel is sent to either a flame fluorinator or a fluidized bed reactor for volatilization. As part of this process, fission gases from the volatization process are captured for disposal. The fluoride volatility method converts LWR used fuel into fluorides and separates the main parts of uranium and plutonium and neptunium. Since the separation of trivalent actinides (Am, Cm) from the majority of fission products (mainly trivalent lanthanides) is not possible using only this method, the transuranics, uranium, and remaining fission products are sent to another pyro-partitioning step for the final separation of transplutonium actinides. Technologies that may be suitable for this pyro-partitioning step include the electrochemical separation similar to the process described above for the chloride-based salt, or a molten-salt/liquid metal extraction process. The separated actinides can then be combined with the fluoride salt and sent to the reactor for burning. Pyrochemical reprocessing for fuel salt cleanup after it passes through the reactor is described further below.

3.4.4 Fuel salt cleanup

Continuous operation of an FS-MSR actinide burner requires fuel salt processing and cleanup. The fuel salt in the MSR primary circuit must be processed to remove fission product gases as they are generated, remove neutron poisons to minimize neutron losses, and possibly remove noble metals that may plate out on reactor system surfaces and interfere with plant operations.[17] Some fission products in a MSR are removed almost automatically, and some can be removed by applying processes to side streams.[29] In addition, it will be necessary to clean the molten salt to remove impurities such as oxygen and to maintain the appropriate fluorine or chlorine (redox) potential in the salt to address material corrosion. Hydrofluorination is commonly used to remove moisture. The salt is kept reduced to control corrosion of structural materials by keeping the ratio of UF_4/UF_3 (($TRU)F_4/(TRU)F_3$) to approximately 0.05 as a buffering agent. Similarly, for a chloride-based salt, the ratio of $PuCl_4/PuCl_3$ (($TRU)Cl_4/(TRU)Cl_3$ will need to be monitored and managed to help control corrosion.

Table 1 summarizes the processes that could possibly be used for fuel salt cleanup in the MSR.[4,29,30]
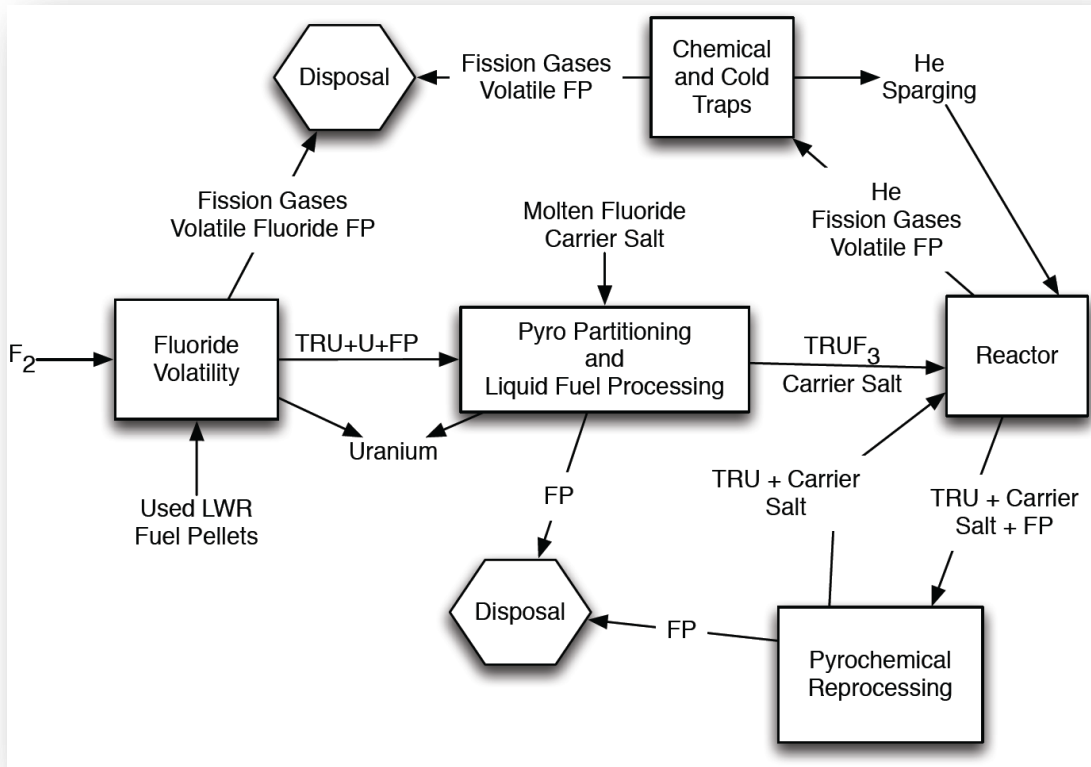
Fig. 8. FS-MSR fuel salt preparation using fluoride volatility and fluoride salt cleanup.

