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A White Paper: Potential Disposition Options for a Liquid-Fueled Molten Salt Reactor At INL

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SUMMARY

The molten salt reactor (MSR) design concept has been the focus of significant research and development efforts, both in the U.S. and around the world, with an emphasis on the reactor core. There are generally two types of MSRs, liquid-fueled MSRs that have the fuel dissolved within the salt and solid fueled MSRs that only use the molten salt as a coolant. The range of conceptual designs and the flexibility of operations of MSRs results in a wide range of potential radioactive waste streams from liquid-fueled MSRs; all of which require safe and secure storage and eventual disposition. This white paper will 1) survey the range of liquid-fueled MSR designs, 2) recognize the implications of design on the resulting radioactive waste streams, and 3) identify the definitions and regulations that apply to the safe and secure management, storage, and disposal of these streams. Research in the management and disposal of MSR radioactive waste streams is young and the focus of intense research that requires more funding and time to become mature enough to fully support design decisions. Once a specific design is selected, a more detailed management and disposition effort must be performed.

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ACRONYMS

AEA – Atomic Energy Act of 1954 as amended

CFR – Code of Federal Regulations

DOE – U.S. Department of Energy

FP – fission product

GMODS – Glass Material Oxidation and Dissolution System

GTCC – Greater-Than-Class C

HLW – high-level radioactive waste

LLW – low-level radioactive waste

LWR – light-water reactor

MSR – molten salt reactor

NWPA – Nuclear Waste Policy Act of 1982 as amended

SNF – spent nuclear fuel

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1. INTRODUCTION

The molten salt reactor (MSR) design concept has been the focus of significant research and development efforts, both in the U.S. and around the world. Much of this effort has been focused on the front end of the MSR fuel cycle, with an emphasis on the reactor core. There are generally two types of MSRs, liquid-fueled MSRs that have the fuel dissolved within the salt and solid-fueled MSRs that only use the molten salt as a coolant. This brief white paper will focus on liquid-fueled MSRs.

These MSRs have been designed to operate in the fast, epithermal, and thermal neutron energy ranges and can be breeder[a] or actinide-burning[b] reactors. This operational flexibility leads to a wide range of potential fuel compositions, such as thorium, uranium, plutonium, as well as the transuranic content from the spent nuclear fuel (SNF) of other reactors. The salts in which the fuel is dissolved can also vary with the fluoride salts and chloride salts frequently used. Each salt presents a different set of chemical and operational characteristics, with chloride salts typically used in fast neutron spectrum MSRs. Another unique aspect of these reactors is the ability to treat (i.e., remove or add isotopes to) the fuel salt while the reactor is online. While volatile fission products (FPs) in the gaseous phase may potentially leave the salt and some transition metals produced as FPs may be insoluble in the salt and naturally plate out, the actinides can be removed from the salt using various chemical processes.

Due to the unique online treatment options, as well as the liquid fuel, these MSRs present a unique set of challenges for disposition, including how to define and categorize the radioactive waste from these reactors. For example, without a fixed fuel geometry the concept of the traditional spent fuel assembly does not apply. As a result of the online treatment, there is also potential for debate regarding whether the spent actinide-bearing fuel salt should be classified as high-level radioactive waste (HLW) or as SNF. Furthermore, depending on the treatment operations, some portion of the MSR fuel salt could qualify as low-level radioactive waste (LLW) based on the radiological hazard/risk represented by the salt waste stream, which would expand the options for final disposition outside of a deep-mined geological repository.

The range of conceptual designs and the flexibility of operations of MSRs result in a wide range of potential radioactive waste streams from liquid-fueled MSRs; all of which require safe and secure storage and eventual disposition. Furthermore, research in the management and disposal of MSR radioactive waste streams is young and the focus of intense research. It requires more funding and more time to become mature enough to fully support design decisions. This white paper will 1) survey the range of liquid-fueled MSR designs, 2) recognize the implications of design on the resulting radioactive waste streams, and 3) identify the definitions and regulations which apply to the safe and secure management, storage, and disposal of these streams. Once a specific design is selected, a more detailed management and disposition effort must be performed.

