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Cradle to grave: the importance of the fuel cycle to molten salt reactor sustainability

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Advanced reactor technologies are being considered for the next-generation of nuclear power plants. These plants are designed to have a smaller footprint, run more efficiently at higher temperatures, have the flexibility to meet specific power or heating needs, and have lower construction costs. This paper offers a perspective on molten salt reactors, promoted as having a flexible fuel cycle and close-to-ambient pressure operation. A complexity introduced by reducing the reactor footprint is that it may require low-enriched fuel for efficient operation, available from enrichment of the feed salt or by reusing actinides from existing used nuclear fuel (UNF). Recycling UNF has the potential to reduce high-level waste, if done correctly. Release limits from UNF processing are stringent, and processes for waste reduction, fission gas trapping, and stable waste-form generation are not yet ready for commercial deployment. These complex processes are expensive to develop and troubleshoot because the feed is highly radioactive. Thus, fuel production and supply chain development must keep abreast of reactor technology development. Another aspect of reactor sustainability is the non-fuel waste streams that will be generated during operation and decommissioning. Some molten salt reactor designs are projected to have much shorter operational lifetimes than light-water reactors: less than a decade. A goal of the reactor sustainability effort is to divert these materials from a high-level waste repository. However, processing of reactor components should only be undertaken if it reduces waste. Economic and environmental aspects of sustainability are also important, but are not included in this perspective.

KEYWORDS

molten salt reactor fuel cycle, salt fuel preparation, waste generation, fuel salt waste disposition, salt processing

1 Introduction

The challenge to develop a large-scale response to increases in CO₂ emissions and climate change has provided an impetus to develop options for nuclear fission for power generation that address the major issues with the prevalent light-water reactor (LWR) designs. The fleet of LWRs has performed with a strong safety record for several decades, especially when considering dose to the public. Yet, accidents have occurred, highlighting the need to develop designs that have enhanced safety margins, passive safety features, and increased tolerance to fault scenarios (Seghal, 2012). These features have become even more desirable as recent events at the Zaporizhzhia power plant in Ukraine indicate the need to include deliberate sabotage to the list of possible conditions that might impair plant operation (Kurando, 2023). Options for passively safe designs include those with

encapsulated tristructural-isotropic (TRISO) fuel (Morris et al., 2004), and low-pressure operation with coolants such as molten metal (King et al., 1991) or molten salt (Holcomb et al., 2021). The latter class of reactors is the subject of this essay. Benefits of molten salt fueled reactors are that they operate slightly above ambient pressure and generally preclude water within containment/confinement, thereby limiting high-energy reactor failure scenarios, airborne transport of radionuclides, and potential dose to the public. Challenges include working with salts that are air sensitive and need to be purified to reduce chemical reactivity (McFarlane et al., 2019).

A decision to adopt a radical change in reactor design must be placed in the context of the production and usage of power. This decision will be made by the electrical utilities and their stakeholders, including customers and government agencies as informed by regulatory review. These decisions also must consider sustainability of the design's supply chain, including the production of nuclear fuel through to the disposition of the waste from the reactor (Krall et al., 2002). Such lifecycle considerations accompany any large-scale adoption of new technologies, one example being the case of electric vehicles and their reliance on new battery technologies (Yang et al., 2022). Molten salt reactors (MSRs) operate at high temperatures with fluoride or chloride salt coolants that can be chemically reactive if not carefully controlled. Development of new materials to contain the salt and research into the chemical stability of these materials in a chloride or fluoride environment are current topics of nuclear research (Raiman and Lee, 2018). The disposition of reactor materials after reactor shutdown and decontamination also must be considered in supply chain feasibility (Riley et al., 2019).