Table 1. Summary methods for fission product removal and TRU recycling

Component	Removal/processing operation
Kr, Xe	Sparging with helium
Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, Te	Plating out on surfaces, filtering, electrowinning, partial removal to off-gas system
Zr	Reductive extraction followed by hydrofluorination or metal transfer
Ni, Fe, Cr	Oxide precipitation
Np, Pu, Am, Cm	Electrodeposition
Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er, Sm, Eu	Distillation
Sr, Ba, Rb, Cs	
Li, Be, Na	Salt discard

Removal of the fission products other than the rare earth elements, including those discussed earlier, may improve the neutron economy of the reactor but may not be necessary from either a chemical or a neutronics perspective. The need for such removal should be reevaluated as the development of the reactor neutronics performance aspects continues.

3.5   Fuel Cycle Options

FS-MSRs have three primary design variants: (1) an LWR-derived TRU burner, (2) a uranium-plutonium breeder, and (3) a natural uranium–fueled minimal-separation converter. Thus FS-MSRs can support waste consumption or fissile resource extension or can serve as modified open-cycle power plants without on-site heavy metal separation. The fuel cycle for the TRU burner FS-MSR is shown in Fig. 9. The equilibrium (following startup fissile material charging) breeder fuel cycle is shown in Fig. 10. The equilibrium minimal processing fuel cycle is shown in Fig. 11. The minimal fuel-processing diagram does not include the eventual disposition of the entire fuel salt core. The fast neutron spectrum enables the configuration involving no heavy metal separation. Fission products absorb neutrons dominantly at low energies. An FS-MSR has few neutrons at these energies. Thus much greater fission product buildup is tolerable in an FS-MSR than in a thermal-spectrum MSR. The uranium addition to the converter cycle reactor may be very low for several years as the roughly unity conversion ratio compensates for the fissile material burnup.
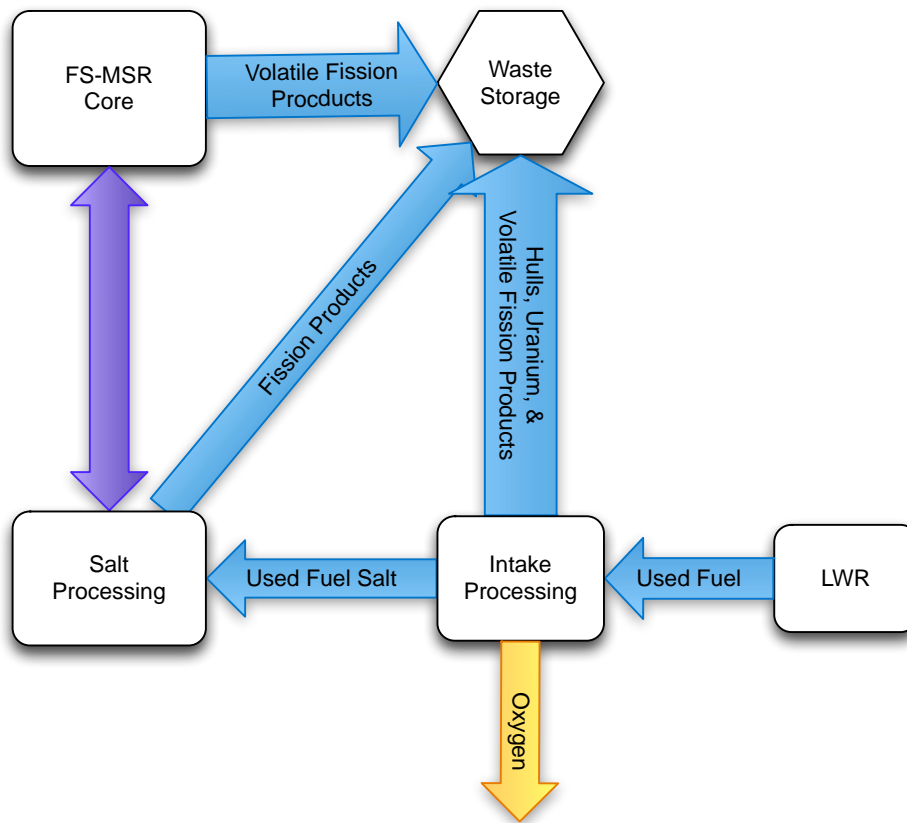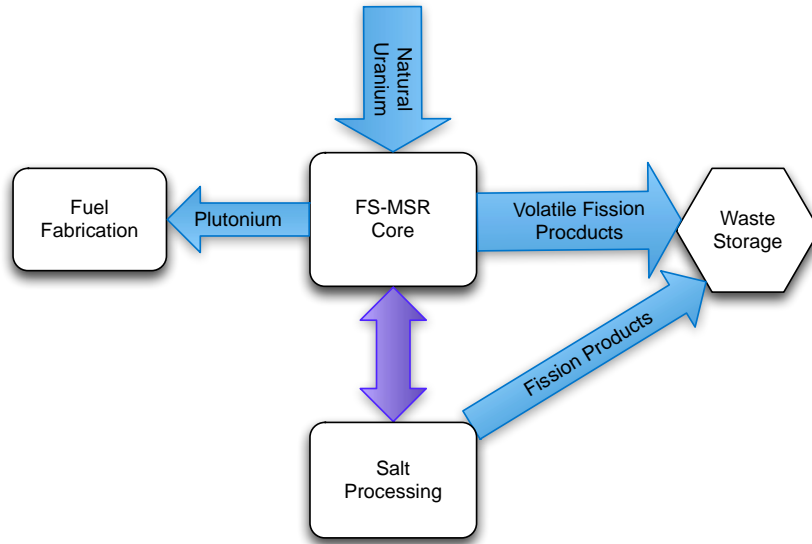
Fig. 9. TRU burner fuel cycle.