Section 2 presents a brief survey of liquid-fueled MSR technologies with attention to the proposed salt, initial fissile, final fissile, and TRU fuel compositions and proposed online treatment operations. Section 3 identifies the various radioactive waste streams from liquid-fueled MSRs and the options for waste management. Section 4 discusses the various classes of radioactive waste and how they may apply to the identified MSR waste streams, and Section 5 presents the conclusions.

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[a] Breeder reactors are designed to produce more fissile material than they consume.

[b] Actinide-burning reactors are designed to incinerate long-lived radioactive isotopes.

2. SURVEY OF MSR TECHNOLOGIES

As mentioned, the range of MSR design concepts feature a variety of fuel and salt compositions, neutron spectra, and operational modes as illustrated in Figure 1. A few examples (but not a complete list) of molten salt technologies are presented in Table 1. The fissile component of the fuel salt is either ^{233}U, ^{235}U, or Pu. This fissile material is either introduced in the form of fresh enriched U, separated Pu, or TRU separated from the SNF from other reactors or in the case of ^{233}U and ^{235}U, bred inside the MSR. The fertile component of the fuel for breeder reactors is either Th or ^{238}U. There are various molten salts into which the fissile and fertile fuel may be mixed. In general, these are either fluoride-based or chloride-based salt systems. The traditional fluoride systems use LiF_2-BeF_2 or NaF-ZrF_4 in the primary loop and LiF-NaF-KF in the secondary loop to avoid the activation of Be. It should be noted that the Li-based salts require enrichment (>99.9%) of ^{7}Li to reduce parasitic neutron capture and limit tritium production, due to the large (n,^{3}H) reaction cross-section of ^{6}Li. Several options exist for chloride salt systems featuring pure compositions or mixtures of alkali- and alkaline-earth chloride salts. Chloride-based salt systems require enrichment of ^{37}Cl to enable high breeding ratios and limit production of ^{36}Cl from the neutron capture reaction (n,γ) in naturally occurring ^{35}Cl. ^{36}Cl has a half-life of 3×10^5 years which poses disposal concerns.

