

Methodology for Source Term Analysis of a Molten Salt Reactor

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Master of Science
Degree

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Abstract

Interest in Molten Salt Reactors ([MSRs](#)) has increased in recent years due to the changing needs and goals of nuclear energy. The Aircraft Reactor Experiment ([ARE](#)) and Molten Salt Reactor Experiment ([MSRE](#)) set the stage early on for [MSR](#) research. There are many different options for [MSRs](#) including different salt compositions, neutron spectrum, fuel choice, and purposes. Thermophysical and thermochemical properties, such as melting point, density, and viscosity must be known for the salt composition chosen for the reactor. Some neutronics differences that set [MSRs](#) apart from Light Water Reactors ([LWRs](#)) are changes in treatment of delayed neutron precursors and point reactor kinetics. [MSRs](#) allow the ability to process the fuel during operation to remove or add desired nuclides. The Nuclear Regulatory Commission ([NRC](#))’s experience is mainly focused on [LWRs](#), but they have started exploring some advanced reactor source terms from a regulatory perspective. A Molten Salt Demonstration Reactor ([MSDR](#)) and [MSRE](#) were modeled in SCALE/TRITON and when compared to a Westinghouse Pressurized Water Reactor ([PWR](#)), the results showed that [MSR](#) source terms were less than 20% compared to [PWR](#) dependent of radionuclides.

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List of Acronyms

AHR	Aqueous Homogeneous Reactor
AHTR	Advanced High Temperature Reactor
ANP	Aircraft Nuclear Propulsion
ARE	Aircraft Reactor Experiment
AST	Alternative Source Term
BWR	Boiling Water Reactor
CANDU	Canadian Deuterium Uranium
CR	Conversion Ratio
GDC	General Design Criteria
GFR	Gas-Cooled Fast Reactor
HALEU	High Assay Low Enriched Uranium
HEU	Highly Enriched Uranium
HTGR	High Temperature Gas Cooled Reactor
HTLTR	High Temperature Lattice Test Reactor
ISG	Interim Staff Guidance
LEU	Low Enriched Uranium

LFR	Lead-Cooled Fast Reactor
LMFBR	Liquid Metal Fast Breeder Reactor
LOCA	Loss of Coolant Accident
LWR	Light Water Reactor
MOSART	Molten Salt Actinide Recycler and Transmuter
MSBR	Molten Salt Breeder Reactor
MSDR	Molten Salt Demonstration Reactor
MSFR	Molten Salt Fast Reactor
MSR	Molten Salt Reactor
MSRE	Molten Salt Reactor Experiment
MST	Mechanistic Source Term
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PERMISS	Process and Effluent Radiological Monitoring Instrumentation and Sampling System
PHWR	Pressurized Heavy-Water Reactor
PRA	Probability Risk Assessment
PSA	Probabilistic Safety Assessment
PWR	Pressurized Water Reactor
R&D	Research & Development
RG	Regulatory Guide

SFR Sodium Fast Reactor

SNF Spent Nuclear Fuel

SNM Spent Nuclear Material

SRP Standard Review Plan

SWCR Supercritical-Water-Cooled Reactor

TMSR-LF Thorium Molten Salt Reactor-Liquid Fuel

TRU Transuranics

USAEC United States Atomic Energy Commission

VHTR Very-High-Temperature Reactor

Chapter 1

Introduction

There has been a resurgence of interest into Molten Salt Reactors ([MSRs](#)) in the past 20 years with more work being done in the past 10 years, and with the most research being done in the past 5 years. Recently there has been an increased focus and emphasis regarding climate change and environmental impacts. [MSRs](#) could support climate change policies by providing a larger contribution from nuclear. The push for lower environmental impacts in the energy production field could also be supported by [MSRs](#).

[MSRs](#) have the ability to be either a breeder or burner reactor, which can reduce impacts on the front end of the cycle, as well as, reduce the current inventory of Spent Nuclear Fuel ([SNF](#)) [\[1\]](#). Below are general characteristics of [MSRs](#) and other Gen IV reactor concepts, providing additional reasons for their well-deserved interest and reevaluation:

- Minimize weapons-usable material in storage
- Make beneficial use of [SNF](#) from Light Water Reactors ([LWRs](#))
- Greatly expand non-carbon based energy at a cost competitive with alternatives
- Minimize the need for high level waste repositories
- Increase resource utilization
- Increase proliferation resistance of nuclear energy [\[2\]](#).

[MSRs](#) have many safety features that are beneficial to their use; these generally are low pressure, chemically stable coolants; negative void coefficients; negative temperature coefficients; and passive shutdown capability. There are several more possible safety mechanisms including freeze valves and chemical processing. The fuel salt is a solid at temperatures below about 500°C and will become a solid in the case of many reactor accidents. This unique feature also allows for pressures near atmospheric to be used instead of the high pressures characteristic of [LWRs](#) and High Temperature Gas Cooled Reactors ([HTGRs](#)).

Figure [1.1](#) shows a break-down of different types of [MSRs](#). The first categorical division is between salt-fueled and salt-cooled. The salt-cooled designs have solid fuel with molten salt only being used as the coolant. For simplicity's sake, [MSRs](#), for the remainder of this discussion, will refer to salt-fueled unless indicated otherwise. In salt-fueled [MSRs](#), the fuel is dissolved in the molten salt creating a mixture, which typically flows from the processing area through the reactor core where fissions occur in the fuel salt with the majority of the neutrons from fission happening in the reactor core. The salt then flows into the primary heat exchanger, where the heat goes into the secondary loop, powering the turbine and producing electricity in a commercial civilian reactor. After that, a portion of the salt can go through processing to remove fission products and introduce new fissile material; the salt then starts all over again by flowing back to the reactor core. After the above divisions of categories of [MSRs](#), they can be further broken down by the neutron spectrum employed and the main salt constituent used in each reactor; these will be discussed later.

Some of the different deployed reactor concepts are shown in Table [1.1](#): Pressurized Water Reactor ([PWR](#)), [BWR](#), Canadian Deuterium Uranium ([CANDU](#)), [HTGR](#), Liquid Metal Fast Breeder Reactor ([LMFBR](#)), and Gas-Cooled Fast Reactor ([GFR](#)). In Table [1.2](#), some concepts for advanced reactors that are part of the Gen IV Initiative are shown, including [MSR](#), [GFR](#), Sodium Fast Reactor ([SFR](#)), Lead-Cooled Fast Reactor ([LFR](#)), Supercritical-Water-Cooled Reactor ([SWCR](#)) and Very-High-Temperature Reactor ([VHTR](#)).

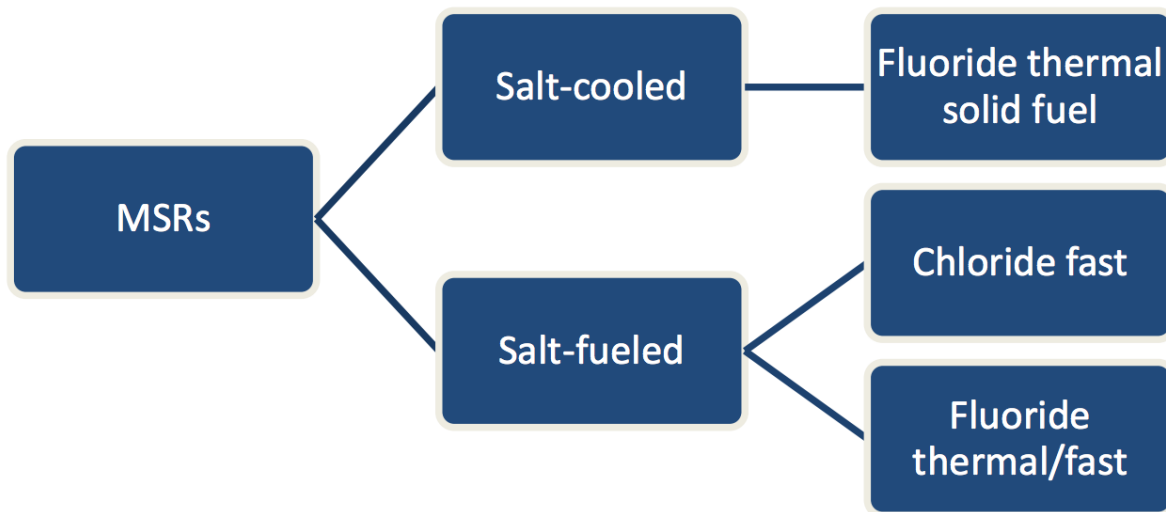


Figure 1.1: Characterization of [MSRs](#) [3]

Table 1.1: Deployed Reactor Classes and Their Properties. Information from [4]

	PWR	BWR/6	CANDU	HTGR	LMFBR	GFR
Manufacturer	Westinghouse					
Fuel	UO ₂	UO ₂	UO ₂	UC, ThO ₂	PuO ₂ , UO ₂	PuO ₂ , UO ₂
Moderator	H ₂ O	H ₂ O	D ₂ O	Graphite	None	None
Coolant	H ₂ O	H ₂ O	D ₂ O	Helium	Sodium	Helium
Thermal output (MW _{th})	3411	3579	1612	3000	2410	2530
Electrical output (MW _e)	1150	1200	500	1170	1000	1000
Burnup (MWD/MTU)	33,000	27,500	10,000	98,000	100,000	100,000
Conversion Ratio (CR)	0.5	0.5	0.45	0.7	1.3	1.5
Efficiency (%)	33.7	33.5	31.0	39.0	39.0	39.5
Fuel Weight (kg)	90,200	138,000	80,000	39,000	19,000	28,000
Specific Power (kW/kgU)	37.8	25.9	20.4	77	126	90
Fuel Enrichment	2.1/2.6/3.1	2.2-2.7	nat U	93.5	10-15	10-15
Inlet Temperature (°C)	300	269	249	337	380	332
Outlet Temperature (°C)	332	286	293	755	552	642
Operating Pressure (bar)	155	72	89	50	14	86

Table 1.2: Generation IV Concepts and Their Properties [5], [6]

Reactor Type	Fuel	Coolant	Temperature (°C)	Size (MWe)
MSR	UF, ThF, SNF	Molten Salts	700-800	1000-1500
GFR	^{238}U	Helium	850	1200
SFR	^{238}U , MOX	Sodium	550	30-150, 300-1500, 1000-2000
LFR	^{238}U	Lead or Lead-Bismuth	480-800	20-180, 300-1200, 600-1000
SWCR	UO_2	Water	510-625	300-700, 1000-1500
VHTR	UO_2 prism or pebbles	Helium	900-1000	250-300

1.1 Characteristics

MSRs have several advantages in relation to other reactors in terms of efficiency and, most importantly, safety. Some of the advantages of MSRs are

- Enables high temperature at low pressure
- Online chemistry adjustment
- Potential for inherent safety depending on design options
 - Fuel salt thermal expansion provides negative reactivity insertion
 - Fuel draining under thermal excursions
 - Low excess reactivity- fuel normally in most reactive configuration
- Potential to substantially reduce actinide waste production
- Can be refueled as "infinite batch" reactors resulting in maximum possible burnup [7].