2 Fuel and salt availability

MSRs can burn a variety of actinides, including the standard ^{235}U (Holcomb et al., 2022), higher actinides such as ^{239}Pu and ^{241}Am (Bhomik et al., 2023) recovered from used nuclear fuel (UNF), as well as ^{233}U derived from a thorium fuel cycle (Bogetic et al., 2016). Depending on the design, an MSR may be operated on a single load of fuel, with online processing or addition of fuel, as a breeder, or as a waste or actinide burner. Thus, there have been descriptions of how MSR technology could be coupled with recycling of UNF from other types of nuclear reactors (Moyer et al., 2022). Many reactor designs are being developed in the United States (US) and around the world: some have already taken regulatory steps towards deployment. An example of the latter includes getting permission to construct a prototype, as detailed on the US Nuclear Regulatory Commission Agencywide Documents Access and Management System website (US Nuclear Regulatory Commission, 2023). These demonstration reactors do not rely on currently available commercial fuel suppliers but are collaborating with fuel development programs at the US Department of Energy national laboratories to provide a supply of ^{235}U in the correct form. The choice of fuel and enrichment level will determine the size and configuration of the reactor, preparation of salt for the reactor, safeguards and security requirements, and downstream waste dispositioning. Therefore, design and planning to scale up both the reactor and the fuel cycle must be done in parallel. The advanced reactor development communities in the US

and globally have started to address this complex problem by holding workshops that include researchers and other stakeholders, the findings from which will be publicly available (i.e., Rose and Ezell, 2024; Espartero and Grassi, 2024).

The other aspect of salt availability that must be considered is that MSRs depend on having a carrier and/or coolant salt that is transparent to neutron flux. Consequently, fluoride salt reactors that have lithium-based carriers need salt that is enriched in ^7Li to reduce the production of ^3H through activation (Harrison et al., 2016). Chloride-based carrier salts need enrichment in ^{37}Cl to reduce the production of ^{36}Cl , a long-lived isotope that complicates waste-form disposal, and other activation byproducts such as sulfur (Pigni, 2023). Thermal diffusion has been studied for chlorine isotope separation (Kranz and Watson, 1953), but commercial production is not yet realized. Recovery of ^6Li from military applications or fusion could be leveraged to supply ^7Li for MSRs, but coupling these different applications may be difficult for both technical and regulatory reasons (US Department of Energy Office of Nuclear Physics, NSAC Isotopes Subcommittee, 2015).

3 Waste generation

One major concern with nuclear power is the accumulation of UNF and difficulties in siting repositories for high-level waste (HLW) (Krall et al., 2002). Although the feasibility of underground disposal of UNF has been thoroughly investigated, public concerns endure regarding the safety and efficacy of permanent disposal at prospective repositories such as Yucca Mountain in the US (Birkholzer et al., 2023). These disparate concerns may arise from the handling of UNF, the safety of transporting UNF canisters long distances to a central location, as well as the indefinite hazard presented by long-lived isotopes sequestered in a dynamic geosphere over many millennia. In the absence of an HLW repository, on-site UNF storage has been the default option (Sindelar, 2022). Although on-site storage is a short-term solution, it has provided the nuclear community time to address the issue of permanent waste disposal.

In the case of MSRs, waste generation will occur at all stages of the lifecycle, as summarized in Table 1. Some of these wastes are analogous to debris from LWR dispositioning, and existing tracking and disposal pathways could be used. Other waste materials are unique to MSRs and will require special consideration (Krall et al., 2002; Riley et al., 2019).

Upstream processing, including salt enrichment, mixing, and transportation to the reactor site, will contaminate vessels and machinery with nuclear materials. Inventories at the production site must be tracked to fulfil material balance accountability (MBA) requirements, which will be stringent if highly enriched uranium or higher actinides are included in the fuel mixture. Production of ceramic fuel requires similar MBA controls. However, waste from fuel production may meet the low-level waste (LLW) acceptance criteria, unless recycled actinides are part of the fuel mixture. UNF has been considered as a source of material for MSRs because of their flexible fuel cycle. Actinides have been recovered from UNF by pyroprocessing at US Department of Energy installations such as Idaho National Laboratory (Fredrickson and Yoo, 2021).

TABLE 1 Waste generation from molten salt reactors.