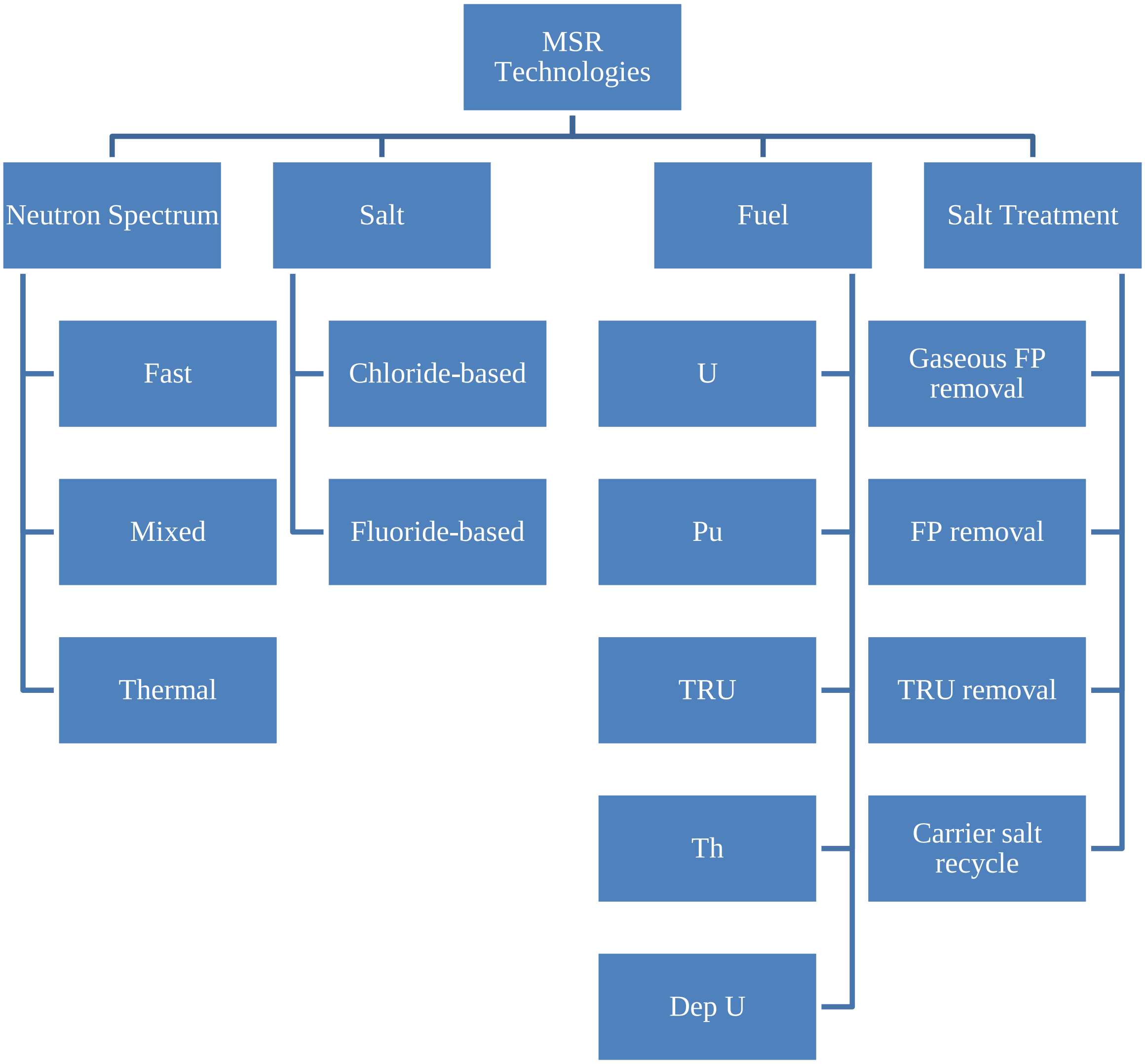

Figure 1: The range of different MSR technologies

Table 1: Examples of MSR designs

Reactor/Developer	Actinide Loading	Salt	Neutron Spectrum	Operational mode
FLiBe Inc. (Electric 2015)	Th and ^{233}U	Fluoride	Thermal	Breeder
Copenhagen Atomics (Pedersen 2017)	Th and ^{233}U/LWR SNF	Fluoride	Thermal	Breeder/ Burner
Alpha Tech Research (Alpha Tech n.d.)	Th and ^{233}U	Fluoride	Thermal	Breeder
Indian Molten Salt Breeder Reactor (Vijayan et al. 2015)	Th and ^{233}U	Fluoride	Thermal	Breeder
Chinese TMSR-LF (Xu 2017)	Th and ^{233}U	Fluoride	Thermal	Breeder
ThorCon Power (Devanney 2015)	Denatured Th and LEU	Fluoride	Thermal	Burner
Terrestrial Energy (LeBlanc 2018)	Denatured ^{235}U	Fluoride	Thermal	Burner
MOSART (Ignatiev 2017)	^{238}U and ^{239}Pu	Fluoride	Fast	Breeder/Burner
Molten Salt Fast Reactor (Serp et al. 2014)	Th and U	Fluoride	Fast	Breeder
TransAtomic (Betzler 2017)	Actinide	Fluoride	Epithermal	Burner
Seaborg Waste Burner (Seaborg 2019)	Th and Actinide	Fluoride	Epithermal	Burner
TerraPower, (TerraPower n.d.)	^{238}U and ^{239}Pu	Chloride	Fast	Breeder
Elysium Industries (Elysium n.d.)	^{238}U and ^{239}Pu	Chloride	Fast	Breeder
Moltex (Moltex Energy 2018)	U, LWR SNF, Pu, Th	Chloride	Fast	Breeder/Burner
Dual Fluid Reactor (Huke et al. 2015)	U	Chloride	Fast	Burner

3. RADIOACTIVE WASTE CLASSIFICATION

The disposition options for radioactive waste streams from MSR operations will be dictated by the classification of the radioactive waste streams as either SNF, HLW, LLW, TRU waste, or byproduct material. The applicable definitions for the various categories of radioactive waste in the U.S. are established by the U.S. Department of Energy (DOE), Atomic Energy Act of 1954 (AEA) (Atomic 1954), and the Nuclear Waste Policy Act of 1982 (NWPA) (Nuclear 1982). The following are the terms as defined in the relevant laws.

