

Initial Calculations for Source Term of Molten Salt Reactors

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Abstract

This paper provides an overview of the current MSR design space and lists unique features of the various designs under consideration. Some general considerations for source terms calculation for Molten Salt Reactors (MSRs) are explained. Applicability and limitations of terminology currently defined for legacy light water reactor (LWR) systems are discussed in the view of MSRs and the need for updated terminology is discussed. Calculations carried out for the Molten Salt Reactor Experiment (MSRE) are discussed with a qualitative comparison to the designs presented. The nature of the fission products (FPs) and actinides for Low enriched uranium, thorium and fast U/Pu fuel cycles employed in representative molten salt reactor systems are discussed. Computational results are obtained from a code (Serpent2) with online reprocessing. Divergence in source terms when fission product bubbling is demonstrated. The source release for each molten salt reactor during postulated accidents is also presented.

Keywords: MSR, Source term, Source release, MSR accident scenarios, Molten salt burnup calculations, LFTR, MCFR

1. Introduction

Generation IV designs are a class of novel nuclear reactor concepts that promise passive safety and improved economics compared to the current large light water reactors (LWRs). One notable representative of Generation IV designs is a class of Molten Salt Reactors (MSRs). These reactors use a liquid salt that dissolve its fuel and circulate it through the core and heat exchangers. The fluid fuel of MSRs provides distinct advantages over LWRs, such as the ability to operate at ambient pressure and high temperatures, increased fuel utilization, and passive safety features. Even though there are many possible advantages to MSRs, they are not yet at the level of technological readiness needed for large scale deployment. As of the writing of this paper, one major area of research that needs to be further explored is MSR source terms' behavior.

The source term is the amount of radioactive material that can be potentially dispersed into the environment from a nuclear reactor [1]. Proper understanding of a reactor's source term is necessary for reactor licensing, but a large portion of previous research has been focused on solid fuel reactors. Solid-fuel reactors keep actinides and fission products (FPs) contained in the fuel pellet. In MSRs, however, the actinide fuel and fission products (FPs) are in a continuous transit through the primary circuit. Due to the flow velocity of the order of meters per second, the salt mixture is considered to be homogeneous. Additionally, the behavior of these elements depends on the chemical characteristics of the various

possible carrier salts. As such, the source terms for MSRs can vary widely – not just from LWRs, but also from each others – depending on the particulars of a specific reactor design and fuel cycle.

There is only one example of power-operated molten salt fuel reactors that provide insight into FP behavior in MSRs, being the Molten-Salt Reactor Experiment (MSRE), which is the only MSR operated at power for an extended time. The MSRE was an 8MW(th) graphite-moderated thermal- spectrum reactor built and operated at the Oak Ridge National Laboratory (ORNL) from 1960 to 1969 [2]. Fuel reprocessing was kept at a minimum, with only an off-gas system for gaseous FPs. The experience gained with the MSRE provides valuable knowledge on how fission products behave in an MSR using a fluoride salt.

FPs in the MSRE can be sorted into three distinct chemical groups. Even though carrier salt chemistry and operating conditions may cause some slight differences in behavior, the categorization can be applied to most MSR designs. The first of these groups is the noble gas fission products. These FPs will bubble out of the fuel-salt to an off-gas system with or without design intervention. Next are molybdenum, technetium, ruthenium, silver, and others that form the noble metals. The noble metals are not expected to form stable fluorides in an MSR, and they tend to deposit on various surfaces as well as the salt-gas interface during operation. Finally, there are those elements that form stable fluorides and remain in the fuel salt such as rubidium, cesium, strontium, barium, and the lanthanides [3]. In the case of salt spillage, these isotopes would remain in the salt and form the major constituent of the source term [4]. Additionally, all actinides are expected to form stable fluorides and remain in the salt under normal operating conditions.

Startup companies in both the US and abroad perusing the MSR technology, and they have proposed a wide variety of designs that use their own fuel cycle, carrier salt composition, and fuel reprocessing schemes. While the underlying physics is similar, these unique design aspects mean that the behavior of FPs, and hence the resulting source terms, differ from design to design. Additionally, for a design to be successfully licensed and brought to market, an understanding of possible radioactive releases for various accident scenarios is needed. This paper presents an overview of three prospective MSR designs/fuel cycles with the objective of providing some preliminary calculations for source terms from a break in fuel salt primary containment. To make this methodology as widely applicable as possible, specific design choices for each reactor are kept to a minimum. Representative calculations from Serpent 2.1.31[5] are presented for fuel burnup of each design. A qualitative overview of the source terms is presented in conjunction with analyzing the degree of applicability of MSRE data with regard to credible accident scenarios in these designs. In addition to presenting the information currently available for studying source terms in MSRs, the purpose of this work is to articulate some gaps in knowledge when it comes to understanding the behavior of these source terms.