According to Holcomb in [8], molten salts are thermodynamically stable, have a high solubility towards uranium, plutonium, and thorium, and also do not chemically react

exothermically with air or water. In addition to all these, they also have excellent heat transfer due to a large heat capacity and natural and forced circulation and are also compatible with chemical processing and nickel-based alloys and graphite [8].

These advantages make molten salt a fantastic candidate for high temperature purposes and for design of safer reactor concepts. The molten salts themselves are stable in radiation fields and can dissolve large concentrations of fission products and actinides [9]. The three main advantages of [MSRs](#) typically are passive cooling and thermal margin in the event of an accident, fuel cycle versatility, and online reprocessing system; these advantages also work together to provide a good economic outlook for [MSRs](#). Although, not all [MSR](#) designs plan to employ reprocessing, or plan to employ varying levels of reprocessing instead of reprocessing everything. Generally one of the main aspects to [MSRs](#) inherent safety features is its simple and passive cooling system during an emergency. The typical [MSR](#) cooling system does not need any power; in fact, it is the lack of power that triggers the [MSR](#) cooling system. In the Molten Salt Breeder Reactor ([MSBR](#)) design, there was a solid plug of salt at the bottom of the reactor vessel that was constantly cooled to below freezing by an electrical fan that would shut off if power is lost; this is still a typical feature in most modern designs. If a power loss to the fan occurs, the salt plug will melt and all of the fuel from the reactor will be gravitationally pulled down into a protected tank that is underground [2], [9], [10], [11]. This safety feature helps prevent a release of radioactivity into the environment and also prevents system damage. Another safety feature that plays on the high melting point of the salt happens when there is a leakage of the salt from the vessel or piping; if the leak is large enough to cause concern, the fuel will freeze quickly halting operations of the reactor [10]. [MSRs](#) have a lot of versatility in the fuel cycle, as well as good fuel utilization and flexibility. With the fuel cycle versatility, there are several fuel options for [MSRs](#), including thorium, plutonium, and uranium, that can be operated as a breeder, burner, or self-sustaining reactor to meet the current goals and adapt to the most cost effective fuel at the time [11].

1.2 Comparison with LWRs

Most of today's regulatory and licensing experience is with LWRs, and thus it is important to point out the differences and compare MSRs to them. MSR and LWR licensing differences will be discussed in Chapter 7, but some of the main differences between the two that cause issues are:

- Nuclear reactivity
- Fissile inventory
- Burnup and Pu isotopes
- Delayed neutron characteristics
- Homogeneous fuel composition [9].

Some other differences in how they operate include the MSR running at atmospheric pressure and a temperature range of 500-1400°C, while LWRs have a high pressure of 150 atm operating at around 315°C. Temperatures up to 1400°C are the absolute maximum temperature for MSRs since it is the boiling point of many of the salts, but material limits will almost always be reached long before then. This results in better thermodynamic efficiency for MSRs due to the ability to maintain low vapor pressure at high temperature. For an MSR with full reprocessing, the core reactivity does not change considerably with time, since the fuel is added continuously and reprocessed continuously to separate radionuclides. Even in an MSR with minimal separations and reprocessing, xenon and krypton continuously escape via the off-gas system [9]. Reprocessing will be discussed in more detail in Chapter 6. Due to these features, the inventory of fissile radioisotopes is low for MSR compared to solid fuel reactors, especially when more reprocessing is utilized. A couple of other reasons for its low fissile inventory include no conventional SNF outside of the reactor and less need to compensate for burnup meaning only a small amount of excess reactivity [9]. Since there is no need for a secondary containment for steam capture and also no pressurized primary-containment, safety features can be less complex and the plant size can be smaller than an LWR [9].

The fraction of delayed neutrons is an important aspect and must be accounted for in all types of reactors to maintain reactor control. For [LWRs](#), all the delayed neutrons occur inside the core, but for [MSRs](#), since the fuel is flowing into and out of the core, some delayed neutrons will occur outside the core. The amount of prompt and delayed neutrons inside the core is important for all reactors, but for [MSRs](#) another aspect for fission neutrons is the fraction outside of the reactor core [9]. For reactors with solid oxide fuel, the fuel must never reach a molten state to ensure safety; whereas to ensure safety for [MSRs](#), the solutions must be in a molten state to keep the FPs and actinides in solution [2].

1.3 Source Term

The source term, as defined by the Nuclear Regulatory Commission ([NRC](#)), is the types and amounts of radioactive or hazardous material released to the environment following an accident [12]. The Health Physics Society defines source term as "the types, quantities, and chemical forms of the radionuclides that encompass the source of potential for exposure to radioactivity." [13]. A mechanistic source term attempts to realistically model the release and transport of radionuclides from the source to the environment for a specific scenario [14]. For [MSRs](#), the mechanistic source term will be more representative since it takes the retention phenomena into account, as well as other safety features not present in a [PWR](#). There is a large investment of both money and time associated with the current licensing process. There needs to be a technology-inclusive process with emphasis on communication throughout the licensing process in order to be more flexible and understand what the advanced reactor community needs to improve regulation on non-LWRs.

The source term is the inventory of radioisotopes in the reactor available for dispersion, and the contribution is two-fold [15]. The source term is both the measure of the radiation that needs to be contained and the measure of the after heat [15]. The energy from the after heat following a heat-removal failure or Loss of Coolant Accident ([LOCA](#)) provides the driving force for the dispersion of the source term [15]. The driving force for dispersion is reduced and there are no major stored energy sources within containment such as high pressure or

reactive fluids [2], [15].

Even with the least amount of fuel processing employed, the volatile gases will be removed, thus removing this from the source term. The gaseous and volatile part is the most likely to be dispersed when there is a breach of containment [15]. The inventory of long-lived isotopes can be reduced by fuel processing, thus reducing the source term since their accumulation is time dependent. These parts of the source term would be unavailable in the event of an accident since they are removed.

Chapter 2

History

2.1 Aircraft Reactor Experiment

In the late 1940s, investigation of [MSRs](#) started as part of the United States' program to develop a nuclear powered airplane [16]. As part of the Aircraft Nuclear Propulsion ([ANP](#)) program at Oak Ridge National Laboratory ([ORNL](#)), molten salts were used for the plane's propulsion system. Fluorides appeared particularly appropriate because they have high solubility for uranium, are among the most stable of chemical compounds, have very low vapor pressure even at high heat, have reasonably good heat transfer properties, are not damaged by radiation, do not react violently with air or water, and are inert to some common structural metals [16]. The Aircraft Reactor Experiment ([ARE](#)) was built in the 1950s to study a circulating fuel system's nuclear stability and investigate molten fluorides as fuels for aircraft propulsion reactors. The [ARE](#)'s properties are listed in Table 2.1. In 1954, the [ARE](#) was operated successfully for 100 hours at steady-state outlet temperatures ranging up to 860°C and at powers up to 2.5MW_{th} [16]. During the operation, it was both stable and self-regulating and there were no chemical or mechanical problems found.

Table 2.1: Main Properties of ARE [17]

Property	Makeup
Fuel Salt	Mixture of NaF, ZrF ₄ , and UF ₄
Uranium Enrichment	93.4% ²³⁵ U
Moderator	BeO
All Piping	Inconel

2.2 Molten Salt Reactor Experiment

Also at ORNL was the Molten Salt Reactor Experiment (MSRE), which ran from 1960-1969. The timeline for the MSRE can be seen in Table 2.2. The MSRE was an 8 MW_{th} reactor with a single core region made up of 33% U-235 fuel [18]. It had an outlet temperature of about 700°C and it had Alloy N vessel and piping. It had several achievements including first reactor to use U-233 fuel, first use of mixed U/Pu salt fuel, online refueling, and over 13,000 full power hours. A single-fluid design was chosen since it resembles a converter in its engineering features, but the fuel salt was similar to what would be used for a two-fluid breeder since it did not contain uranium. Some properties of the MSRE can be seen in Table 2.3.

A Schematic of the MSRE reactor can be seen in Figure 2.1. For the MSRE, graphite blocks were used in the reactor core for neutron moderation [19]. The fuel salt circulated throughout an external pump into the reactor core then through the salt-salt heat exchanger. The secondary coolant was a similar salt but without the actinides seen in the fuel salt. The coolant loop circulated the salt through a radiator in order to dissipate heat to the atmosphere. Xenon and krypton were removed by gas sparging, a technique used to remove volatile components or dissolved gases by bubbling a gas through a liquid, via a pump bowl [18]. In high radiation conditions and at high temperatures, the salt chemistry was concluded to behave well. The reactor’s dynamic behavior correlated well with predictions as reported by [20]. The successful MSRE operation helped show that using molten salt for reactors is

Table 2.2: MSRE Demonstrations of Key MSRE Technology [18], [21]

Date	Event
Oct. 1964	Salt First Loaded into Tanks
Jan 1965	Salt First Circulated through Core
June 1965	First Criticality
May 1966	Briefly Reached Full Power
Dec. 1966	First Full Power Operation on U-235
Oct. 1968	First Critical with U-233 Fuel
Dec. 1969	Shutdown

Table 2.3: Main Properties of MSRE [18]

Property	Makeup
Fuel Salt	LiF-BeF ₂ -ZrF ₄ -UF ₄ (65-29.1-5-0.9mol%)
Lithium Enrichment	99.99% ⁷ Li
Uranium Enrichment	33% ²³⁵ U
Moderator	Unclad Graphite
All Piping	Hastelloy-N
Cover Gas	Helium