Fig. 10. Uranium–plutonium breeder equilibrium fuel cycle.
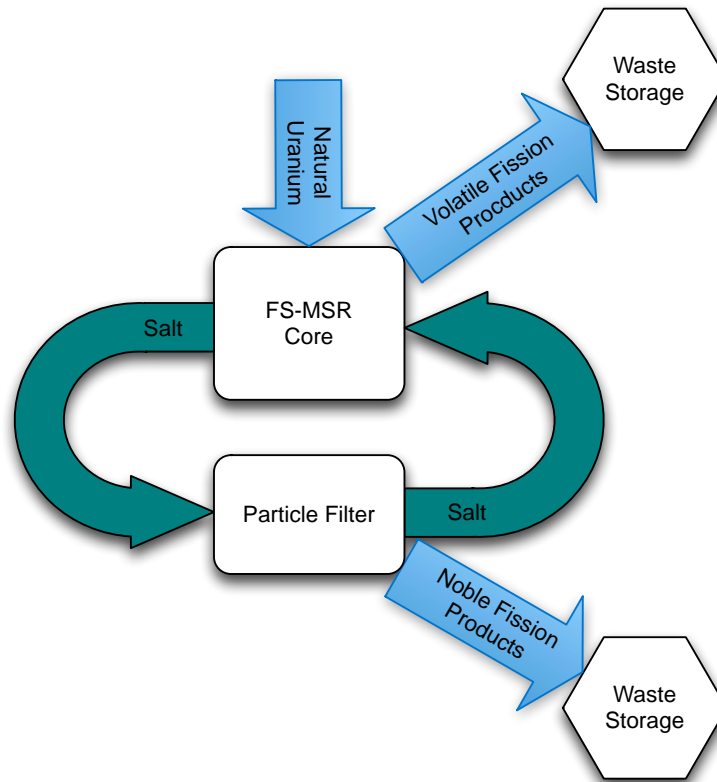

Fig. 11. Minimal fuel salt processing equilibrium fuel cycle.

3.6 High-Temperature Reactor Product Options

FS-MSRs are high-temperature reactors. Fluoride salts have high viscosity near their melting points, essentially exhibiting a glass transition as opposed to a sharp melting point. The same is generally true, to a lesser extent, for chloride melts. In order to avoid an overcooling transient, it is recommended that the lowest temperature in the primary circuit exceed the primary salt melt point by at least 50°C and preferably, for fluoride systems, by 100°C. The leading candidate chloride salt melts below 500°C, and the leading candidate fluoride salt melts slightly above 500°C. Consequently, the representative minimum fluoride salt reactor hot and cold temperatures are 650 and 600°C, respectively. Similarly, the representative minimum chloride salt reactor hot and cold temperatures are 600 and 550°C, respectively.