2. MSR Design Space Considerations

The MSR design space encompasses many important choices both inside and outside the core that could affect the source term. For this paper, only the primary fuel salt circulating inside the reactor core is considered, while any on-site or off-site reprocessing or waste storage is not considered and would have separate source terms associated with them. Nevertheless, given the variety of fuel types, the hardness of the neutron spectrum, and chemical characteristics of the carrier salts under consideration, there is potential for divergence in the source terms when comparing reactor core designs. For example, long-

lived transuranic isotopes existing in the reactor, which form a significant portion of the source term, are expected to be lower for designs that employ the thorium fuel cycle. Additionally, the chemical behavior of FPs can be significantly different given the composition of the carrier salt used. For the purposes outlined in this report, three reactor designs representative of the fuel cycle and neutron spectrum parameter space are considered. While particular design choices will reflect the specifics of the means for source release, the qualitative source terms should not differ significantly, thus providing a means for evaluating radioactive release for postulated accidents. The rest of this section presents an overview of these reactor designs and details the characteristics that are important for determining their respective source terms.

2.1. Thermal Spectrum, Single Fluid MSR

As the name implies, a thermal spectrum, single fluid (TSSF) MSR has only a single salt mixture that goes into the moderated core region, and the primary fissile is typically LEU. These reactors tend to be graphite-moderated and use a fluoride carrier salt. A significant portion of the source term is the result of FP accumulation. Given the liquid nature of the fuel, all MSRs have the potential to remove much of these FPs through reprocessing. If there is no reprocessing, then FPs and actinide source terms will increase as the reactor is operated; however, the fuel composition will depend on the spectrum, salt composition, and other operational parameters. If this salt contains lithium, there would be significant tritium production via Li-6 neutron absorption. One way to mitigate this source would be to enrich the lithium used in the carrier salt. However, there is still tritium production due to neutron capture in the salts and ternary fissions.

2.2. Liquid Fluoride Thorium Reactor

As part of ORNL's work on MSRs during the 1960s, two breeder reactor designs were studied. These were namely the single-fluid [6, 7] and two-fluid [8] Molten Salt Breeder Reactor (MSBR). These designs feature a modular reactor core consisting of graphite moderator columns with machined channels to allow for salt circulation. The single-fluid design employs a 7LiF-BeF₂-ThF₄-UF₄ (71.6-16-12-0.4 mole%) salt in the primary system. A mechanical pump circulates the salt through the graphite columns of the core and then through an intermediate counter-current heat exchanger. The two-fluid design, on the other hand, circulates a 7LiF-BeF₂-UF₄ (68.5-31.3-0.2 mole%) fuel salt through the core along with a 7LiF-BeF₂-ThF₄ (71-27-2 mole%) fertile salt through interstitial channels in the core matrix and a surrounding under-moderated blanket region. In both cases, the Th-232 converts to U-233 upon absorbing a neutron and undergoing two beta decays. Since U-233 is not readily available, these reactors would probably start with an initial load of low-enriched uranium fuel before transitioning to burning the bred U-233. These designs aim at utilizing the thorium fuel cycle, and they have the advantage of much lower long-lived actinide production. Furthermore, the designs elaborated in Ref [6, 7, 8] provide a continuous fuel reprocessing system to remove FPs. In the two-fluid design, the presence of two separate salts in the primary system means that there are different source terms associated with each fluid. The fuel salt source term consists mainly of FPs and some actinides. The U-233 formed in the fertile salt is contaminated with U-232 formed via (n, 2n) reactions whose daughters are strong gamma emitters and

constitute the primary source terms of concern in the fertile salt. However, this study only considers the fissile salt mixture of a two-fluid design.