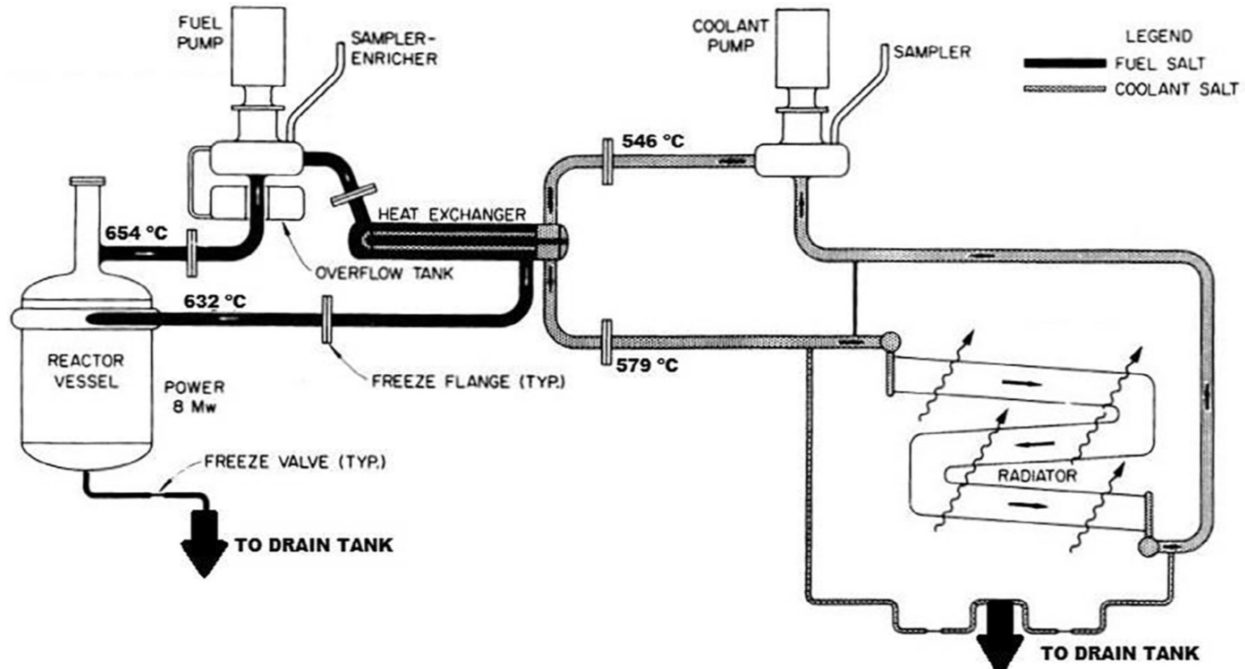


Figure 2.1: MSRE schematic [20]

practical, corrosion of graphite and metal is very low, and, under reactor conditions, the salt is stable.

2.3 Source Term History

2.3.1 TID-14844

The United States Atomic Energy Commission (USAEC) released a technical information document titled "Calculation of Distance Factors for Power and Test Reactor Sites", known as TID-14844, in 1962 [22]. The deterministic methodology for calculating radiological hazards and corresponding site boundary distances at commercial nuclear power plants was outlined in this report. This source term in this document was based on deterministic assumptions for a maximum credible accident in an LWR. This maximum credible accident was defined as substantial core melt due to a LOCA. RGs 1.3 and 1.4 have incorporated these assumptions for the evaluation of radiological consequences at power reactors. It included guidance on assumed fractional release to containment, atmospheric transport and

Table 2.4: Assumed release percentage according to TID-14844 [14], [22]

Nuclide Group	Assumed percentage released to containment
Noble gases	100%
Halogens	50%
Remaining solids	1%

dispersion behavior, and calculation of offsite consequences [14]. There was only explicit guidance for LWR and conservative assumptions and parameters were utilized in the absence of appropriate data. These assumptions included instantaneous release to containment, no credit to engineered safety features, and conservative assumed release fractions. These release fractions can be seen in Table 2.4. RGs 1.3 and 1.4 derived their assumptions from TID-14844.

2.3.2 WASH-1400

In 1974, the NRC released WASH-1400, which provided the first mechanistic treatment for accident progression and used more realistic bases and assumptions than TID-14844 [23]. It established 7 BWR and 9 PWR classes of fission product release that were based on probability of accident progression and mechanistic accident [23]. The Probabilistic Safety Assessment (PSA) was based on fault tree and event tree analysis.

2.3.3 NUREG-0956

NUREG-0956 released in 1986 provided reassessment of the source terms for severe accidents and introduced the NRC source term code package which included ORIGEN, MARCH, CORSOR, MERGE, and others [24]. It also provided major improvements in modelling of fission products by including fission product chemistry and the interaction of fission product behavior and thermal hydraulics.

2.3.4 NUREG-1465

In 1995, the NRC released a new document relating to source term named "Accident Source Terms for Light Water Reactor Plants," referred to as NUREG-1465 [25]. There are unique

PWR and BWR source terms based on a range of accident scenarios derived from NUREG-1150 [14], [26]. NUREG-1465 provided a realistic assessment of source term including uncertainties instead of the deterministic bounding value in TID-14844, as well as it gave credit to engineered safety features for fission product removal. A large break LOCA was chosen since it results in the earliest onset of radionuclide release to the fuel-clad gap [14]. The releases in this document were not necessarily bounding, like in TID-14844, and were intended to represent typical releases from a core melt event at low pressure [14]. The release fractions used in NUREG-1465 can be seen in Tables 2.5 and 2.6.

2.3.5 Regulatory Guide 1.183

RG 1.183 named "Alternative Radiological Source Terms for Evaluating Design Basis Accidents at Nuclear Power Reactors" was released by the NRC in 2000 [27]. RG 1.183 contains formal guidance for developing acceptable source term and offsite consequence and siting analyses. It is intended to only be used in conjunction with an acceptable Alternative Source Term (AST). A large break LOCA is used as the representative accident in this document. It also gives a quantitative assessment of the fractional fission product composition in the gap, which only applies to non-LOCA scenarios [14]. The release fractions are similar to NUREG-1465 except only the gap release and early in-vessel phases are considered; this is due to the design basis source term criteria in 10 CFR 50.67, which considers the total effective dose to only be over the first two hours of the accident since those are the worst [14], [28]. The release fractions match those of NUREG-1465, but the onset of each phase is explicitly defined in RG 1.183. The updated release fractions can be seen in Tables 2.7 and 2.8.

2.3.6 NRC's Perspective on Mechanistic Source Term

In the early 1990s, the NRC began addressing the use of Mechanistic Source Term (MST) in advanced reactor licensing by issuing SECY-93-092 [29]. NRC defined the MST as "the result of an analysis of fission product release based on the amount of core damage, fuel damage, and cladding damage resulting from the specific accident sequences" [29].

Table 2.5: BWR Fractional Release Assumptions in NUREG-1465 [14], [25]

Nuclide Group	Gap Release	Early In-Vessel	Ex-Vessel	Late In-Vessel
Noble gases	0.05	0.95	0	0
Halogens	0.05	0.25	0.30	0.01
Alkali metals	0.05	0.20	0.35	0.01
Tellurium group	0	0.05	0.25	0.005
Barium, Strontium	0	0.02	0.1	0
Noble metals	0	0.0025	0.0025	0
Cerium group	0	0.0005	0.005	0
Lanthanides	0	0.0002	0.005	0

Table 2.6: PWR Fractional Release Assumptions in NUREG-1465 [14], [25]

Nuclide Group	Gap Release	Early In-Vessel	Ex-Vessel	Late In-Vessel
Noble gases	0.05	0.95	0	0
Halogens	0.05	0.35	0.25	0.1
Alkali metals	0.05	0.25	0.35	0.1
Tellurium group	0	0.05	0.25	0.005
Barium, Strontium	0	0.02	0.1	0
Noble metals	0	0.0025	0.0025	0
Cerium group	0	0.0005	0.005	0
Lanthanides	0	0.0002	0.005	0

Table 2.7: BWR Fractional Release Assumptions in RG 1.183 [14], [25]

Nuclide Group	Gap Release	Early In-Vessel
Noble gases	0.05	0.95
Halogens	0.05	0.25
Alkali metals	0.05	0.20
Tellurium group	0	0.05
Barium, Strontium	0	0.02
Noble metals	0	0.0025
Cerium group	0	0.0005
Lanthanides	0	0.0002

Table 2.8: PWR Fractional Release Assumptions in RG 1.183 [14], [25]

Nuclide Group	Gap Release	Early In-Vessel
Noble gases	0.05	0.95
Halogens	0.05	0.35
Alkali metals	0.05	0.25
Tellurium group	0	0.05
Barium, Strontium	0	0.02
Noble metals	0	0.0025
Cerium group	0	0.0005
Lanthanides	0	0.0002

MST take into account mitigation features, whereas earlier source terms had not. It reviewed the source term proposed by the vendor for the advanced reactors in the preapplication phase [14]. Only one of these advanced reactor designs proposed the use of MST out of the four under consideration [29]. It recommended that advanced reactor source terms should be based on mechanistic analysis and provided the following three items that must be met:

- "The performance of the reactor and fuel under normal and off-normal conditions is sufficiently well understood to permit a mechanistic analysis. Sufficient data should exist on the reactor and fuel performance through research, development, and testing programs to provide adequate confidence in the mechanistic approach.
- The transport of fission products can be adequately modeled for all barriers and pathways to the environs, including specific consideration of containment design. The calculations should be as realistic as possible so that the values and limitations of any mechanism or barrier are not obscured.
- The events considered in the analyses to develop the set of source terms for each design are selected to bound severe accidents and design dependent uncertainties" [29].

In 2003, the NRC released SECY-03-0047, which recommended using MSTs, specifically for licensing decisions related to the containment and siting [14], [30]. They recognized that unique reactor design features that could affect the source term would be considered using MST instead of a deterministic, bounding source term.

Chapter 3

Salt Composition

At this time, most research is not focused on one specific [MSR](#) technology, but rather developing a generic method that can be used for different salt compositions and materials. The potential source term for an [MSR](#) includes fission products, their decay daughters, activation products, and transmutation products, in addition to the initial fissile material and salt composition. In a [PWR](#), [BWR](#), and most other reactor types of the present, the source term is contained within the core, whereas with an [MSR](#), the source term can be anywhere in the primary system including within the salt, in a processing system, absorbed or adhered to reactor materials, or lost to the containment area. In order to solve the challenge of both accounting for and tracking the source term, the chemistry of the salt needs to be well understood. Salt chemistry behavior is related to the following:

- Thermophysical properties of salts and their dependencies on operating conditions and compositional changes
- Thermochemical nature of salts, including their phase relationships dependent on operating conditions and salt composition changes
- Impact of salt composition and operating conditions on corrosion of primary structural materials [\[31\]](#).