Process	Waste form	Disposition
Fuel salt preparation (from unirradiated material)	Contaminated equipment	<ul style="list-style-type: none"> Decontaminate and reuse equipment, LLW
Fuel salt preparation (from irradiated material)	Contaminated equipment	<ul style="list-style-type: none"> Decontaminate and reuse equipment
	Radiological waste streams (solid debris, liquid, or captured in off-gas)	<ul style="list-style-type: none"> Consolidate and stabilize waste streams, LLW, HLW
Reactor operation	Contaminated equipment	<ul style="list-style-type: none"> Decontaminate and reuse equipment if possible, LLW, HLW
Fuel handling	Unused fuel salt	<ul style="list-style-type: none"> Recover and recycle unused salt, LLW
	Used fuel salt	<ul style="list-style-type: none"> Stabilize and contain used fuel salt or separated components, HLW
Reactor operation	Materials replaced during operation	<ul style="list-style-type: none"> Out-of-core, minimal contamination, LLW
Maintenance		<ul style="list-style-type: none"> In-core, high contamination, Stabilize and contain for storage/disposal
Reactor operation	Volatile radionuclides	<ul style="list-style-type: none"> Confine radionuclides until activity is minimal. Capture and stabilize long-lived isotopes. Control decay heat
Off-gas	Spent filters, capture media for off-gas	<ul style="list-style-type: none"> Off-gas components and in-core vessels require remote handling as HLW debris. Consolidate and stabilize for storage/disposal
Shutdown & Maintenance	Materials replaced during shutdown (sensors, off-gas filters)	<ul style="list-style-type: none"> From out-of-core, minimal contamination, LLW
	Volatile products of radiolysis	<ul style="list-style-type: none"> From in-core, high contamination, Stabilize and contain for storage/disposal Minimize conditions contributing to radiolysis
Decommissioning and Decontamination (D&D)	Fuel, carrier, flush salts drained from facility	<ul style="list-style-type: none"> Recycle valuable salt components, stabilize used salt against radiolysis, convert to insoluble waste form(s)
	Salt contacted metals and structural materials	<ul style="list-style-type: none"> Decontaminate salt wetted materials if feasible. Compact to reduce volume
	Salt contacted carbon wastes	<ul style="list-style-type: none"> Off-gas components handled as described for reactor operation
	D&D wastes	<ul style="list-style-type: none"> D&D LLW, HLW, depending on contamination
	Off-gas wastes	

Reactor operation will generate wastes from maintenance of the reactor systems such as the off-gas and online processing. The design will dictate the waste generation. For instance, the off-gas system could be enclosed for some reactor designs or have limited throughput (Dunkle et al., 2023), but most will have a flowing cover gas that continuously removes volatile radionuclides similarly to the Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (Guymon, 1973). Online processing could include removal of insoluble materials such as graphite particles and noble metals or actinide recovery from the liquid phase. The rationale for online processing has been reviewed in regard to molten salt fuel qualification (Holcomb et al., 2020; 2022). Implementation will depend on a number of factors including relevant regulations and so will be specific to each jurisdiction. Because of salt contamination, wastes from maintenance and operation are likely to be HLW and must be managed accordingly.

Decommissioning, shutdown, and decontamination wastes from MSRs will include materials that are wetted with salt and those that are not. The latter can be handled similarly to LWR reactor materials: volume minimization will be an important consideration (Vestal et al., 2023). The salt-wetted components from MSRs may include the reactor vessel, graphite moderator, filters, and equipment for fuel introduction and sampling. A review of wastes from MSRs delves into each of these categories and

suggests ways of managing them for disposal or recycle (Andrews et al., 2021). The feasibility of these options must include safe operation, resistance to diversion, and HLW volume. The salts are water soluble, which may permit decontamination to remove some of the more active fission products (e.g., intermediate half-lived ^{137}Cs and ^{90}Sr) to reduce the decay heat load associated with HLW.