2.3. Molten Chloride Fast Reactor

One of the earliest chloride salt based fast breeder designs was carried out by Dr. M. Taube at the Swiss Eidg. Institut für Reaktorforschung (EIR), Wrenlingen [9]. Taube's reactors utilize fuel in the form of molten chlorides of PuCl_3 as the fissile material, UCl_3 as fertile, and NaCl as carrier. Some multi-fluid designs, such as Taube's design, would be required to be coupled with a continuously operating reprocessing plant, while other designs that have only one fluid would not. Fast spectrum molten salt designs are not limited to these design choices. Ref [10] explores the options for fuel carrier salt in alone. This paper however focuses on fast spectrum, molten chloride designs.

Recently, the U.S. DoE awarded Southern Company and its affiliates a grant for development of Molten Chloride Fast Reactor (MCFRs). While the technical specifics of the design remain undisclosed, the award marked the first time the DoE has committed any funding towards an MSR concept [11]. Chloride fast reactors share many of the same advantages of thermal-spectrum fluoride salt reactors discussed above. Most importantly, a U/Pu breeding cycle can only be realized in the fast spectrum due to improved capture-to-fission ratio in plutonium and other actinides. A representative salt composition, coupled with the knowledge that these designs employ a fast neutron spectrum, can be used to make high quality estimates of the potential actinide and fission product concentrations and thus the resulting source terms.

3. Chemical Behavior of Source Material

Unlike in a solid fuel reactor, the FP in an MSR are flowing, reacting, and possibly volatilizing during normal operation. This can lead to significant divergence of the source term from an LWR fuel cycle. Crucial to understanding these differences is the chemical behavior of the various elements present in the salt. However, a complete prediction of the behavior of each element is beyond the scope of this paper. There are many design minutia and operating conditions that could have a significant impact on species transport. Additionally, the high amount of radiation, fissioning, and short-term decay in an actively used fuel salt further complicates predictive models [12]. However, the chemical behavior of source material can be understood in broad terms where key design features can be accounted for. For instance, the fluid fuel of MSRs allows for online fuel processing to remove FP products. Specifically, MSRs that are fueled with uranium fluoride have the fluoride volatility process available to them, which enables simple, proven, full processing [13].

The MSR designs considered in this study utilizes online fueling, but the online reprocessing of FPs is not evaluated in detail. In liquid fuel reactors, burnt fuel is continually replenished either by feeding new fuel or breeding in the fertile material, and the fuel is continuously mixed in solution. Therefore, the term fuel burnup does not apply to MSRs in the way that it does to LWRs since there is no itemized amount of energy that can be directly associated with a specific mass of fuel. Related to burnup is the energy produced per unit mass of fuel. The standard unit for power density for many burnup calculators is energy produced per tonne heavy metal. This is misleading for MSRs since there may be a significant amount of fuel salt circulating outside of the core region. In addition to these terms, there is other reactor

terminology that is primarily used in an LWR design space that is not applicable to liquid fueled reactors. For instance, a reactor meltdown in LWRs is not possible in MSRs, as the fuel is already molten. By the same token, since the primary coolant is the fuel, MSRs do not have a Loss-of-Coolant accident in the traditional LWR sense. These are severe accidents for LWRs since FPs trapped in the solid fuel pellets have a chance to volatilize if the solid fuel melts. Liquid fueled reactors, such as MSRs, would have some continuous volatilization. This difference in operation makes the primary points of interest for severe accident study a break in either the core where large inventories of radionuclides are kept (explored in this study) or in the off gas removal and storage (not explored in this study). As a result, there is a need to update technical vocabulary to be more applicable to MSRs.

Additionally, in all cases, the xenon bubbles out of solution. Thus, there may be no need for excess reactivity to compensate for fuel burnup and poisoning from FPs [4]. There can also be spectral effects when Xe-135 is removed, and this should be accounted for in burnup calculations. For this reason, the refuel regime of each reactor design is to maintain criticality and to minimize the need for control reactivity.

Figure 1 is a periodic table of elements, color-coded to show the classification of fission products (FPs) based on their general chemical behavior. The legend indicates four categories:

- Gaseous:** Light blue background.
- Always soluble:** Green background.
- Sometimes soluble:** Yellow background.
- Insoluble:** Red background.