- **Spent nuclear fuel (NWPA 1982)** – *Fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing.*
- **High-level waste (NWPA 1982)** – *(A) the highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and (B) other highly radioactive material that the Commission, consistent with existing law, determines by rule requires permanent isolation.*
- **Transuranic waste (DOE Order 435)** – *radioactive waste containing more than 100 nanocuries (3700 becquerels) of alpha-emitting transuranic isotopes per gram of waste, with half-lives greater*

than 20 years, except for: (1) High-level radioactive waste; (2) Waste that the Secretary of Energy has determined, with the concurrence of the Administrator of the Environmental Protection Agency, does not need the degree of isolation required by the 40 CFR Part 191 disposal regulations; or (3) Waste that the Nuclear Regulatory Commission has approved for disposal on a case-by-case basis in accordance with 10 CFR Part 61.

- **Low-level waste (NWPA 1982)** – *radioactive material that (A) is not high-level radioactive waste, spent nuclear fuel, transuranic waste, or by-product material as defined in section 11e(2) of the Atomic Energy Act of 1954 [42 U.S.C. 2014(e)(2)]; and (B) the Commission, consistent with existing law, classifies as low-level (C) radioactive waste.*
- **Byproduct material (AEA 1954)** – *(1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to the radiation incident to the process of producing or utilizing special nuclear material.*

There are four classes of LLW: Class A, Class B, Class C, and Greater-Than-Class C (GTCC). These classes are based on waste form requirements and the activity per unit volume of specific radioisotopes, as defined in the Title 10, Part 61 of the Code of Federal Regulations (10 CFR 61). Depending on the online treatment operations and subsequent fuel salt separation processes, it is possible that some of the waste streams from MSRs could be classified as LLW, which would expand the options for final disposition outside of a deep-mined geological repository.

There are four categories of byproduct material: (1) radioactive material that results from the fission, or splitting apart, of enriched uranium or plutonium in nuclear reactors; (2) tailings or waste produced by processing uranium or thorium from ore; (3) certain processed radium-226 or material that becomes radioactive in a particle accelerator used for a commercial, medical, or research activity; and (4) a naturally occurring radioactive source that is processed to increase its concentration and that the Commission decides could pose a threat to people and the environment similar to that of radium-226.

Disposition of some MSR radioactive waste as TRU or LLW may be possible following the recent DOE interpretation of HLW (DOE 2018, DOE 2019). DOE interprets that reprocessing waste may be determined to be non-HLW if the waste meets either of the following two criteria: *1) does not exceed concentration limits for Class C low-level radioactive waste as set out in section 61.55 of title 10, Code of Federal Regulations, and meets the performance objectives of a disposal facility; or 2) does not require disposal in a deep geologic repository and meets the performance objectives of a disposal facility as demonstrated through a performance assessment conducted in accordance with applicable requirements*.

MSR radioactive waste streams may also contain hazardous waste, in which case it would be classified as a mixed waste. Hazardous wastes are materials known or tested to exhibit one or more of the following hazardous traits: ignitability, reactivity, corrosivity, or toxicity as per 40 CFR 261. Fuel salt mixtures containing hazardous compounds will be designated as mixed waste. Mixed wastes are regulated by both the Resource Conservation and Recovery Act (Resource 1976) and AEA. For example, lead in its metallic form is one of the eight metallic elements that RCRA monitors along with arsenic, barium, cadmium chromium, mercury, selenium, and silver due to their toxicity.

4. MSR RADIOACTIVE WASTE STREAMS

Regardless of the MSR design concept and materials selected, the radioactive waste streams from MSRs can be categorized in the following groups:

1. Off-gas streams
2. Salt waste streams[c]
    a. Direct disposal without separation
    b. Salt separation
3. Other waste streams.