3.1 Fluorides vs. Chlorides vs. Nitrates

Fluorides have low vapor pressure, high solubility for uranium, reasonably good heat transfer properties, excellent chemical stability, radiation resistance, and good compatibility with air, water, and some common structural materials [32]. Fluoride has the least corrosion issues out of all the alkali-halide carrier salts due to its extreme electro-negativity. Chloride salts are also a good candidate, but there is less data and experiments available for them. Also, chloride salts enable a harder neutron spectrum and have enhanced breeding [33]. ^{35}Cl has a moderate capture cross-section, making isotopically separated chlorine preferable [33]. ^{35}Cl produces ^{36}Cl in absorption reactions, which is a long-lived radionuclide. Some chloride salts have a higher plutonium solubility than fluoride salts, as well as a lower melting point. There has been no in-core testing of chloride salts, whereas fluoride salts have multiple in-pile loops, capsule tests, and two operating MSRs. Both chloride and fluoride salts are used as heat transfer fluids since they have high heat capacity and a high boiling point. They have low thermal conductivity, as well as being insensitive to fission products. Both types of salts, under mildly reducing conditions, are reasonably compatible with graphite and high temperature structural alloys [33]. Chlorides have more corrosion concerns than fluorides based salts. Nitrates have been used in Aqueous Homogeneous Reactor (AHR), but are not being heavily considered for the current MSR designs [34]. Some properties of a few typical salt composition can be seen in Table 3.1. As can be seen, the nitrate salt has a much lower boiling point, which in the case of MSRs is typically undesirable.

3.2 Stoichiometry

According to Grimes, fuel-containing salts are required to have freezing points below 525°C to be considered useful [40]. However, this is not necessarily true in some current cases where other properties are ideal, but the melting point is higher than 525°C . Using a salt with the lowest freezing point possible is of great interest in order to simplify the system. To achieve this, salts with multiple components will be necessary since there are no single component

Table 3.1: Properties for Typical Fluoride, Typical Chloride, and Typical Nitrate Molten Salts [35]. $\text{NaNO}_3\text{-NaNO}_2\text{-KNO}_3$ is at 400°C .

Property	LiF-NaF-KF	KCl-MgCl ₂	$\text{NaNO}_3\text{-NaNO}_2\text{-KNO}_3$
Composition (mol %)	46.5-11.5-42	67-33	7-49-44
Melting Point ($^\circ\text{C}$)	454 [36]	435 [36]	142 [36]
Boiling Point ($^\circ\text{C}$)	1570 [37]	>1418 [38]	Decomposition at 538 [39]
Specific Heat Capacity ($\text{J/g/}^\circ\text{C}$)	1.88 [36]	1.15 [36]	1.56 [36]
Thermal Conductivity (W/m-K)	0.92 [37]	0.445 [35]	0.51-0.605 [36]
Density (g/cm^3) at 600°C	2.02 [37]	1.60 [35]	1.84 [35]
Viscosity (cP) at 700°C	2.9 [37]	1.4 [38]	1.2 [38]

salts that freeze at a low enough temperature; adding 1 additional component to the pure component to form a binary mixture can lower the freezing point by as much as 500°C , but adding an additional component in order to have a ternary mixture does not significantly effect the freezing point and is normally added for other reasons [41].

Phase diagrams are useful to help determine the relevant temperatures and best compositions. The phase diagrams for LiF-BeF₂ and LiF-BeF₂-UF₄ are given in Figures 3.1 and 3.2; phase diagrams for other salts of interest can be found in Appendix A.

It can be seen in Figure 3.1 that the binary system LiF-BeF₂ has one eutectic, which is where a liquid cools directly into a solid 2-phase region, at 355°C with a composition of 52 mole % BeF₂. There is also one peritectic, which is where a solid phase and liquid phase will form a second solid phase with no liquid, at 454°C with a composition of 33.5 mole % BeF₂. According to several sources, 33 mole % BeF₂ is the most suitable for nuclear applications due to its favorable properties at that composition [42], [43], [44], [45]. If this composition is used, the melting point of the salt will be around 454°C .

The ternary phase diagram for LiF-BeF₂-UF₄ is given in Figure 3.2. It can be seen that there are 2 eutectics; one at 426°C with a composition of 69-23-8 mole % and one at 350°C with a composition of 48-51.5-0.5 mole %. The design for MSBR used LiF-BeF₂-UF₄ as the

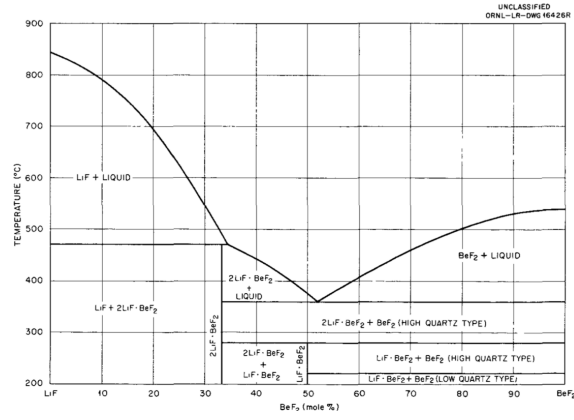


Figure 3.1: Phase diagram of LiF-BeF₂ [46], [47]

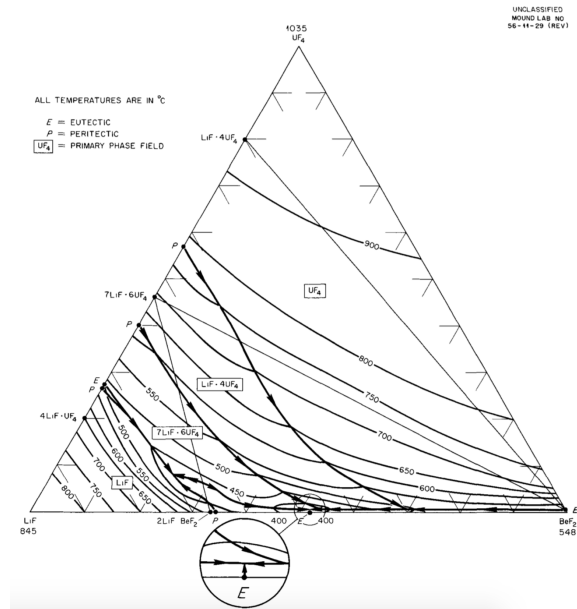


Figure 3.2: Phase diagram of LiF-BeF₂-UF₄ [46], [48]

fuel salt; the composition proposed was 68.5-31.3-0.2 mol % and has a melting point of 450°C.

The melting points of LiF and BeF₂ are 845°C and 555°C respectively. By adding them together into a binary mixture the melting point decreases about 400°C for lithium and about 100°C for beryllium to 454°C, whereas adding UF₄ to LiF-BeF₂ only decreases the melting point by 4°C to 450°C.

3.3 Neutron Spectrum

A range of neutron energies can be used in nuclear reactors ranging from thermal to fast neutron energies. In a thermal spectrum [MSR](#), neutrons with energy equal to the average kinetic energy of the other surrounding particles are used to maintain the chain reaction. In order to do this, fast neutrons must be slowed down to thermal energies, which is achieved using a moderator. [LWRs](#) typically use light water as the moderator, however this is not a good option for [MSRs](#). Graphite is typically used as a moderator in an [MSR](#) with the fuel salt moving around them versus in [LWRs](#), where the moderator surrounds the fuel rods. Thermal spectrum [MSRs](#) typically have a lower amount of fissile material needed for start-up. A thermal neutron spectrum looks more promising for burner designs since the use of a moderator and the lower initial fissile material makes breeding more difficult.

Meanwhile, a fast reactor uses fast neutrons to maintain the chain reaction. Since no moderator is required to slow down the neutrons, fast [MSRs](#) typically have a simpler core design. Typically for a fast spectrum reactor, a higher fissile load is required, losses to fission products are low, and there is a shorter prompt neutron lifetime, whereas for a thermal spectrum, losses to fission products are high, a very low fissile inventory is needed, and there is an increased prompt neutron lifetime. A fast neutron spectrum usually increases the breeding ratio. The salt will need to contain a higher concentration of fissile material in order to breed. Currently, fast spectrum [MSRs](#) will need more Research & Development ([R&D](#)) than thermal spectrum [MSRs](#) since there is a lot of uncertainty surrounding the necessary

technologies.

3.4 Fuel Choice

There are several fuel cycle options available including those using uranium, plutonium, thorium, and SNF. Using Low Enriched Uranium (LEU) enriched to less than 5% as fuel will have fewer challenges from the NRC since there is no thorium and the enrichment can be lower [49]. A lot of experience has been accumulated for LEU as reactor fuel since it is used in most LWRs. Low fuel enrichment and decent Conversion Ratio (CR) are attractive, while maintaining the advantages of the fluid fuel such as low pressure and high temperature operation, and passive safety inherent to fluid core design [49]. The CR is the ratio of number of fissile nuclei produced to the number of fissile nuclei consumed [50]. Eventually, a SNF recycler or efficient thorium breeder reactor could be possible, but the focus should be on LEU first since the NRC has dealt with LEU as reactor fuel more than SNF or thorium. Highly Enriched Uranium (HEU) was the fuel of choice for the MSRE with an enrichment of 33%, but it is not as favored now due to concerns involving economics, material availability, and safeguards. High Assay Low Enriched Uranium (HALEU), which is 5-20% U-235 content, is being investigated as a possible fuel as well [51].

Thorium has several advantages over uranium, such as a greater abundance of natural thorium, lower capture-to-fission ratio, reduced plutonium and actinide production, and potentially better non-proliferation as well [52]. U-233 is bred from thorium-232 in the thorium fuel cycle and can be bred from Th-232 in any neutron spectrum [7], [53]. Thorium has been used as fuel at both Shippingport and Fort St. Vrain, so it is not completely uncharted territory.

Designs using uranium fuel, thorium fuel, and Transuranics (TRU) fuel can be seen in Table 3.2 along with their neutron spectrum, thermal power, thermal efficiency, and operating temperature.

Table 3.2: Comparison of Different [MSR](#) Designs [\[54\]](#).

MSR Design	Fuel Cycle	Neutron Spectrum	Operating Temperature (°C)	Thermal Power (MW _{th})
MOSART	TRU -Th- ²³³ U [55]	Fast [55]	600-720 [55]	2400 [55]
Flibe Energy	Th- ²³³ U [56]	Thermal [56]	653 [56]	600 [56]
Terrestrial Energy	LEU [57]	Thermal [57]	600 [57]	400 [57]

3.5 Purpose (Breeder vs. Burner)

There are two main types of fuel strategies: breeder and burner. Some of the specifications of different breeder and burner reactors can be seen in [Table 3.3](#). In a breeder reactor, more fissile material than is needed to continue the chain reaction is generated resulting in production of its own fuel. Neutron economy is important and the parasitic neutron absorption must be low for a breeder reactor [\[58\]](#). Usually fuel processing is employed to remove fission products with high neutron absorption cross sections. Breeders tend to be fast neutron spectrum and are also more likely to use thorium as fuel.