For the fuel salts, indefinite storage is not an option as it is for ceramic fuel. Experience with MSRE has shown that salts drained from the reactor can undergo radiolysis for decades, requiring active monitoring and degassing (National Research Council, 1997). The salts' water-soluble nature complicates waste disposal: they must be processed to an insoluble form. Because some processing is necessary, a once-through fuel cycle is unlikely to be feasible for commercial operation. Reprocessing methods were reviewed by Fredrickson and Yoo, and they highlight the effectiveness of electrorefining for uranium recovery from the converted Experimental Breeder Reactor II (Fredrickson and Yoo, 2021). Capture and reuse of isotopically enriched chlorine and lithium salts may also be desirable. Chemical processing may be avoided by incorporating the UNF salt into an insoluble matrix, such as an intimate mixture of salt and metal, termed a halmet (Del Cul et al., 2018). This approach may be a simpler alternative to chemical or pyrochemical processing. Cermet technology has been developed

TABLE 2 Comparison of MSR and LWR considerations for UNF recycle and waste handling.

	LWR	MSR
Interim storage	On-site dry cask	Drain tank
Incentives for back-end processing	Removal of high-decay heat producing fission products, or long-lived fission products to ameliorate repository requirements	Recovery of enriched carrier salts, value-added fission products, or high-decay heat producing fission products
	Recovery of value-added radionuclides	
UNF Processing	Requires several steps including removal of the matrix surrounding the fuel, cladding and dissolution of the ceramic fuel	Salt can be heated for pyroprocessing or electrorefining
Waste form	Robust clad ceramic fuel matrix can be made more resistant to radionuclide leaching by further encapsulation and geoengineered barriers	Fuel salt is not suitable for permanent disposal without additional processing and stabilization
Volatile and semivolatile fission products (Isotopes of I, Kr, and Xe, Te, Tc, ^3H , ^{14}C)	Need to be tracked during back-end processing	Largely released and confined in the off-gas system during reactor operation. Disposition of semi-volatile fission products with complex redox chemistry requires further study

over several decades and has been applied to UNF as well as irradiation targets (Kobisk et al., 1981; Robinson et al., 2020). Using halmets to stabilize UNF salt is at an early stage of development. For example, copper, used as the metal phase for a cermet, is typically heated to 1,085°C when forming a mixed phase with UO_2 . At this temperature, salt components will volatilize, so a lower temperature process must be considered with a different metal, such as aluminum. Preparation of UNF powder for halmet production will be an engineering challenge because of the requirement for contamination control and MBA considerations. Although halmets have been proposed and patented (Aaron et al., 2012), the durability of these materials has not yet been tested.

4 Conclusion

This paper briefly surveys considerations for the fuel cycle of MSRs from a US perspective. Reactor design and prototype development is more advanced than the supporting fuel preparation and waste management constructs. The choice of fuel to be burned in an MSR will directly affect its design, so these aspects must be considered in tandem. Because irradiated salts used in MSRs undergo radiolysis during storage, the ability to store UNF indefinitely is more complicated for MSRs than for spent LWR fuel, requiring active monitoring and mitigation of acidic gases that may be generated during storage. Because salts are water soluble, they must be processed before disposal, so a once-through fuel cycle is unlikely to be feasible. Stabilizing matrices such as halmets have been proposed to permit direct disposal of salts with minimal processing. Table 2 summarizes the differences in fuel cycle considerations for LWR and MSR designs. In the case of MSRs, the fuel cycle cannot be decoupled from the reactor design and has the potential to be as complex as the reactor itself. Consequently, research and development efforts in all aspects of the MSR fuel cycle—including linkages to the current nuclear materials supply chain, development of fuel processing facilities with or without recycled actinides, and development of insoluble waste forms for permanent disposal—must be considered in tandem. Sharing of information globally is important to addressing complex fuel cycle issues related to MSR technology.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

Author contributions

JM: Conceptualization, Funding acquisition, Writing—original draft, Writing—review and editing.

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Conflict of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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