Some of these radioactive waste streams have important differences from light-water reactor (LWR) radioactive waste streams. For example, LWR waste management strategies do not have to directly account for many short-lived isotopes that decay away long before the LWR SNF is ready for disposal. However, for some MSRs, these short-lived isotopes are volatile and will separate from the salt in the off-gas stream and, as such, must be directly addressed. Some concepts have no off-gas system and plan to let pressure build up in a plenum above the core. Other designs feature delay tanks that allow the radioactive component of these gases to decay before release. Others have cryogenic systems to capture and store volatile components in tanks. Nevertheless, waste management approaches and technologies with the potential for application in disposition have been investigated for these potential types of waste (Riley et al. 2019). The following paragraphs present brief overviews of these approaches.

4.1. Off-gas Streams

The removal of volatile FPs such as ^{135}Xe from the salt stream must be effectively managed. The off-gas stream contains volatile FPs and their decay daughter nuclides, particulates, tritium, water, oxides, nitrides, halides, trace amounts of salt in the form of aerosols, and noble gases. Processes exist for the capture, immobilization, and disposal or release of all off-gas waste species. This includes scrubbing with aqueous hydroxide and non-aqueous molten hydroxide systems. The halides may also be captured using solid sorbents. The noble gases may be captured using cryogenic distillation, silver-zeolite, activated carbon, and metal-organic frameworks. Additionally, a delay bed could be used to allow the short-lived isotopes to decay away before treating the remaining gases. Several of the noble gases could be separated for commercial value. The helium could also be recycled back into the MSR as a headspace purge gas. Tritium production can be limited by enrichment of ^{7}Li as mentioned. Several options exist for the removal of the remaining tritium produced including capture in core carbon components, capture in packed beds of porous carbon outside the core, conversion and removal as T_2O, and/or sparging with Ar or He. For a more complete review of these technologies please see (Riley et al. 2019). Most of the off-gas wastes would likely be classified as LLW.

4.2. Salt Streams

There are two general approaches for management of the fuel salt. The entire fuel salt can be disposed of as a single stream without separation (i.e., direct disposal), or the fuel salt can be separated into different streams with each stream treated and dispositioned as appropriate based on the stream characteristics (ie.,

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[c] Including fuel-bearing salt streams

salt separation). While increasing operational complexity, the separated salt stream approach is advantageous as it allows the removal of FPs; recycling of U, Pu, and TRU; and recycling of F and Cl in the salt. The recycling of F and Cl is attractive due to the high effort and cost of producing high purity ^{7}Li and ^{37}Cl for use in these systems. For a more complete review of these technologies please see (Riley et al. 2019). Unseparated fuel salts intended for direct disposal would be classified as HLW. Common waste forms for HLW includes glass, ceramic, glass ceramic, glass-bonded ceramics, and ceramic-metal composites. When considering separated salts, the FP-bearing salt waste would most likely be classified as HLW. A separated TRU-bearing salt waste could be classified as TRU waste. It is possible to make a case that a separated salt stream with low amounts of radioactivity could be handled solely as an industrial chemical waste, although this is unlikely to be accepted. It is more likely that these waste streams would be treated as LLW.

4.2.1. Direct disposal without separation

The water solubility of certain species in the fuel salt requires immobilization of the fuel salt prior to disposal in a deep-mined geological repository to prevent the migration of radioactive species into groundwater. Fabrication of a single waste form that can retain all the various species within the fuel salt would be very difficult (Riley et al. 2019). Waste forms suitable for the direct disposal of fuel-bearing salts include mineral-based waste forms, glass waste forms, and metal-composite waste forms.

4.2.1.1. Mineral-based waste forms

The three most studied mineralogical waste forms include titanate, sodalite, and apatite. Of the three, sodalite has been demonstrated as a waste form option for wastes from pyroprocessing Experimental Breeder Reactor II fuel; the resulting product from this process is a glass-bonded sodalite ceramic waste form. The ability to incorporate different species into mineral-based waste forms varies for the halides, alkali earth, alkaline earth, rare earth, and actinide elements in the fuel salt (Riley et al. 2019).