The elements are arranged in the standard periodic table format, with the Lanthanide and Actinide series shown at the bottom. The color-coding is as follows:

- Gaseous (Light Blue):** Hydrogen (1), Helium (2), Neon (10), Argon (18), Krypton (36), Xenon (54), Radon (86), and Oganesson (118).
- Always Soluble (Green):** Lithium (3), Sodium (11), Potassium (19), Rubidium (37), Cesium (55), Francium (87), Barium (56), Radium (88), Strontium (38), Calcium (20), Scandium (21), Yttrium (39), Zirconium (40), Niobium (41), Molybdenum (42), Technetium (43), Ruthenium (44), Rhodium (45), Palladium (46), Silver (47), Cadmium (48), Indium (49), Tin (50), Antimony (51), Tellurium (52), Iodine (53), and Astatine (85).
- Sometimes Soluble (Yellow):** Beryllium (4), Magnesium (12), Calcium (20), Strontium (38), Barium (56), Radium (88), and Rf (104).
- Insoluble (Red):** Boron (5), Carbon (6), Nitrogen (7), Oxygen (8), Fluorine (9), Silicon (14), Phosphorus (15), Sulfur (16), Chlorine (17), Vanadium (23), Chromium (24), Manganese (25), Iron (26), Cobalt (27), Nickel (28), Copper (29), Zinc (30), Gallium (31), Germanium (32), Arsenic (33), Selenium (34), Bromine (35), Krypton (36), Xenon (54), and Radon (86).

Figure 1: Fission product classification in the MSRE

As stated in the introduction, FPs can be classified by their general chemical behavior. These groups defined during the MSRE are gaseous, soluble, semi-soluble (depending on operating conditions), and insoluble. This organization is shown in Figure1. In the event of a release of fuel salt, for the most part, gaseous fission products will volatilize and possibly escape the liquid fuel. The soluble will remain in the salt. Insoluble FP will plate out through the reactor system and remain in the vessel. Finally, the semi-soluble will either remain in the salt or plate out on the system [3]. Actinides are expected to behave similarly to soluble FP; Tritium is a notable exception to every one of these groups. At the operating temperature of an MSR, the hydrogen will most likely traverse through the metal of the vessel along the

heat gradient during continuous operation of the reactor. These assumptions can be applied to each fluoride reactor described in this paper. For added simplicity, it is assumed that FPs in the chloride salt fast reactor also have a similar chemical behavior during operation. A detailed exploration of the redox chemistry is beyond the scope of this paper.

4. Burnup Calculations

The models utilized in a previous study [14] utilize infinite lattices in a versatile computational system SCALE 6.1[15], which treats the fuel salt as a solid, and this leads to inaccuracies when accounting for gaseous FPs. Although, fission yield is not directly affected by salt chemistry, the neutron absorption rate of poisons such as Xe-135 is, and gaseous daughters in decay chains impact methods for FP calculations. Recent advancements in computational tools enable online reprocessing in ChemTriton, but as of the writing of this paper, it is still under development [16]. In this paper, the models are updated and run in Serpent 2.1.31 [5]. Serpent allows online and continuous reprocessing through the use of Chebyshev Rational Approximation Method (CRAM), which is used in this study. This section will elaborate on the assumptions used in modeling the three MSR designs. An overview of the design parameters used in Table 1. For all cases, the burnup steps are 7 days and the numerical parameter are set such that at the beginning of cycle, the errors on fission and captures of the major actinides is less than 1%. All input and output files are stored in a Github repository¹.

Inputs	TSSF MSR	LFTR	MCFR
Pitch	15cm	14cm	-
Moderator	Graphite (1.8 g/cc)	Graphite (1.8 g/cc)	-
Carrier salt	LiF-BeF ₂	LiF-BeF ₂	NaCl
Primary fissile	LEU	U-233	U/PU
Temperature of fuel	900K	900K	900K
Density of fuel	3.353 g/cc	2.034 g/cc	3.509 g/cc
Breeder salt	-	LiF-BeF ₂	-
Gas removal rate	1%/s	1%/s	1%/s
Other FP Removal	-	0.1%/s	-

Table 1: Design parameters used in Serpent

In the thermal spectrum, U-235 fueled MSR case, a hexagonal lattice with a pitch of 15 cm and a fuel salt channel of radius 2.7 cm is modeled with periodic boundary conditions to simulate an infinite lattice. The moderator is graphite with a 15ppm boron impurity. The fuel salt is LiF-BeF₂-UF₄(72-16-12 mole%). The lithium is highly depleted such that Li-7 isotope represent 99.998% of the lithium mass, to reduce neutron absorption and tritium production, as is customary in MSRs with lithium bearing carrier salt. The initial

¹ Github repository: https://github.com/awheel13/MSR_source

enrichment of the uranium in the fuel salt is 1.3%. This lattice is critical at startup with a conversion ratio approximately 0.6. Since new fuel is mixed with burnt fuel salt, the refueling salt needs to be a higher enrichment than the fuel used to start up the reactor. For the calculations presented in this paper, the refueling salt has the same chemical makeup as the startup fuel salt but with a uranium enrichment of 10%. The results from calculations show that a mass flow rate of refueling salt should be 3.5%/day of the core volume to maintain criticality for the power density of 20 MW/MTHM.