There is no excess fissile material generated in a burner reactor so it cannot sustain operation on its own for an indefinite amount of time. A burner normally has a simplified core design and it does not necessarily have fuel processing, although it could. Neutron economy is not important because absorption by the fission products is not likely to hurt the reactor since it is not expected to breed. A burner is more likely to use uranium as fuel rather than thorium and can potentially be used to destroy actinides in [SNF](#). One big advantage for burners versus breeders is the lower amount of [R&D](#) needed.

3.6 Salt Composition Requirements

There are several requirements for the fuel salt components; according to [\[59\]](#), these are:

- Must be able to dissolve more than the critical concentration of fissionable material at operating temperatures

Table 3.3: Design Variants for MSR with Fluoride Salts [58]

Concept	Neutron Spectrum	Fuel	Use	Fissile Separations
Terrestrial Energy	Thermal	LEU	Burner	Offsite
Thorcon	Thermal	LEU & Thorium	Burner	Offsite
Flibe Energy	Thermal	Thorium	Breeder	Onsite
Indian MSBR	Thermal	Thorium	Breeder	Onsite
Chinese TMSR-LF	Thermal	Thorium	Breeder	Mixed Onsite and Offsite
MSRE	Thermal	HEU	Burner	Onsite
MOSART	Fast	TRU/natural U	Breeder	Onsite
MSFR	Fast	Thorium	Breeder	Onsite

- Have a low neutron capture cross section for the chosen energy spectrum
- Must be thermally stable
- Have low vapor pressure for the operating temperature range
- Must possess heat transfer and hydrodynamic properties adequate for heat exchanger fluid, but with minimal power needed for circulation
- Relatively non-corrosive
- Must be stable in radiation environment of the reactor
- Must tolerate fission product accumulation without losing its useful properties.

The following information was gathered for fluoride systems largely from two papers by Williams [41], [59]. The molten salts commonly used are chloride- or fluoride-based, mixed with beryllium, sodium, potassium, zirconium, or lithium. Some fluoride compounds and compositions along with their melting points can be seen in Tables 3.4 and 3.5. For fluoride systems, the first process of elimination for possible salt components is their neutronics

Table 3.4: Different Fluoride Salt Components [60]

Salt Component	Melting Point (°C)
${}^7\text{LiF}$	845
NaF	995
KF	856
RbF	775
BeF_2	555
ZrF_4	903

Table 3.5: Different Fluoride Salt Compositions [41], [60]

Salt Composition	Molar Composition (%)	Melting Point (°C)
LiF- BeF_2	67-33	458
NaF- BeF_2	57-43	340
LiF- ZrF_4	51-49	509
RbF- ZrF_4	58-42	410
KF- ZrF_4	58-42	390
NaF- ZrF_4	59.5-40.5	500
LiF-NaF- BeF_2	31-31-38	315
LiF-NaF-KF	46.5-11.5-42	454
LiF-NaF- ZrF_4	42-29-29	460
LiF-NaF- ZrF_4	26-37-37	436
NaF-RbF- ZrF_4	33-24-42	420
KF-KBF ₄	25-75	460
RbF-RbBF ₄	31-69	442
NaF-NaBF ₄	8-92	384

including their moderating properties and parasitic neutron absorption in combination with the limits associated with the reactor's mission and the neutron energies in the reactor [59]. Salts containing enriched lithium will certainly have better neutronic performance than their analogs containing Na, Rb, or K, but at a significant extra expense associated with isotopic enrichment since the lithium used must be almost isotopically pure Li-7 since Li-6 has a high neutron absorption cross section [41], [61]. Some other salt compositions were eliminated due to a variety of reasons: BeF₂ rich systems are very viscous due to BeF₂'s behavior in the system, making them undesirable candidates, salts containing potassium have a high parasitic capture cross section usually eliminating them, and rubidium has epithermal absorption bands knocking it out as an ideal candidate [41].

Summing up these issues with specific fluoride salt components:

- ⁷Li- high initial capital cost of salt
- Lithium and Beryllium- tritium production
- Rubidium- industry start-up expenses [49].

Chapter 4

Thermochemical and Thermophysical Properties

In order to operate an [MSR](#), thermophysical properties need to be known for relevant salt compositions at temperatures from 500-1000°C. These properties can be estimated through first principle predictions and extrapolation and interpolation of measured data with appropriate data and models, but the data sets and correlations found in the literature have a lot of discrepancies and gaps in the available information.

Historically, many conceptual designs used a neutron balance table to demonstrate the neutron behavior at equilibrium [\[62\]](#). The generic version of this equation is listed below.

$$\begin{aligned} \text{Rate of Energy Accumulation} = \\ \text{Rate of Energy Entering the System} - \text{Rate of Energy Leaving the System} \end{aligned} \tag{4.1}$$

Table [4.1](#) shows how well the prediction method works and how well experimental measurements work for various thermodynamic properties. The prediction methods in this table consist of extrapolation or interpolation of known data and the measurement column refers to experimental values of the properties. For viscosity, the measurement is accurate for molten salts at low temperatures, but it is not accurate at higher temperatures and in material that is highly corrosive [\[63\]](#).

Table 4.1: Prediction Methods for Properties of Fluoride Salts [41]

Property	Prediction Method	Measurement
Density	Accurate	Accurate
Heat Capacity	Approximate	Database is deficient
Melting Point	Difficult to Predict	Accurate
Vapor Pressure	Difficult to Predict	Accurate
Viscosity	Difficult to Predict	Accurate at low temperatures
Thermal Conductivity	Difficult to Predict	Poor Database, Difficult to Measure

4.1 Use of Properties in Reactor Design

A diagram of the Molten Salt Demonstration Reactor ([MSDR](#)) design is shown in [Figure 4.1](#). Thermophysical properties need to be known for many different reasons: for example, thermal conductivity is needed for heat exchanger design, and viscosity affects the speed of the salt flow as well as its resistance to flow. Density is important to determine heat transfer capabilities. Melting and boiling point are important to the liquid fuel flow by insuring the salt stays a liquid during operation. Viscosity, density, and thermal conductivity are all important for heat exchanger performance. Values for some of these properties for [MSRE](#) and [MSBR](#) can be seen in [Table 4.2](#).

4.2 Melting Point and Heat of Fusion at Boiling Point

In order to simplify the system, using the salt with the lowest freezing point possible is of interest. As can be seen in [Table 4.3](#) for alkali fluorides, the melting point depressions for ternary mixtures are very modest compared to the binary systems with a change of about 40°C. There is a 25°C depression for LiF-NaF-BeF₂ compared to NaF-BeF₂, but this is not a significant advantage and the addition of LiF is costly. LiF-NaF-ZrF₄ has a melting point less than 500°C and a Na:Zr ratio equal to 1, making it a possible candidate. NaF-RbF-ZrF₄ ternary system also has some low melting point compositions. Phase diagrams can be found in [Section 3.2](#) and [Appendix A](#), and melting points of some useful salt compositions can be found in [Table 4.3](#). Also, the melting points as determined by Thoma in [64] for [MSRE](#)

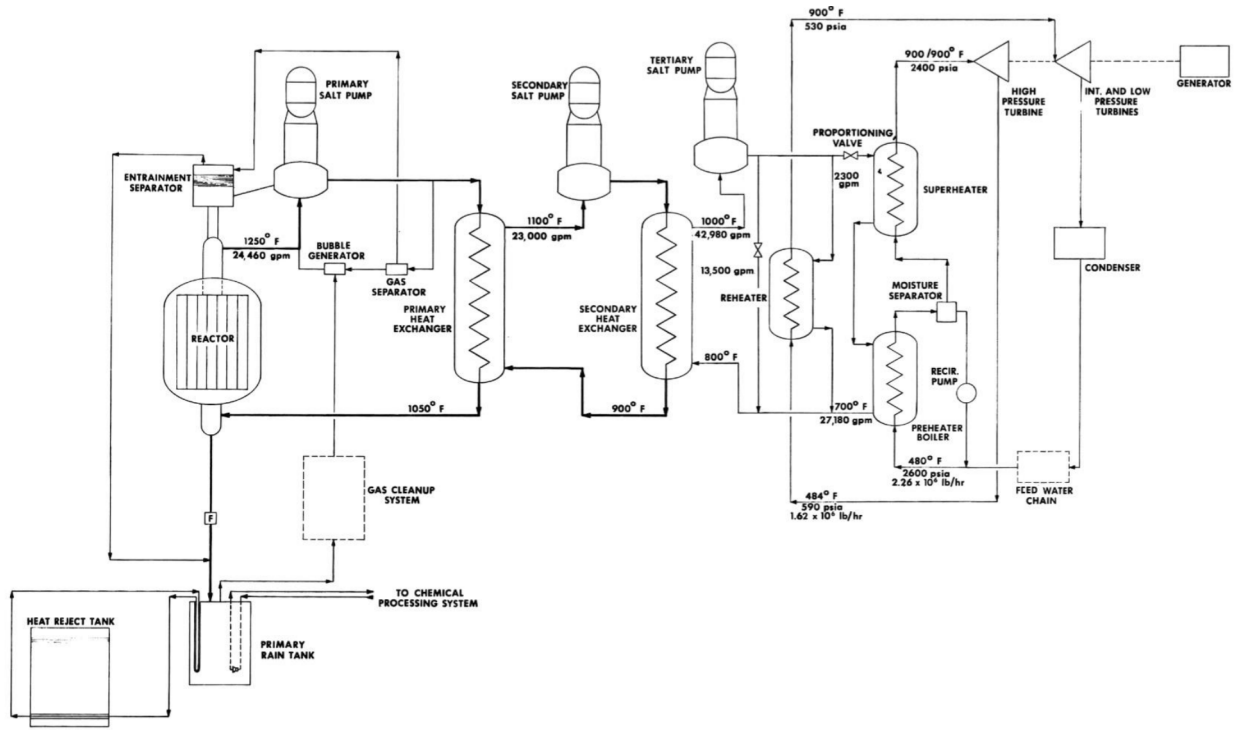


Figure 4.1: Flowsheet of MSDR [3]

Table 4.2: Some Properties of MSRE and MSBR Fuels at 600°C [65], [66]