4.2.1.2. Glass waste forms

The types of glass for direct disposal are limited for several reasons. For example, the traditional silicate-based glass waste forms have low solubility limits for halogens at the operating temperatures used for making silicate glass, resulting in a low retention of halides. Thus, non-traditional, non-silicate glass would be required to increase salt loadings. Many of the properties needed for waste form qualification are undefined for these systems and require further study. Tellurite glass has been used for the immobilization of salt waste from pyrochemical operations. These systems have shown potential to accommodate chloride and iodide salts, but other salts tend to phase-separate into immiscible phases. Furthermore, tellurite glasses are expensive to make, reducing their attractiveness as a waste form. It is unlikely that fluoride-based salts could be used for a suitable waste form due to low chemical durability in these systems (Riley et al. 2019).

4.2.1.3. Metal-composite waste forms

Ceramic-metal (commonly referred to as cermet) composites are made by dispersing the waste particles homogenously within a metal matrix. The metal matrix performs both a containment and heat dissipation function and results in a higher-density waste form, all advantages over the glass or mineral-based waste forms. The metal matrix can consist of several metals such as Ti, Al, Mg, Ni, Cu, Fe, Co, Mo, Nb, Zr, as well as mixtures of these metals. Metal-composite waste forms appear promising as a waste form for disposal of salt wastes (Riley et al. 2019).

4.2.2. Salt separation

Several salt separation techniques can be used to partition the MSR fuel salt for the purpose of separating FPs, recycle TRU, recycle portions of the salt, or facilitating the fabrication of waste forms. These include reductive extraction, oxide precipitation, distillation, melt crystallization, dehalogenation, phosphorylation, ion exchange, and the Glass Material Oxidation and Dissolution System (GMODS) (Riley et al. 2019).

4.2.2.1. Reductive extraction

Reductive extraction can be used to selectively remove certain components in the salt mix, such as the actinides. Several studies have demonstrated high separation efficiency for actinide and actinide surrogates in chloride and fluoride salts. It has been shown that the TRU component can be coextracted and separated from the rare earths due to high separation factors and differences between the species (Riley et al. 2019).

4.2.2.2. Oxide precipitation

Oxide precipitation has been shown to remove rare earth FPs from molten chloride salts by forming oxychlorides and oxides, which are insoluble in the salt and so precipitate and settle at the bottom. A similar process with fluoride salts seems unlikely, due to no formation of oxyfluorides or oxides in calculations involving fuel salt species. With the addition of species with a high affinity for fluorides, formation of oxyfluorides or oxides can be promoted (Riley et al. 2019).

4.2.2.3. Distillation

Vacuum distillation is the process of separating species within a mixture based on the differences in the vapor pressures under certain conditions. As part of the Molten Salt Reactor Experiment program, vacuum distillation was applied to volatize and recover the ^{7}LiF and BeF_2 for recycling the salt. With modern manufacturing capabilities, the possibility now exists for multistage separation. Reactive distillation uses an additive to react the target species into a species that be separated in vacuum distillation (Riley et al. 2019).

4.2.2.4. Melt crystallization

Melt crystallization is the process of separating species based on differences in melting temperatures. FP separation from chloride salts has been demonstrated, and experimental data collected for fluoride salts (Riley et al. 2019).

4.2.2.5. Dehalogenation

Dehalogenation is the process of removing the chloride or fluoride species. Since these species represent the major molar component of the salt mixture, dehalogenation offers the advantage of using existing waste forms that have already been demonstrated at the industrial scale and reducing the potential for chlorine or fluorine generation by radiolysis in the waste form. Dehalogenation in chloride-based salts has been demonstrated via the addition of phosphate precursors (e.g. H_3PO_4 and $NH_4H_2PO_4$). While this has not been demonstrated for fluoride-based salts, the thermodynamic favorability of fluorine over chorine in the reactions suggest the approach is likely to be successful. Dehalogenation resulting in oxide-based FPs enables a wide range of superior waste form options compared to halide-based species (Riley et al. 2019).

4.2.2.6. Phosphorylation

Phosphorylation is the process by which species within the fuel salt can be converted into phosphates with the advantage of having phosphate-based waste forms for rare earth and actinide phosphates. These phosphates can be pressed into a waste form and then mixed with a glass binder to create a glass composite or be immobilized in a phosphate glass form (Riley et al. 2019).