The LFTR is also modeled as an infinite hexagonal lattice for this paper, and the geometry and materials are obtained from ORNL-4528 [8]. The resulting effective multiplication factor (k) and conversion ratio are both 1. Fresh fuel salt with pure U-233 is used for refueling. Since the neutron economy is extremely important for breeding in a thermal spectrum, fission products other than gaseous FP are removed online. Noble metals are removed at the same rate as gaseous FPs (1%/s). Lanthanides, Halogens, and Alkalines excluding lithium are removed at an order of magnitude lower rate (0.1%/s). The calculated refueling rate is 2.6 %/day of fuel salt (for a power density of 20 MW/MTHM) with pure U-233 as the fissile. For this study, only the salt containing the primary fissile material is considered for the source term. The breeding blanket has its own source term from actinides and from a small amount of fissioning which is not considered in this paper. The MCFR, on the other hand, is not modeled with an infinite geometry. Since fast reactor fissile concentration is related to the neutron leakage, the MCFR was modeled as a sphere with a radius of 300 cm with a 150 cm thick stainless steel reflector. The initial fuel salt mixture is NaCl-UCL₃ (54.1-45.9 mole%). For the purpose of neutronic calculations, all of the chlorine is Cl-37, and the uranium enrichment is 11%. With this design, there is a plutonium atom created or a U-238 atom fissioned for every U-235 consumed. As a result, the core remains in a critical condition with minimal compensation for buildup of FPs and actinides that diminish neutron economy, even for a hard-spectrum reactor. As a result, this model does not have any material flowing into the core save the gaseous FPs.

There are many challenges common to all MSRs when considering the fuel cycle. When the core is refueled, the refuel volume is added to the core fuel salt mixture. However, Serpent 2.1 does not account for volume changes during burnup. The way the addition of material is handled in Serpent is that the mass will increase while the volume remains the same. This causes the density to increase if not mitigated. Changing density also impacts the results of the calculations in a way that would not be physical. To compensate for this, an overflow tank (or expansion volume) is employed to maintain density. This is a volume that is within the serpent model but is not under a neutron flux, and the material in it is not reintroduced to the core fuel salt. For this study, the LFTR and single fluid, thermal MSR designs have an overflow tank within the model. It may be possible that an MSR reactor facility would have such an overflow tank. In which case, the overflow tank would have a separate source term, and the overflow fuel salt could be used to start another reactor. The management of an overflow tank is not explored in this paper. Another issue is that fuel salt oxidizes during operation. One way to manage oxidation is by adding pure uranium metal, which would have some impact on the source term result. The method for managing oxidation needs to be specific to the operational details for a particular design. The goal of this study is to make equitable comparisons for potential releases of radioisotopes among various reactor designs, and in order to accomplish this, energy produced per mass of heavy metal exposed to neutrons (e.g., GWd/MTHM) is used. For solid fueled reactors, GWd/MTHM, is a well-defined quantity since the fuel remains in the reactor during its time of exposure to neutrons. In the case of liquid fueled reactors, heavy

metal fuel spends time exposed and unexposed to neutrons. In addition, makeup fuel may be added during the time of consideration for the determination of energy produced.

The energy produced is determined by the power density multiplied by the time that volume of the critical portion of the system has produced energy, which is the most common method for expressing burnup in solid fueled LWRs. The mass of heavy metal is the amount in the critical volume under consideration plus the amount of makeup fuel during the same time under consideration. The radioisotopes produced during any particular duration of time, depending on the fuel composition and on the neutron energy spectrum. The accuracy of models that determine the impact of variations of the neutron spectrum and composition of the critical volume if dependent on computational methods and on data used in these models. For the three MSR designs considered in this study, the computational methods are the same. However, for two cases, the neutron energy spectrum is obtained based on an infinite geometry and a finite model for the other (MCFR). It is assumed that variations in the production of fission products and on transmutation are not significantly impacted by neutron leakage. The reference volumes for the infinite- and finite-media models are one liter, but the neutronic properties are averaged over the critical volume for the finite media case.