The high reactor temperature increases the efficiency of electricity production. As a high-temperature reactor class, FS-MSRs can support high-efficiency electricity production or thermochemical cycles. Electric power production cycle options include supercritical water, supercritical carbon dioxide, helium Brayton, and direct air Brayton cycles. The supercritical water power cycle is especially promising in that the requisite technology is being developed and deployed at fossil-fueled power plants. The uranium carbonate cycle (Fig. 12) for hydrogen production appears to be particularly well suited for coupling to high-temperature, low-pressure reactors as it requires heat input in the 650°C temperature range and does not involve high-pressure caustic chemicals.[31]
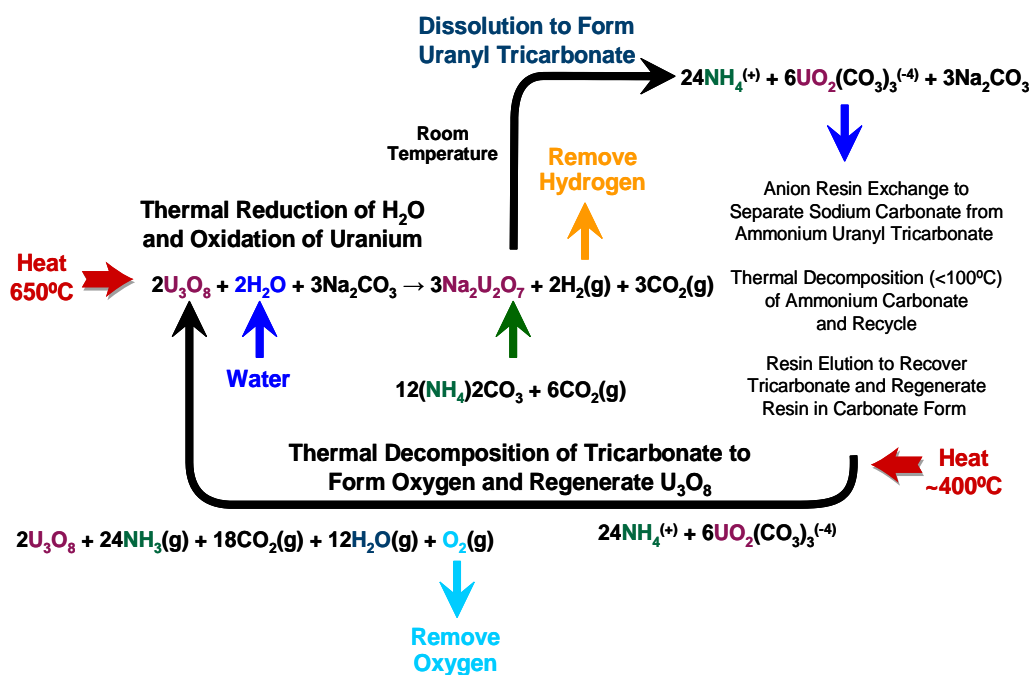
Fig. 12. Uranium carbonate cycle for hydrogen production.

The capability to efficiently produce large amounts of hydrogen enables high-temperature reactors to expand their role in meeting world's energy needs into hydrocarbon energy systems. An example thermochemical cycle for the production of gasoline using a high-temperature reactor, water, and carbon dioxide from a coal-fired power plant is shown in Fig. 13. The production of methanol from carbon dioxide (e.g., from flue gas) and hydrogen, as well as the conversion of methanol and additional hydrogen into gasoline, is already proven technology.[32, 33] Thus, a potential route to minimizing U.S. dependence on imported oil for gasoline is economically producing large quantities of hydrogen, which large-size, high-temperature reactors makes possible.
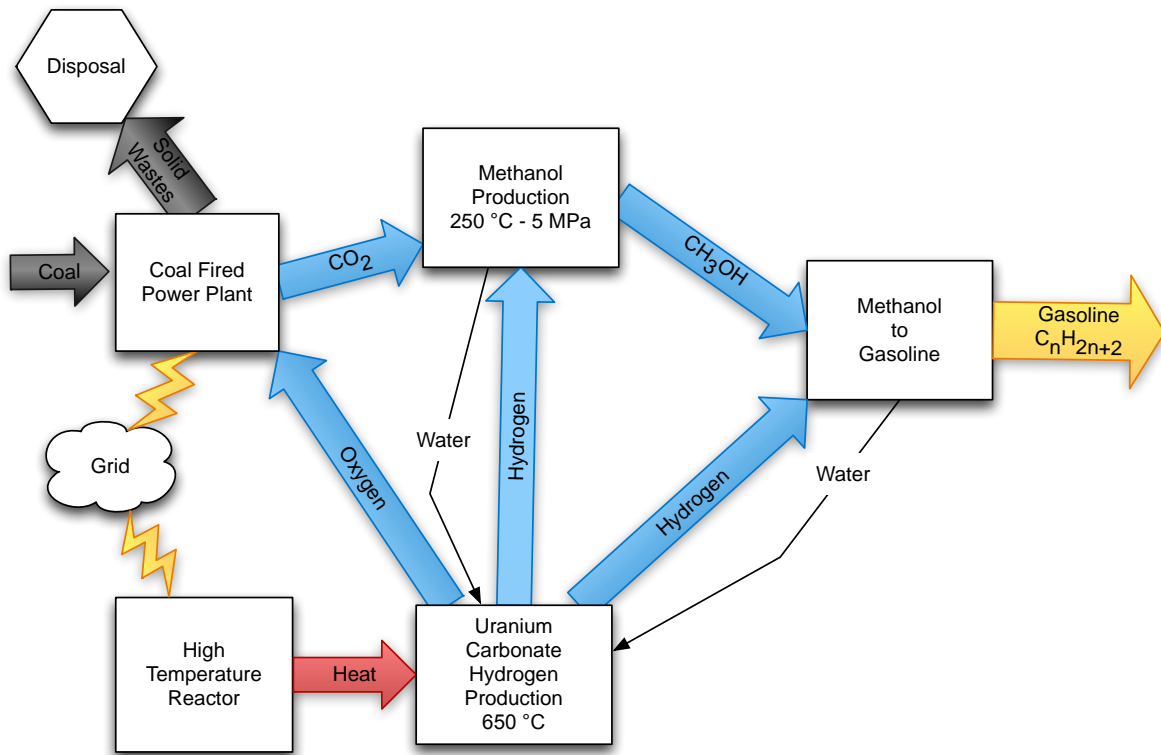
Fig. 13. High-temperature reactor thermochemical power cycle for the production of gasoline.