	MSRE fuel	MSBR fuel
Composition (mole %)	LiF 65 BeF ₂ 29.1 ZrF ₄ 5 UF ₄ 0.9	LiF 71.7 BeF ₂ 16 ThF ₄ 12 UF ₄ 0.3
Liquidus (°C)	434	500
Density (g/cm ³)	2.27	3.35
Heat Capacity (cal/(g·°C))	0.47	0.324
Viscosity (centipoise)	9	12
Vapor Pressure (torr)	<0.1	<0.1
Thermal Conductivity (Watt/(°C-cm))	0.014	0.012

Table 4.3: The Freezing and Boiling Points for Chosen Salts [60]

Salt constituents	Freezing point °C	Boiling point °C
LiF	845	1681
NaF	995	1704
KF	856	1502
RbF	775	1408
BeF ₂	555	1327
ZrF ₄	903	600 (sublimes)
LiF-NaF-KF (46.5-11.5-42)	454	1570
LiF-BeF ₂ (67-33)	458	1400
NaF-BeF ₂ (57-43)	340	1400
NaF-ZrF ₄ (59.5-40.5)	500	1350
RbF-ZrF ₄ (58-42)	410	1450
LiF-NaF-BeF ₂ (67-33)	315	*
NaF-RbF-ZrF ₄ (59.5-40.5)	420	*
LiF-NaF-ZrF ₄ (59.5-40.5)	436	*

* Data not available

fuel salt and LiF-BeF₂ salt can be found in Tables 4.4 and 4.5. In terms of source term, the melting point is important for safety aspects since the salt will become solid and fission products will be contained within the salt if the temperature gets too low. This will greatly reduce the source term since fission products, actinides, and other constituents in the fuel salt will be trapped and unable to escape in the event of an accident.

4.3 Vapor Pressure and Vapor Species

Most fluoride salts exhibit very low vapor pressures [60]. The boiling points of some salt components and salt compositions can be seen in Table 4.3. Salts with very low vapor pressures (<1 mmHg at 900°C) are favored, and Forsberg suggests in [9] using salts with boiling points in excess of 1400°C. In addition, salts where the vapor species generated will melt easily after condensing are favored. The vapor produced needs to be evaluated even for low-pressure systems due to worries around the nature and magnitude of the vapor,

Table 4.4: Physical Properties for MSRE Fuel Salt [64]

Property	Value	Uncertainty
Melting Point	434°C	± 3°C
Vapor Pressure	$\log P(\text{torrs})=8.0-10,000T(^{\circ}\text{K})$	Factor 50 from 500-700°C
Density (Liquid)	$\rho=2.575-5.13 \times 10^{-4}T(^{\circ}\text{C})$ =139.9 lb/ft ³ at 650°C	± 1%
Viscosity	$\eta(\text{centipoises})=0.116 \exp [3755/T(^{\circ}\text{K})]$	± 7
Thermal Conductivity	0.010 watt cm ⁻¹ °C ⁻¹	± 10%
Heat Capacity		
Liquid	$C_p=0.57 \text{ cal g}^{-1} \text{ }^{\circ}\text{C}^{-1}$	± 3%
Solid	$C_p=0.31+3.61 \times 10^{-4}T(^{\circ}\text{C}) \text{ cal g}^{-1} \text{ }^{\circ}\text{C}^{-1}$	± 3%

Table 4.5: Physical Properties for LiF-BeF₂ Salt [64]

Property	Value	Uncertainty
Melting Point	459.1°C	± 0.2°C
Vapor Pressure	$\log P(\text{torrs})=8.0-10,000T(^{\circ}\text{K})$	Factor 50 from 500-700°C
Density		
Liquid	$\rho=2.214-4.2 \times 10^{-4}T(^{\circ}\text{C})$ =122 lb/ft ³ at 650°C	± 1%
Solid	$\rho=2.1953 \text{ g/cm}^3$	
Viscosity	$\eta(\text{centipoises})=0.116 \exp [3755/T(^{\circ}\text{K})]$	± 7
Thermal Conductivity	0.010 watt cm ⁻¹ °C ⁻¹	± 10%
Heat Capacity		
Liquid	$C_p=0.57 \text{ cal g}^{-1} \text{ }^{\circ}\text{C}^{-1}$	± 3%
Solid	$C_p=0.31+3.61 \times 10^{-4}T(^{\circ}\text{C}) \text{ cal g}^{-1} \text{ }^{\circ}\text{C}^{-1}$	± 3%

which is very important to source term since some elements are more volatile than others under different conditions. Experience with ARE and MSRE shows that very low salt vapor pressures (<1 mmHg) simplify the off-gas system design and that certain vapor species can present problems [60]. The Lewis acid-base theory explains the effect of the salt composition on vapor pressure. If there are significant salt vapor amounts in the cover gas systems, problems with transport can happen. For ARE, the vapor was almost pure ZrF_4 ; since ZrF_4 sublimates, ZrF_4 snow was found in the exhaust piping [41]. Snow traps were designed to mitigate this issue, but this issue could be eliminated completely by choosing salt composition wisely [41].

Helium cover pressure for the MSRE was 0.35 atm, and for the MSBR, it was designed to be 2.75 atm. For MSRE, the vapor pressure was 100 times lower than for ARE. The vapor above the MSRE salts contained BeF_2 and LiF in a proportion that would melt at a low temperature. If this happened, this condensate would drain as a liquid to the reservoir. The vapor pressure for the MSRE fuel salt and LiF-BeF_2 can be seen in Tables 4.4 and 4.5.

4.4 Density

Density is used to indicate the heat transfer capabilities in forced and natural convection, and it has been shown that with increasing temperature comes a linearly decreasing density. Density can be predicted accurately using established methods and can also be relatively easily measured [40], [67]. Salts that have a large coefficient of thermal expansion may remove heat by natural convection better, resulting in better cooling [41]. One method for prediction uses the relation in Equation 4.2 from [41] to calculate the approximate density of the mixture.

$$\rho_{\text{mix}}(T) = \frac{\sum_i x_i M_i}{\sum_i x_i V_i(T)} \quad (4.2)$$

ρ_{mix} is the density of the mixture (g/cm^3)

x_i is the mole fraction of component i

Table 4.6: Density Equations for Some Salt Compositions [60]

Salt Composition	Molar Composition (%)	Density equation with temperature in °C (kg/m^3)
KF-ZrF ₄	58-42	3416 - 0.887 x T
NaF-ZrF ₄	59.5-40.5	3650 - 0.880 x T
LiF-NaF-KF	46.5-11.5-42	2530 - 0.730 x T
LiF-NaF-ZrF ₄	26-37-37	3533 - 0.517 x T
KF-KBF ₄	25-75	2258 - 0.803 x T
RbF-RbBF ₄	31-69	2946 - 1.047 x T
NaF-NaBF ₄	8-92	2252 - 0.711 x T

M_i is the molecular weight of component i (g/mole)

V_i (T) is the molecular volume of component i at temperature T.

According to [61], the density of the MSRE fuel salt at 600°C is 142 lb/ft³ (2.27 g/mL) and for the MSRE flush salts the density at 600°C is 120 lb/ft³. Some density equations for various salts can be found in Table 4.6. Density values, according to Thoma in [64], for the MSRE fuel salt and LiF-BeF₂ salt can be found in Tables 4.4 and 4.5.

4.5 Viscosity

Viscosity quantifies the resistance to flow for a mixture and is dependent on temperature with an exponential increase in viscosity happening with decreasing temperature. According to Samuel, the viscosity of coolant salts should be <10 centipoise at operational temperatures as a general rule [68]. One equation to calculate viscosity from [41] is shown in Equation 4.3.

$$\mu = A \exp\left(-\frac{B}{T}\right) \quad (4.3)$$

μ is the dynamic viscosity in centipoise (cP)

T is the temperature in Kelvin

A and B are empirical constants.

Currently, there are no methods that have been established to determine the constants used in the above equation. This means the viscosity values have to be determined experimentally, but for some salt mixtures, using a mole fraction weighted sum as in Equation 4.4 from Kendall and Monroe in [69] to estimate viscosity may be appropriate if experimental data does not exist, but it can only be used if the salt is close to an ideal mixture.

$$\mu_{\text{ideal mixture}} = [\sum X_i \mu_i^{1/3}]^3 \quad (4.4)$$

X_i is the mole fraction of component i

μ_i is the dynamic viscosity of component i in centipoise

$\mu_{\text{idealmixture}}$ is the dynamic viscosity of the ideal mixture of elements

Viscosity values for the MSRE fuel salt and LiF-BeF₂ salt, according to Thoma in [64], can be found in Tables 4.4 and 4.5.

4.6 Thermal Conductivity

In molten salts, there are two methods of thermal conduction: the vibration mechanism, due to the mechanical behavior of liquid molecules, and the diffusion mechanism, due to movement of ions [70]. Thermal conductivity is needed for heat exchanger design. According to Williams in [41] and [38], thermal conductivity is the most challenging thermophysical property to measure for the relevant molten salt mixtures for MSR designs with more experimental data needing to be obtained to sufficiently understand thermal conductivity.

Cornwell in [70] derived the following expression from the most reliable experimental data despite the difficulty with uncertainties:

$$k = 0.119 \frac{T_m^{0.5} \nu^{0.667}}{(M/n)^{1.167}} \quad (4.5)$$

k is the thermal conductivity in W/m K

T_m is the salt mixture melting point in Kelvin

ν is the molar volume of the salt in cm^3/mol

M is the average formula weight of the salt

n is number of ions per salt formula

The much simpler correlation equation below was recommended by Khoklov [71] and Ignatiev et al. [72]

$$k = 5 \times 10^{-4}T + \frac{32.0}{M} - 0.34 \quad (4.6)$$

Thermal conductivity values for the MSRE fuel salt and LiF-BeF₂ salt, according to Thoma in [64], can be found in Tables 4.4 and 4.5.

4.7 Heat Capacity

Molten salts have relatively large heat capacities, and after taking density differences into account, some of the fluoride mixtures have a heat carrying capacity similar to water [41], [38]. Empirical relationships have to be used to estimate heat capacities for salt compositions since first principles methods are not fully developed. The empirical relations normally involve an estimation based on mole fraction-weighted contributions from the pure components of the mixture and taking mixing enthalpies into account when available [41], [38]. This relationship by Cantor in [67] can be seen in Equation 4.7.

$$c_p = 8 \frac{\sum_{i=1}^n X_i N_i}{\sum_{i=1}^n X_i M_i} \quad (4.7)$$

C_p is the specific heat capacity in cal/g-K

X_i is the mole fraction of the component i

N_i is the number of i atoms per formula salt component in g/mole (the value for N_i is 2 for alkali halides, 3 for BeF₂, and 5 for ZrF₄ [41], [38])

M_i is the molecular weight of component i in g/mole.