Since two reactor design models are infinite, the source terms are expressed in units of volumetric activity. Volumetric activity has the benefit of making design comparison intuitive, and accident release calculations straight forward since releases would be in terms of volume. The resulting specific activities from each of the fission product chemical groups (as in Figure1) and actinides after one full-power year of operation (power density of 20MW/MTHM) are presented in Table2. As can be seen, there are some expected behaviors, such as the low actinide activity in a LFTR design. Altogether, each reactor has a similar activity level since the majority of the source term is the result of FPs. However, there is some divergence due to the fission yield of the different fissile, initial salt chemistry, and refueling strategies. The numbers presented here are representative of the activity of currently fissioning fuel salt. It should be noted that what is shown in Table 2 is only the available radioactive inventory in the fuel salt. This does not include any of the reprocessing streams or refueling salt storage. There will be other accidents, sources and risks associated with the other inventories.

Source term (Ci/cm ³)	TSSF MSR	LFTR	MCFR
Gaseous	6.80	12.01	11.39
Semi-soluble	32.40	43.27	-
Soluble	136.10	184.5	204.47
Insoluble	49.89	19.83	56.08
Actinides	30.24	4.93*10 ⁻⁴	59.08

Table 2: The specific activity of important fission products

4.1. Burnup Calculations both with and without Gaseous Fission Product

The liquid nature of the fuel requires the online removal of gaseous FPs. Many legacy codes used to calculate reactor burnup do not have any form of online extraction or reprocessing. Reducing the

concentration of gaseous fission products after the calculation is complete is not a viable option since several decay chains have a parent or at least one daughter isotope that is gaseous. In Table 3, the specific activity of each chemical group in the case of 1%/s gaseous FP removal and without gaseous FP removal is presented. As can be seen from Table 3, gas removal can affect many different FPs beyond the gaseous chemical group. Actinides are also affected despite not in any gaseous fission product decay chain. This is mainly due to the removal of Xe-135, which has two primary effects. The first, is the change in neutron spectrum caused by Xe-135's high cross section for thermal neutrons. The second is the need for excess reactivity to compensate for xenon poisoning.

Source term (Ci/cm ³)	Gas Removal	No Gas Removal	Difference
Gaseous	6.80	20.86	+307%
Semi-soluble	32.40	32.48	-
Soluble	136.10	146.70	+8%
Insoluble	49.89	48.37	-3%
Actinides	30.24	28.74	-5%

Table 3: The change in specific activity of important fission products in an TSSF MSR when considering gaseous fission product removal

5. Source Terms for Postulated Accident Scenarios

As part of the MSRE research, various activity release scenarios were explored [17]. Results from some of these calculations for source terms are specific to the MSRE, while others (such as leakage of fuel salt following a rupture in any of the primary circuit pipes or vessels) can be scaled to most other MSRs. MSRE reports contain a wealth of information about radiation doses at different part of the reactor plant and at various locations in the surrounding environment following a breach in the primary/secondary containment. The parameters used in the dose calculation models may not be applicable for regulatory requirements to date (2020); thus, this study will only consider the activity available for dispersal following an accident by the reactor and not the destination of the radioactive material following release. It is illuminating to consider the differences in the behavior of MSRs when compared to LWRs in accident scenarios. As mentioned before, a loss- of-coolant accident as described for an LWR does not apply to MSRs. A loss-of-coolant in MSRs implies either a breach in the primary system or the secondary system. In the former case, the fuel salt would leak out into the surroundings and freeze over time depending on the rate of decay heat production. In the second case, the reduction in heat removal from the core would lead to a temperature excursion which would insert significant negative reactivity owing to the strongly negative density coefficient of reactivity for such systems. Hence, MSRs generally contain an overfill tank and associated piping to accommodate density changes in the fuel salt. In solid-fuel reactors, “meltdown” episodes (like those at Chernobyl, Fukushima, Three-Mile Island) could result in fuel pellets changing to a uncontrollable critical configuration. This is not the case with MSRs, as the fuel is already in a liquid state. However, exposure of the molten salts to water or oxygen could lead to the release of certain radioactive species. Furthermore, exposure to oxygen containing compounds could oxidize the uranium and plutonium chlorides to form oxide precipitates [17, 18]. This could lead to the formation of a critical mass