4.   FS-MSR PERFORMANCE CHARACTERISTICS

4.1   Key Parameter Overview

No fast-spectrum reactor has ever been cost-competitive with large LWRs. The resource sustainability and actinide-burning capabilities of fast-spectrum reactors have never been judged to be sufficiently economically important to bear the burden of the additional capital and operating costs necessary to provide the additional service. Fast-spectrum reactors also have not been able to enter the high-temperature carbon cycle market, as they have not been at the required temperature to efficiently generate the hydrogen necessary to re-hydrogenate petroleum or carbon dioxide. Additionally, the required fuel processing technologies, which have often been intended for integral deployment, have been judged to present a larger proliferation hazard than the open LWR fuel cycle.

FS-MSRs have the potential to address all of the negative aspects of fast-spectrum reactors while preserving their outstanding fissile resource utilization and waste disposal characteristics. FS-MSRs eliminate the expensive solid fuel fabrication and qualification aspects of heterogeneous core fast-spectrum reactors. Also, as higher-temperature reactors, they will have higher efficiency electricity generation. Additionally, because of the low reactivity of the salt and water, FS-MSRs are candidates for coupling to supercritical water power cycles, which are by far the most mature of the high-temperature power generation cycles. With the recent invention of the lower-temperature uranium carbonate–based

hydrogen production cycle, FS-MSRs would be able to efficiently generate hydrogen, enabling the plant to have a near-zero carbon balance while producing hydrocarbon fuel.

FS-MSR fuel has a uniform isotopic concentration of actinides, including highly burnt plutonium or uranium isotopes along with other minor actinides and fission products, making it undesirable for weapons production. The local fuel processing of the breeder and burner configurations also eliminates the possibility of diversion during transport. The fission product–saturated fuel salt of the no-heavy-metal separation converter reactor is highly self-guarding during transportation. In general, diversion of molten salt materials is difficult. The reactor operates as a sealed system with an integrated salt processing system that is technically difficult to modify once contaminated. The hot salt freezes at relatively high temperatures (450–500°C), requiring heated removal systems.

The no-heavy-metal separation converter cycle FS-MSR reactor presents a distinctive capability for a highly proliferation-resistant resource-sustaining fast-spectrum reactor. The potential lack of fissile material separation technology within a converter cycle FS-MSR has the potential to enable a fast-spectrum reactor that is exportable to nonfuel-cycle states without requiring a fuel return. Because of the ability of a fast-spectrum reactor to tolerate the accumulation of significant amounts of fission products, the only fuel processing that appears necessary for many years of FS-MSR converter cycle operation is capture of the fission gases (possibly extracted via helium sparging) and mechanical filtering of the noble metal fission products particles as they accumulate in the fuel salt.

4.2   Performance Comparison with Existing Reactor Classes

As liquid fueled, fast-spectrum reactors with several design options, FS-MSRs have several performance differences from the current LWR open fuel cycle. The liquid salt fuel also results in several performance differences from the sodium fast reactors.

The most obvious difference for an FS-MSR is that a solid fuel fabrication plant is not required. While a fissile material fluoride or chloride will need to be fabricated initially for any of the FS-MSR design variants, fabricating and qualifying solid fuel forms that include the varying amounts of minor actinides found in used LWR fuel is technically challenging, expensive, and as yet unproven. FS-MSRs essentially avoid the entire fuel qualification issue in that they are tolerant of any fissile material composition, with their inherent strong negative thermal reactivity feedback providing the control necessary to accommodate a shifting fuel feed stream.

FS-MSRs provide a unique potential for a fissile resource extension without requiring a fissile material separation process step. The potential lack of fissile material separation technology within a converter (or small breeding gain) cycle FS-MSR has the potential to generate a fast-spectrum reactor that is exportable to nonfuel-cycle states without requiring fuel return and extensive monitoring. As explained in the design options section of this report, the only fuel processing that appears necessary for many years of FS-MSR converter cycle operation is capture of the fission gases (possibly extracted via helium sparging) and mechanical filtering of the noble metal fission products particles as they accumulate in the fuel salt. The limited-separation FS-MSR is conceptually similar to the denatured MSR studied in the late 1970s (but with a slightly positive breeding ratio) in that fission products are allowed to build up in the salt, and the salt as a whole eventually is disposed of as waste.[34] The noble metals may actually be beneficial to FS-MSR operation (and so may not need to be removed) if they plate out as a protective layer on the structural material pressure boundary.