4.2.2.7. Ion exchange

Ion exchange can be used to selectively remove specific species from the fuel salt mix. Results of studies in this area have shown that FPs (Cs and Sr) can be removed from a chloride-based salt mixture (Riley et al. 2019).

4.2.2.8. Glass material oxidation and dissolution system

The GMODS process was developed to directly vitrify radioactive, mixed, and chemical wastes in a single-step process. In the GMODS process, organics are destroyed, and heavy metals and radionuclides are oxidized and immobilized in a glass waste form (Riley et al. 2019).

4.3. Other Radioactive Waste Streams

Other radioactive waste streams include metal, carbon, operating, and decontamination and decommissioning waste streams. The various metal components used in MSRs will become activated and coated with salt and FPs, which are expected to plate out of the salt. Many of the FPs can be removed using various treatments. The carbon streams from most MSRs will largely be composed of polycrystalline graphite used as moderator or reflector material. During operation, the contact between the graphite and fuel salt mixture will result in some salt penetration into the pores of the graphite as well as the embedding of FPs, decay products, and small amounts of uranium. Operating radioactive waste includes equipment, material containers, personal protective equipment, air filters, waste cleanup resins, and glove box gloves. For a more complete review of these technologies please see (Riley et al. 2019). In general, these radioactive wastes can be treated and disposed of in the same manner as similar wastes from other nuclear reactors. These waste streams are likely to produce some HLW, but with a majority being LLW. It is possible to produce some TRU waste if a separated TRU salt stream was produced as part of MSR normal salt treatment operations.

5. CONCLUSIONS

In conclusion, the MSR design concept has been the focus of significant research and development efforts both in the United States and around the world. Much of this effort has been focused on the front end of the MSR fuel cycle, with an emphasis on the reactor core. The MSR design concept provides reactor design options that can produce very different designs for liquid-fueled MSRs. These MSRs have been designed to operate in the fast, epithermal, and thermal neutron energy ranges and can be breeder or actinide-burning reactors. The salts systems used in liquid-fueled MSRs are typically fluoride-based or chloride-based salt systems. Multiple options for online treatment of the fuel salt via addition or removal of various isotopes are available. As a result, these MSRs present a unique set of challenges for disposition, including how to define and categorize the waste from these reactors. The wide range of waste streams from MSRs can be categorized into three groups: (1) off-gas waste streams, (2) actinide and fission product-bearing salt waste streams, and (3) other waste streams. Research in the management and disposal of MSR waste streams is young and the focus of intense research that requires more funding and time to become mature enough to fully support design decisions. Off-gas waste streams may present the highest technological risk since the mobility of gaseous components require a thorough understanding of the chemical and physical processes which influence their behavior. Continued research directed toward the study of glass and ceramic formulations especially related to loading capacities is required. Finally, waste form development needs to be performed while being focused on the predicted final regulatory disposal requirements.

The off-gas waste stream contains volatile FPs, their decay daughter nuclides, particulates, tritium, water, oxides, nitrides, halides, trace amounts of salt in the form of aerosols, and noble gases. Processes exist for the capture, immobilization, and disposal or release of all off-gas waste species. The off-gas wastes would likely be classified as LLW. The actinide and FP-bearing salt streams are likely to be HLW and would require disposal in a deep-mined geological repository. For direct disposal, the water solubility of certain species in the fuel salt requires immobilization of the fuel salt to prevent migration of radioactive species into groundwater. Fabrication of a single waste form that can retain all the various species within the fuel salt would be very difficult, although suitable waste forms for direct disposal exist for certain species in the fuel-bearing waste form. Several salt separation techniques can be used to partition the MSR fuel salt into different waste streams, for which suitable waste forms can more easily be produced. Here, FP-bearing waste streams would be classified as HLW, waste salts bearing only actinides may be classified as TRU waste. Salt separation provides the option of greatly reducing the HLW volume with most of the non-FP and actinide-bearing salt waste likely to be classified as LLW, for which more disposition options exist. Other waste streams include metal, carbon, operating, and decontamination and decommissioning waste streams. In general, these wastes can be treated and disposed of in the same manner as similar wastes from other nuclear reactors.

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