of fuel and such reactions should be thoroughly investigated. For thermal spectrum reactors, the fuel leaking out of the containment would render it in a subcritical state in the absence of moderating elements. For a fast spectrum reactor, the criticality of the fuel salt upon leakage would depend solely on the settling geometry of the liquid. Hence the design of fast reactor facilities should factor in such a sequence of events. All MSR designs are equipped with some sort of storage tanks for storing the various salts when they are not in use in any of the reactor systems. These storage tanks are often coupled with a freeze-valve based system for automatic drainage of salts into safe, non-critical configurations in the event of unforeseen accidents. This freeze-valve is simply a portion of the piping which is maintained at a lower temperature than the rest of the circuit where a mass of salt is held frozen. In the event of an excessive temperature excursion or loss of power, the freeze valves melt and the salt drain into designated storage tanks passively due to the action gravity. If a leak is detected in the system, the freeze valves can be melted to drain the salts thereby reducing the amount of radioactivity released.

As mentioned earlier, many accident scenarios that apply to LWRs does not apply to MSRs. Therefore, it is imperative to explore possible initiating events. Table 4 summarizes the accidents and their corresponding causes and consequences investigated by the authors. For this study, however, only events that result in a release of radioactive material from either the primary containment (e.g., reactor vessel and external fuel salt piping) or the secondary containment (e.g., concrete annulus or other shielding material that house the components of the primary system) are considered. Much of these events were taken from Ref. [17] with some additional scenarios that are plausible for modern designs.

Results obtained during this study can be scaled to the maximum credible scenario presented for the MSRE in the case of a leakage in the primary system and for an event where all the fuel salt is drained into the secondary containment area. Ref. [17] describes such episodes for the MSRE where (i) a spray or drip type leakage occurs in the primary system with adequate operator response, and (ii) a major rupture in the reactor vessel unloads all the fuel salt into the secondary containment area which has experienced a breach. It is important to note that some of the activity presented in this paper is the result of isotopes whose half-life is less than the ex-core transit time. In the case of a relatively long fuel salt circuit, the activity of the fuel salt leaving the core would be higher than the fuel salt entering the core. This could result in different terms when considering where a drip or rupture occurs relative to different design components in an MSR.

5.1. Spray or drip leakage in primary circuit

Although this accident scenario is considered improbable, it is estimated that the most probable type of leakage would be a spray or drip type leakage into the secondary containment area. It is assumed that the rate of fuel salt released would be only a few cubic centimeters a minute, and that this leak would be detected and stopped after no more than four liters have leaked. This will likely be different between the designs given the particulars of the reactor facility. The activities from Table 2 can be coupled with any MSR design study, however. As stated before, predicting exact chemical behavior of a molten salt reactor is beyond the scope of this study. However, some assumptions are made as to which FPs volatilize. In the previous studies [17, 14], about 10% of the FPs dissolved in the salt, 10% of the iodine, and all the gaseous FPs would volatilize and be dispersed into the cell environment. It is assumed that all insoluble FPs plate

out in the system. For this scenario, the calculated activities at the end of one year of operation for the described reactor systems are presented in Table 5.

Potential Hazard	Causes	Consequences
1. Power excursion beyond design limits	a. Uncontrolled rod withdrawal b. Premature criticality due to excess fuel addition during filling c. “Cold-slug” due to overcooling or insufficient heating of frozen salt at startup	a. Excessive temperatures causing damage to equipment b. Potentially compromised primary containment c. Dose to workers
2. Pressure in the pump/off-gas system above design limits	a. Rapid unchecked expansion of fuel salt following thermal expansion due to excess temperature (see 1.) b. Failure of off-gas system valves and pressure regulation system	a. If unchecked, damage to pump seals leading to leaks b. Exceed design capacity of off-gas system c. Possible loss of primary containment in the off-gas system d. Dose to workers if gases leak
3. Exceed overflow tank capacity	a. Overfill caused by malfunction in fill system b. Excessive temperatures leading to uncontrolled expansion in fuel salt	a. Loss of capacity to handle fuel expansion b. Unintended dynamic behavior c. Possible breach of primary containment due to excessive mechanical stress
4. Loss-of-flow/pump-trip	a. Malfunction in pump system b. Power loss	a. If secondary salts still operational, salts freeze b. Small reactivity insertion due to delayed neutron precursors remaining in core c. Minimal risk of barrier breach
5. Puncture in piping or vessel	a. Excessive corrosion of piping and/or reactor vessel b. Malfunction in auxiliary systems c. Unforeseen acts of nature d. Intentional sabotage	a. Maximum release scenario for primary containment damage b. Reactor system rendered inoperable c. Potential source release into the secondary containment area d. Possible damage to secondary containment e. Dose to workers