The high-level waste stream for breeder, burner, and converter FS-MSRs is substantially different from that for other reactor classes. Neglecting separation inefficiencies, only the short-lived fission products are removed from the FS-MSR fuel cycle for U-Pu breeders and TRU burners. For the minimal-separation small breeding gain reactor, the long-term waste stream contains only the reactor fuel salt at final shutdown. The shutdown salt will contain the remaining fissile materials necessary for criticality, the

dilutant salt, and the built-up fission products from long-term generation. Somewhat smaller amounts of fissile materials (compared with multiple LWR cores) will need to be disposed of from the shutdown limited-separation converter FS-MSR salt, as the continuous on-line refueling enables operation with minimal excess reactivity, and many years of operation are anticipated before the fuel salt must be discarded or reprocessed.

FS-MSRs can provide a high degree of passive nuclear safety while enabling fissile resource extension, maintaining high power output, and achieving high power density. This set of characteristics compares favorably with all other proposed reactor classes. The high degree of negative thermal reactivity feedback due to the large fuel salt coefficient of thermal expansion combined with the negative void reactivity feedback is a unique reactor characteristic. Also, the ability to passively drain the core into geometrically subcritical decay tanks that provide for passive decay heat removal (likely via heat pipes to the surrounding soil) provides a highly robust severe-accident response that compares favorably with the capabilities of solid-fuel reactors.

The containment wall thickness and consequent capital costs for FS-MSRs will be lower than those for other reactor types because mechanisms to generate pressure or explosive chemical mixtures within containment are lacking. The containment walls are only required to contain a low-pressure internal environment and endure when subjected to external seismic and impact stressors. Halide salts are chemically inert, so they do not have exothermic reactions with the environment (oxygen, water) as would hot sodium or hot zirconium. With a greater than 500°C margin to boiling, the halide salts also do not have a credible route to pressurizing containment as would a water-cooled reactor. FS-MSRs also do not have any hydrogenous material within containment; thus they cannot generate hydrogen.

FS-MSRs will require more expensive structural materials than LWRs because of their higher reactor temperatures and fast neutron flux tolerance requirement. However, because of the lower pressure, smaller amounts of the materials will be required. Overall, the material-expense-balance economics are as yet unknown. FS-MSRs will also require more expensive components and instruments because of both the higher temperatures and the requirement to accommodate remote maintenance.

FS-MSRs will have an increased potential for small-scale radioactive materials leaks because the highly radioactive fuel material is liquid and comparatively more accessible than solid fuels. The leak probability will be increased for on-line reprocessed reactor design variants as a result of more intensive fuel salt manipulation. The fuel salt reprocessing manipulation will need to take place within a hot-cell type environment, providing an additional containment structure within the primary reactor containment. The additional hot-cell containment structure will increase the plant capital costs.

FS-MSRs will have increased operation costs, especially for on-line salt processing design variants, because remote handling is required for maintenance. Long-handled tools were demonstrated during the MSRE program; and, after the primary coolant loop was flushed (as would be required for maintenance), only small amounts of fuel would remain within the loop. Nonetheless, the containment environment for an FS-MSR would be more radioactive than that for a solid-fuel reactor, making increased remote handling and inspection technology necessary.

The spent fuel transport would be different for a minimal-separation FS-MSR converter reactor. The salt would be solidified into canisters that would be temporarily stored locally in storage pool and then placed within a dry transport cask for either long-term storage or transport to a fuel processing facility.

4.3 Used Fuel Disposition, Separations, and Waste Management

Only the minimal-separation converter reactor has a modified open fuel cycle and thus a fuel disposition requirement. The solid fission products from all design variants of the reactor are captured on zeolite beds that can be cemented into solid form and allowed to decay in near-surface repositories. The FS-MSR minimal-separation converter reactor would need to be shut down when the reactor vessel reached

maximum allowed exposure, a fissile material solubility limit was reached, or sufficient amounts of fission products built up in the salt to raise its melting point beyond ~550°C. Upon final shutdown, the fuel salt would be first heavily poisoned and then pumped into subcritical geometry decay canisters. The canisters would be allowed to decay in a local light-water or inexpensive salt spent pool storage pool. The salt would then be sent for either deep geologic disposal or centralized fuel processing.

The fuel salt separation technologies proposed for FS-MSRs are based upon the technologies developed and demonstrated in the MSR program for fluoride salt options and in the IFR program for chloride salt options.

4.4 Resource Sustainability

The breeder and waste burner design variants represent fully closed fast-spectrum reactors. As such, they make use of the nearly the entire uranium resource (neglecting system losses). Homogeneous reactors may require a slightly smaller initial charge of fissile material to initiate the cycle because they lack structural absorbing materials in the core. However, FS-MSRs do have nearly half of their fissile material inventory outside of the core in the heat transfer loop.