In Equation 4.7, the value of 8 cal/(K-gram-atom) of each atom in the mixture is assumed from the Dulong-Petit law. This method is generally accurate to within 10% for BeF₂ and ZrF₄ bearing salts and to within 20% for other Advanced High Temperature Reactor (AHTR) and MSR coolant candidate salts. In general for temperature ranges relevant to MSR designs, heat capacities do not vary considerably [31]. More experimental data is needed to understand heat capacity for molten salts. The heat capacities for MSRE fuel salt and LiF-BeF₂ salt can be found in Tables 4.4 and 4.5.

4.8 Material Solubility

The standard unit for solubility is g/L, which is grams of solute per liter of solution. In this case, the solute would be the fission products and the solution would be the molten salt.

4.8.1 Soluble fission products

- Rb, Ce, Sr, Ba, Y, the lanthanides, and Zr all form stable fluorides that are relatively soluble in the fuel salts.
- Br and I are expected to appear as soluble in the fuel especially if the fuel contains an appreciable concentration of UF₃.
- I-131 deposits on metal or graphite surfaces in the core region. Analysis of the MSRE core showed a large fraction of iodine present in the fuel. Material balances for I-131 were generally low. It is possible that some of the precursor, Te-131 (25min), was volatilized and sparged with Kr and Xe. Also I-131 produced by decay of Te-131 in complex metallic deposits (as in the heat exchanger) may not have been able to return to the salt [73].

4.8.2 Noble and Semi Noble Fission Products

- Some FP metals have fluorides that are unstable toward reduction by fuel mixtures with appreciable concentrations of UF_3 ; thus it must be expected to exist entirely in the elemental state in the MSR. These FPs are Ge, As, Nb, Mo, Ru, Rh, Pd, Ag, Cd, Sn, and Sb [73].
- Selenium and tellurium were expected to be present as elements within the reactor circuit [73].
- Precipitation on the metal surfaces such as the heat exchanger will be insufficient to impede fuel flow, but radioactive decay of the deposited material contributes to heat generation during reactor shutdown.
- Operation of the MSRE implicated tellurium fission products as responsible for the embrittlement of the metal surface exposed [73].
- Kr and Xe form no compounds under conditions existing in an MSR. They are only sparingly soluble in molten fluoride mixtures. This low solubility is a distinct advantage because it enables the easy removal of Xe and Kr from the reactor by gas sparging [73].

The groupings and typical elements for different solubilities are given in Table 4.7.

Table 4.7: Volatility of Different Elements. Adapted from [74].

Solubility	Elements
Gaseous	Kr, Xe
Highly Volatile	Cs, I, Sb, Te
Moderately Volatile	Ru, Mo, Sr, Ba
Lowly Volatile	Eu, Zr, Ce
Actinide	Pu

Chapter 5

System Analysis

MSRs potentially present many advantages in regards to neutronics such as low excess reactivity due to batch refueling being an option, online removal of fission products, and also a strong potential for inherent safety and stability [7]. In order to model an MSR, several categories pertaining to the reactor must be explored including thermal hydraulics, mass transport, thermochemistry, reactor physics, and corrosion. Thermophysical properties of the fuel salt influence reactor performance. Also, multiphysics simulations are needed to understand how the salt behaves during the lifetime of the reactor, as well as how it affects the reactor components. This chapter begins with an overview of the aspects important to systems analysis with a discussion about modeling provided at the end.

5.1 Delayed Neutron Precursors

Delayed neutron precursors are radioactive fission products that release delayed neutrons upon decay [75]. In LWRs, the delayed neutron emission is very close to the site of the fission resulting in an effect on neutron source in the core. In MSRs, the delayed neutron emission location and site of the fission are different compared to solid-fueled reactors since it can happen inside the core or elsewhere within the loop or system. The effective delayed neutron fraction (β_{eff}) for MSRs is affected by the flow loop size, pipe diameter, flow rate, and other flow characteristics. Delayed neutron production happens on a large enough time

scale to allow a reactor to be controlled, implying that β_{eff} has reactor safety implications [76]. For time-dependent analysis of an MSR, it is vital to account for the delayed neutron precursor drift since it changes the energy spectrum and strength of the neutron source which affects the depletion calculation. The distance it travels and the time of neutron emission are directly related to reactor behavior since the distance is determined by the flow rate of the fuel salt; the flow rate could potentially be changed giving more control. Due to the delayed neutron fraction, decay heat will be deposited outside the core changing how reactor kinetics will need to be treated for salt-fueled reactors. Table 5.1 shows the 8 delayed neutron groups and their half-lives and yields. The delayed neutron fraction is typically small for circulating-fuel reactors. The MSBR's calculated values are in the range of 0.0010-0.0015 [65], whereas it is around 0.006 for LWR and 0.005 for CANDU [4].

5.2 Point Reactor Kinetics

The point kinetics equation used for typical solid fueled reactors is below [4]:

$$\frac{dn}{dt} = \frac{\rho(t) - \beta}{\Lambda} n(t) + \sum_{i=1}^6 \lambda_i C_i(t) \quad (5.1)$$

$$\frac{dC_i}{dt} = \frac{\beta_i}{\Lambda} n(t) - \lambda_i C_i(t), i = 1, \dots, 6 \quad (5.2)$$

Table 5.1: Delayed-Neutron Group Data for U-235 thermal [77]

Group	Half-life (s)	Relative Yield
1	55.6	0.033
2	24.5	0.154
3	16.3	0.091
4	5.21	0.197
5	2.37	0.331
6	1.040	0.090
7	0.424	0.081
8	0.195	0.023

$\rho(t)$ is the reactivity as a function of time

β is the total fraction of delayed neutrons

Λ is the prompt neutron generation lifetime

$n(t)$ is the neutron density as a function of time

λ_i is the decay constant of the delayed neutron precursor group i

$C_i(t)$ is the concentration of delayed neutron precursor group i as a function of time

β_i is the fraction of delayed neutrons of group i

$\lambda_i C_i(t)$ is the reactivity rate, which is the radioactive decay of the delayed neutron precursors

In [MSRs](#), the transport of delayed neutron precursors and subsequent emitted delayed neutrons outside of the core must be taken into account for reactivity effects; the reactor kinetics equations will need to be modified to account for this. Equation 5.3 from [\[78\]](#) and 5.4 from [\[79\]](#) show the kinetic equations for [MSRs](#)

$$\frac{dn}{dt} = \frac{\rho(t) - \beta}{\Lambda} n(t) + \sum_{i=1}^6 \lambda_i C_i(t) \quad (5.3)$$

$$\frac{dC_i}{dt} = \frac{\beta_i}{\Lambda} n(t) - \lambda_i C_i(t) - \frac{C_i(t)}{\tau_c} + \frac{C_i(t - \tau_l) \exp(-\lambda_i \tau_l)}{\tau_c}, i = 1, \dots, 6 \quad (5.4)$$

τ_c is fuel residence time in the core

τ_l is fuel residence time in the primary loop

The neutron equations, Equations 5.1 and 5.3, are the same for both solid-fueled and liquid-fueled nuclear reactors since where the neutron is formed, whether inside the core or outside the core, the amount of delayed neutrons stays the same. The only difference is all the delayed neutrons happen inside the core for solid-fueled reactors, whereas some of the delayed neutrons will happen in the primary loop of an [MSR](#). When comparing Equations 5.4 to 5.2, the additions are the residence times in the core and the primary loop accounting for the delayed neutron precursor drift. The core transit time and primary loop transit time for the [MSRE](#) can be seen in Table 5.2.

5.3 Reactivity Controls and Effects

The temperature coefficient of reactivity is the effect of the core temperature on the multiplication of the core and can be expressed as Equation 5.5 given by [4]:

$$\alpha_T = \frac{\partial \rho}{\partial T} \quad (5.5)$$

α_T is the temperature coefficient of reactivity

$\partial \rho$ is the change in reactivity

∂T is the change in temperature

A negative temperature coefficient of reactivity is preferred, as it indicates that the reactivity of the system decreases with increasing temperature, preventing any sort of runaway reaction. This is a great safety feature which helps to restore the system to a safe temperature.

Reactivity control is different for [MSRs](#) than what is done for most solid fueled reactors including adjusting fuel salt composition to compensate for long-term reactivity changes [65]. The temperature coefficients of reactivity for a single-fluid [MSBR](#) is not significantly large compared to other fluid-fuel reactors [65]. Some of the larger individual effects are the Doppler effect and moderator coefficient [65]. For the [MSRE](#), the measured and calculated temperature coefficients showed agreement within 10% [81]. However, the temperature coefficients found for the [MSRE](#) showed a large dependence on neutron leakage, this analysis

Table 5.2: Neutronics Parameters for U-235 for the [MSRE](#) [20], [80]

Property	Value
Prompt neutron lifetime (Λ)	4×10^{-4} s
Delayed-neutron fraction (β)	0.0065
Core Transit Time (τ_C)	8.46 s
External Loop Transit Time (τ_l)	16.73 s
Fuel salt reactivity coefficient (α_f)	$-8.71 \times 10^{-5} \delta \rho / ^\circ\text{C}$
Graphite reactivity coefficient (α_g)	$-6.66 \times 10^{-5} \delta \rho / ^\circ\text{C}$

may not be applicable to a large-scale power [MSR](#). The reactivity coefficients are similar at least qualitatively to those for a natural-uranium, graphite moderated, gas-cooled reactor.