6. Compromised secondary containment	a. Unforeseen acts of nature b. Corrosion due to spilled salt (see 5.) c. Intentional sabotage d. Missile damage	a. Most severe accident scenario b. Reactor rendered inoperable c. Significant release of source terms possible d. Dose to workers d. Possible dose to nearby populations
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Table 4: Accident scenarios and resulting consequences

	TSSF MSR	LFTR	MCFR
Activity Released (Ci)			
Total Released	$8.22 \cdot 10^5$	$9.59 \cdot 10^5$	$1.29 \cdot 10^6$
Volatilized Isotopes	$6.74 \cdot 10^4$	$9.11 \cdot 10^4$	$1.00 \cdot 10^5$
<i>Volatilized Iodine</i>	$7.01 \cdot 10^3$	$5.48 \cdot 10^3$	$9.67 \cdot 10^3$

Table 5: The activity released from primary containment during a drip or spray based on energy production of 7300 MW-days per MTHM of fuel salt in the critical region of the core.

5.2. Major rupture of reactor vessel and breach of secondary containment

The MSRE study included another estimate for what the authors thought the worst-case scenario would be. In it, all the fuel salt would have been drained out of the core, and the secondary containment would be breached causing radioactive nuclei to be released to the environment. All the salt in the core would be released in this accident. The assumptions for volatilization are the same as in the previous scenario. If one were to assume the same heavy metal power density of 20 MW/MTHM, then each reactor design would have a different amount of fuel salt to produce the same amount of energy. Table 6, contains the amount of salt needed to yield one gigawatt of power over the year and activity released in such a sequence of events for each reactor. Note that for the LFTR design this is only the fuel salt and does not include the blanket salt, reprocessing streams or refueling stock. These parts of a reactor facility would have separate barriers of release.

	TSSF MSR	LFTR	MCFR
Volume of Salt (cm³)	$1.47 \cdot 10^8$	$1.61 \cdot 10^8$	$1.13 \cdot 10^8$
Activity Released (Ci)			
Total Released	$3.02 \cdot 10^{10}$	$3.87 \cdot 10^{10}$	$3.63 \cdot 10^{10}$
Volatilized Isotopes	$2.47 \cdot 10^9$	$3.67 \cdot 10^9$	$2.84 \cdot 10^9$

<i>Volatilized Iodine</i>	$2.58 \cdot 10^8$	$2.21 \cdot 10^8$	$2.73 \cdot 10^8$
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Table 6: The activity and amount of fuel salt released in the case of a major rupture based on energy production of 1 GW-year in the critical region of the core

6. Conclusions

Of the reactor designs evaluated in this study, the actinide source term is lowest for the LFTR, followed by the TSSF MSR. The U-233 cycle has inherently lower transuranic production, however this does not include the actinides in the blanket salt. The fast spectrum MCFR facilitates more actinide burning due to a harder neutron spectrum, but the high actinide density required to maintain criticality results in a higher production rate of actinides than the rate at which actinides are transmuted due to spectral effects. As a result, the MCFR has the highest amount of actinide activity. For large scale accidents, the LFTR had the most amount of salt available for release, which leads to it having the most substantial source term for the major rupture. This is a result of the power density being in terms of heavy metal present, which is the low density for the near-pure U-233 in the LFTR. From the calculations presented, the TSSF MSR seems to have the least amount of activity available for release after one year of operation. However, most of the source term in an actively burning fuel salt is mainly dictated by the number of fissions taking place and their related FPs. Therefore, MSRs with similar carrier salt, fissile, and power operation history should have nearly the same source term. However, a significant divergence can be introduced with the management of the overflow fuel salt. Ideally, this study would be paired with an in-depth reactor design study, a more rigorous evaluation chemical volatilization, and an accident frequency assessment. This study in combination with each of these other studies would give a complete understanding of the source term. Altogether, the predictions in this paper provide a reasonable jumping off point for source term calculations for a wide variety of MSR designs.

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Source term (Ci/cm ³)	TSSF MSR	LFTR	MCFR
Gaseous	6.93	12.01	11.39
Semi-soluble	32.55	43.27	-
Soluble	137.39	184.5	204.47
Insoluble	49.16	19.83	56.08
Actinides	29.70	4.93*10 ⁻⁴	59.08