4.5 Proliferation Resistance and Physical Protection

In the context of safeguards, the fuel has a uniform isotopic concentration of actinides, including highly burnt plutonium or uranium isotopes along with other minor actinides and fission products. Other aspects of an FS-MSR design also reduce the proliferation risk, including the following:

1. There is little fertile material in the fuel (<1%) for the low-conversion-ratio concepts, so only very small amounts of additional TRU are created by the reactor.

2. MSRs generally have online processing, which returns TRU to the reactor, requiring no off-site transportation.

3. Front-end processing could be co-located with onsite LWR used fuel processing, with separated products directly introduced into the reactor.

Diversion of molten salt materials is difficult. The reactor operates as a sealed system with a highly integrated salt processing system that makes it difficult to modify the system. The hot salt has been shown to freeze, requiring heated removal systems. A loss of fuel salt would remove fissile material from the reactor and could impact reactor operation. During operation, the TRU always remains in the hot, radioactive salt. However, FS-MSRs, as described earlier, may be poor candidates for use in nonfuel-cycle states because the integral processing system may raise concerns. Also, methods of inspection and materials accountability for liquid cores have not been fully developed.

4.6 Economics

A confident assessment of the economic performance of an FS-MSR is not yet possible. Technology, regulatory requirements, and market conditions have changed significantly over the 40 years since the economic assessments accompanying the MSBR; therefore, the cost inferences drawn from the earlier work have such large error bands that they provide little guidance. Additionally, the neutron spectrum of the present evaluation alters the fuel cycle both in and outside the power plant site sufficiently that direct analogies to other reactor concepts are challenging. The most challenging aspect of reporting a cost for an FS-MSR, however, arises from the concept flexibility. A no-heavy-metal reprocessing design variant has a plant layout much different from that of a full-recycle plant intending to directly accept used LWR fuel as its fuel source. Similarly, a plant intending to produce gasoline as its primary product has an entirely different power cycle compared with an electricity generator.

Overall economic tendencies, however, can be estimated by comparing FS-MSR attributes with those of other nuclear power systems. A summary of FS-MSR attributes and their cost implications is provided in Table 2. A primary cost metric for any power plant is its thermal efficiency. FS-MSRs, as high-temperature power plants, are anticipated to have 45–48% thermal efficiencies, a 12–15% efficiency advantage over LWRs. As refueling for an FS-MSR would be performed on-line, the plant availability would be expected to eventually, once maintenance techniques were developed and matured, surpass that for an LWR.

Table 2. FS-MSR economic performance attributes and cost implications

Difference relative to existing LWRs	Consequence	Effect on costs/revenues
Homogeneous liquid fuel/primary coolant	No fuel testing, qualification, or fabrication	Lower fuel acquisition cost
	No cladding as fission product barrier	Potential higher capital cost for replacement fission product barrier
	No cladding-based burnup limits	Higher electricity generation revenue per unit mass of fuel
	No fuel handling equipment or pool storage facilities	Lower capital cost without fuel handling/storage
		No operations and maintenance (O&M) cost attributed to spent fuel handling/storage
	No irradiated cladding or matrix material in ultimate waste stream	Lower disposal cost at end of plant life
	Multiple potential fuel sources (LWR UNF, DU, NatU, RepU, Th)	Higher fuel acquisition cost if separations required (LWR used nuclear fuel)
		Lower fuel acquisition cost if natural material used (NatU, Th)
	Large temperature reactivity coefficient	Lower capital cost for passive and simpler active reactor safety systems
	No cladding- or matrix-based temperature limits in accident scenarios	Lower O&M cost attributed to active reactor safety systems
	Safe shutdown possible through geometry control in accident scenarios	
	Higher operating temperature	Higher capital cost for compatible materials
		Higher electricity generation revenue per unit of heat generation
		Potential thermochemical industrial use
	Higher primary coolant volumetric heat capacity	Lower capital cost for pipes, pumps, and heat exchangers

Difference relative to existing LWRs	Consequence	Effect on costs/revenues
	Highly radioactive, fissile-bearing primary coolant	Higher capital cost for compatible materials and safe-geometry system design
		Higher O&M cost attributed to primary coolant system
	Visually transparent, low-pressure, chemically stable coolant	Lower capital cost for pipes and vessels
		Lower O&M attributed to overall system maintenance
On-line fuel processing	Greater control of fuel/primary coolant chemistry	Higher capital cost for salt treatment plant
		New O&M cost attributed to salt treatment plant
		Potential lower O&M cost attributed to chemistry control
	Flexible input fuel chemical form	Potential lower fuel acquisition cost based on market availabilities
	Flexible input fuel isotopic content	
	Continuous separation of fission products (and reduction of source term in accident scenarios)	Higher capital cost for waste handling facilities
		New O&M cost attributed to waste handling facilities
		Lower capital cost for passive and simpler active reactor safety systems
		Lower O&M cost attributed to active reactor safety systems
		Potential lower disposal cost at end of plant life
	Potential for safeguards concerns with separated material	Higher capital cost for security improvements
		Higher O&M cost attributed to security
Fast neutron flux	Material corrosion	Higher capital cost for compatible materials

When a thermal energy cycle's high temperature is further from its low temperature, an increase in the low temperature has proportionately lower impact on the cycle efficiency. Because FS-MSRs are high-temperature reactors, they can reject heat through comparatively smaller, hotter dry cooling systems than can LWRs. The increased compatibility with dry cooling systems also decreases the land acquisition cost for an FS-MSR in that riparian property is significantly more expensive than land without water access.

Another cost difference for an FS-MSR is the lack of uranium enrichment required to operate the fuel cycle. Further, having a low-pressure, fully passive shutdown and decay heat removal system is anticipated to significantly reduce overall FS-MSR plant costs. However, development of a plant design

is required to develop a system capital cost model based upon actual component and system costs, and the magnitude of the savings is thus not yet known.

As much of the fuel reprocessing technology for a chloride salt FS-MSR is similar to that developed for the IFR (although perhaps configured in continuous instead of batch form), the cost for fission product separation is expected to be similar. FS-MSRs, unlike sodium fast reactors, do not require solid fuel fabrication (approximately 10% of the total cost) or fuel qualification and thus have a fuel cycle cost advantage.

4.7   Safety

FS-MSRs have the potential for excellent passive safety characteristics. FS-MSRs have a negative salt void coefficient (expanded fuel is pushed out of the core) and a negative thermal reactivity feedback that avoids a set of major design constraints in solid-fuel fast reactors. A passive core drain system activated by a melt plug enables draining the radioactive inventory into geometrically subcritical drain tanks that are passively thermally coupled to the environment. FS-MSRs have a low operating pressure even at high temperatures; and FS-MSR salts are chemically inert, thermodynamically lacking the energetic reactions with environmental materials seen in other reactor types (hot zirconium and sodium with water). FS-MSRs do involve more intensive manipulation of highly radioactive materials than other reactor classes and thus small spills and contamination accidents appear to be more likely with this reactor class.

5.   CONCLUSIONS AND FUTURE WORK

FS-MSRs have the potential for highly desirable fuel cycle characteristics with excellent resource sustainability, actinide waste disposal, favorable economics, and desirable safety characteristics. The minimal-separation converter form of the FS-MSR also represents a unique fast-spectrum concept that does not involve transport of fissile materials (after the initial reactor charge) or on-site fuel separation. Further, the liquid fuel avoids the fuel radiation damage material limitations that severely hamper alternative fast-reactor long-lived core concepts. The FS-MSR reactor class limitations remain unknown at present because of the concept immaturity.

Overall, FS-MSRs are quite immature with the only exception being a smattering of the component technologies at commercial or near-commercial levels of development. Perhaps most important, no FS-MSR system conceptual design has been developed. Without at least a preconceptual level integrated system design, it is not possible to evaluate the reasonableness of design options, and considerably uncertainty accrues to the overall system viability.

As a homogeneous core reactor, the FS-MSR has among the simplest of any core design models. However, the on-line refueling and waste separation processes complicate the neutronic modeling. Although the Multiregion Processing Plant Code [35] was developed during the MSBR program, it appears to have been lost; its functionality will need to be recreated to evaluate material balances in the primary salt system.

All of the safety features have been evaluated only on a general-principles basis. No accident scenarios or general design criteria have been evaluated. Also, FS-MSRs do not have an accepted safety modeling methodology or evaluation codes.

Chloride salts appear to be advantageous for producing a harder neutron spectrum and thus improving actinide burning and breeding. However, the structural material information underpinning a chloride salt reactor is uncertain. Chloride salt alloy compatibility testing is recommended as an early-phase activity. Chloride salt isotope separation would improve the eventual repository compatibility of the carrier salt.

More detailed technical and economic analysis of chloride isotope separation technology is recommended to assess the viability for selectively using ^{37}Cl.

The salt reprocessing technologies presented have been demonstrated only with similar salts. EBR-II fuel processing employed lithium chloride. A chemical process analysis of the fuel processing steps needs to be performed to determine the reasonableness of the overall schemes proposed.

Phase diagrams of actinide-bearing chloride salts have not been published. Thermochemical modeling of the salt thermal and hydraulic parameters (e.g., melt point, viscosity, thermal conductivity, density) is necessary to enable development of a reliable reactor system performance and safety models.

The fuel-processing portion of an FS-MSR will have among the highest radiation doses of any ex-core environment. Remote handling processing equipment, as well as maintenance approaches and instrumentation, will need to be developed and demonstrated.

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