The multiplication factor, k , is directly related to the neutron balance and can be approximated using the six-factor formula seen in Equation [5.7](#). k is the ratio of the number of neutrons in the next generation over the number of neutrons in the current generation. The relationship between reactivity and the multiplication factor can be seen in Equation [5.6](#). If there is no leakage out of the system k becomes k_∞ , which uses the four-factor formula for approximation; the four factor formula can be seen in Equation [5.8](#). A comparison of calculated and measured k_∞ values are shown in Table [5.3](#). The reactivity effect of fuel circulation was measured to be $0.212 \pm 0.004\% \delta k/k$ and the calculated value was 0.222% for U-235 fuel [\[81\]](#).

$$\rho = \frac{k - 1}{k} \quad (5.6)$$

$$k = \eta f p \epsilon P_{FNL} P_{TNL} \quad (5.7)$$

$$k_\infty = \eta f p \epsilon \quad (5.8)$$

ρ is the reactivity k is the multiplication factor k_∞ is the infinite multiplication factor

η is the reproduction factor

f is the thermal utilization factor

p is the resonance escape probability

ϵ is the fast fission factor

P_{FNL} is the probability that a fast neutron will not leak out of the system

P_{TNL} is the probability that a thermal neutron will not leak out of the system

5.4 Neutron Flux

Due to the adjustable fuel composition and fluid movement in [MSRs](#), flux calculations are not important on their own but they do serve the purpose of establishing reactor conditions

Table 5.3: Comparison of k_{∞} for the High Temperature Lattice Test Reactor (HTLTR)-MSBR Lattice [65]

	20°C	300°C	627°C	1000°C
measured	1.0291±0.0012	1.0127±0.0010	1.0065±0.0010	1.0036±0.0012
calculated	1.0300	1.0138	1.0057	1.0030

[65]. There is sensitivity in the calculated concentrations; almost all of this is due to neutron leakage [65]. The MSRE had unusually high neutron leakage with 31% for U-235 and 35% for U-233 fuel; but the neutron leakage would be considerably lower (factor of 20-30) in a large molten salt power reactor [65].

5.5 Frequency Response and Reactor Stability

The dynamic behavior of a multiple loop system depends on the properties of all parts of the system and how they are linked to form the system [65]. Power densities, heat transfer coefficients, heat capacities, and salt circulation rates are just as important as the neutronic characteristics in an MSR [65]. To determine where controls need to provide intervention within the system, a study to determine the system's inherent dynamic behavior needs to be conducted to understand the self-controlling parts of the system [65]. Transient response to various perturbations and analysis of frequency response are some techniques that have been used to study the dynamic characteristics of the MSRE and other prospective molten salt power reactor designs [65]. All of these studies involving standard techniques and sensitivity studies have shown that both the MSRE and MSBR are stable, tractable systems with increased power levels resulting in increased stability [65]. Although there is not any direct power density experimental data for MSR, some relevant experience exists for heavy water reactors and large gas-cooled graphite reactors [65]. A high sensitivity of power distribution is expected with small variations of k_{∞} [65].

The fuel is constantly mixing and maintains a homogeneous mixture, although potential changes in chemical behavior during downtime in the reactor is a major concern and should

be studied further. The composition of the fuel salt does change through the lifetime of the reactor due to fissioning of the fuel, fuel additions or withdrawals, chemical reactions between the walls and the fuel, composition adjustments, and actinide changes due to neutron transmutation, but stays homogeneous.

5.6 Thermochemistry and Corrosion

Alloys with high concentrations of chromium, aluminum, or silicon are typically used in most high temperature applications since they readily form passive oxide films, which slows the transport of species between the metal and the environment preventing further corrosion; however, these oxides are either unstable due to the low oxygen activities or do not form at all in most halide salts, making degradation dependent on reactions between the bare metal surface and the molten salt [82], [83], and [84].

Alloy constituents tend to leech into the salt from the alloy according to the stability of the halides in the salt [82]. The leeching of the elements in the molten fluoride salts corresponds well with the stability of the metallic fluorides in the salt [82], [85]. Alloy constituents that form halides with the most negative Gibbs free energies are the most likely to deplete into the melt from the alloy; selective depletion of chromium is the dominant form of material degradation in molten halides for most common structural alloys since it is the major constituent with the most stable halide [82].

Due to the large effect of purity of the salt on corrosion rate, only pure salts are included in this discussion. In order to include the most relevant data, only salt corrosion experiments lasting 500 hours or longer are listed in Table 5.4. The data in Table 5.4 was compiled by

Table 5.4: Corrosion [82], [86], [87]

Salt	Container Material	Time (h)	Temp (°C)	Material	Method	Mass Change (mg/cm ²)	Corrosion Rate (mg/cm ² /day)	Salt Notes	Reference
LiF-BeF ₂ -ThF ₄	Alloy N	1610	663	Alloy N	Loop	-0.4	-5.96E-03	Hydrofluorinated	[88]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	17719	685	Alloy N	Loop	-0.34	-4.61E-04	Hydrofluorinated	[88]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	17719	705	Alloy N	Loop	-0.7	-9.48E-04	Hydrofluorinated	[88]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	2776	690	Alloy N	Loop	-0.15	-1.30E-03	Hydrofluorinated	[88]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	4984	704	Alloy N	Loop	-3.07	-1.48E-02	Hydrofluorinated	[89]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	3206	704	Alloy N	Loop	-0.94	-7.04E-03	Hydrofluorinated	[89]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	10000	704	Alloy N	Loop	-3.35	-8.04E-03	Hydrofluorinated	[90]
LiF-BeF ₂ -ThF ₄ -UF ₄	Alloy N	4309	704	Alloy N	Loop	-2.31	-1.29E-02	Hydrofluorinated	[90]
LiF-NaF-KF	Graphite	2000	700	Alloy N	Capsule	1.84	2.21E-02	Sparged with H ₂ and argon	[91]
LiF-NaF-KF	Graphite	2000	700	Alloy N	Capsule	0.38	4.56E-03	Sparged with H ₂ and argon	[91]
LiF-NaF-KF	Graphite	500	850	Alloy N	Capsule	-2.3	-1.10E-01	HF purified salt from ORNL	[86]
LiF-NaF-KF	Graphite	500	850	Alloy X	Capsule	-13.2	-6.34E-01	HF purified salt from ORNL	[86]
LiF-NaF-KF	Graphite	500	850	Haynes 230	Capsule	-51.4	-2.47E+00	HF purified salt from ORNL	[86]
LiF-NaF-KF	Graphite	500	850	Alloy 617	Capsule	-29.7	-1.43E+00	HF purified salt from ORNL	[86]
LiF-NaF-KF	Graphite	500	850	Alloy 800H	Capsule	-28.6	-1.37E+00	HF purified salt from ORNL	[86]
LiF-NaF-KF	Graphite	500	850	Alloy 800H	Capsule	-10.8	-5.18E-01	Purified with Mg addition	[86]
LiF-BeF ₂	Graphite	3000	700	SS 316	Capsule	0.47	3.76E-03	Hydrofluorinated	[87]
LiF-BeF ₂	Graphite	1000	700	SS 316	Capsule	-0.18	-4.32E-03	MSRE salt, Hydrofluorinated	[92]
LiF-BeF ₂	Graphite	1000	700	Alloy N	Capsule	0.17	4.08E-03	MSRE salt, Hydrofluorinated	[92]

Raiman and Lee in [82].

The salts seen in Table 5.4 are $\text{LiF-BeF}_2\text{-ThF}_4$, $\text{LiF-BeF}_2\text{-ThF}_4\text{-UF}_4$, LiF-NaF-KF , and LiF-BeF_2 at temperatures between 663-850°C. The length of these experiments varied from 500-3000 hours. The corrosion testing in [86], [87], [91], and [92] was done in sealed capsules. Sealed capsules allow for better control of impurity compared to open crucibles; the capsules, typically metallic, are filled with the solid salt, welded shut, and heated in a furnace. Keiser et al. [89], [90] and Koger et al. [88] used thermal convection loop experiments. Loop experiments are more realistic than capsule experiments since it allows for the temperature gradient typical of reactors to be taken into account [82]. The temperature difference drives both the salt flow through the loops and corrosion due to differential solubility of corrosion products with temperature [82].

Keiser et al. [89], [90] and Koger et al. [88] were the only experiments that ran for over 3000 hours; these were loop experiments at ORNL; these exhibited low corrosion rates and the salt composition was clean, fuel containing fluoride [82]. There were some more experiments run with MSRE salts purified by hydrofluorination before the corrosion testing experiment by Olson [86] and Zheng [87], [92]; these were capsule experiments and show low corrosion rates as well [82].

5.7 Radiation Stability

Radiation transport causes different problems in an MSR compared to traditional reactors since the fuel is circulated throughout the reactor instead of just being within the core. Design problems such as neutron or gamma heating, radiation damage, and shielding are due to radiation transport, but currently there are not any unique problems present for the MSR transport problem; rather the problems will deal with the distribution of radiation sources throughout the plant [65].

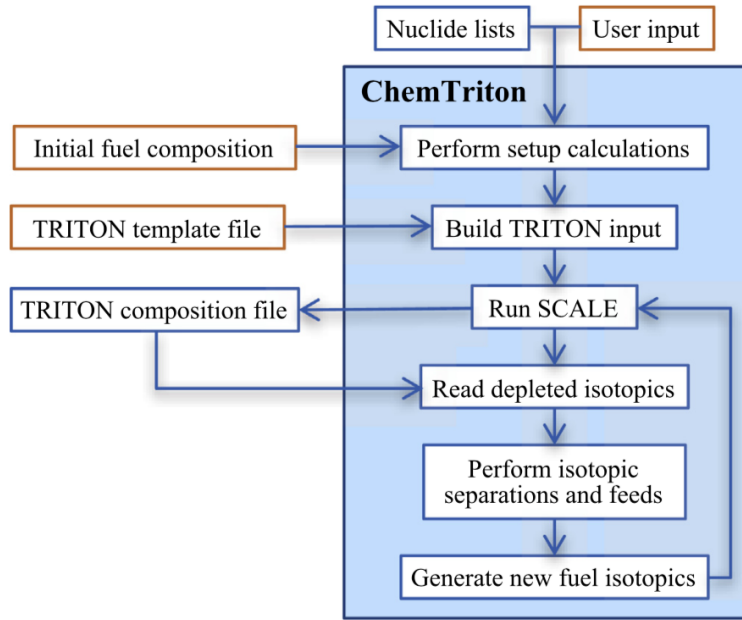


Figure 5.1: ChemTriton flow chart [76]

5.8 Modeling and Simulation

In the modeling tool SCALE/TRITON, ORIGEN solves a set of Bateman equations using spectrum-averaged fluxes and cross sections provided from a transport calculation [76]. For solid fuel, fuel is stationary so there is no material removal or feed term in the equation. For liquid fuel, this term is likely not zero, and the feed/removal term must be included; this term represents the removal of FPs and addition of fertile and fissile material. Reactor design and liquid fuel flow rates must be taken into account to get an accurate removal/feed rate. For these to be modeled, the simulation must stop at the given time and then start again with the new composition. This process can be seen in Figure 5.1. The process starts by creating a TRITON input using a template file, nuclide lists, and other user input. Next, SCALE is run to produce a TRITON composition file and depleted isotopics are read from this. From there, the separations and feeds of isotopes are calculated to generate new fuel isotopics, which then starts the process over again at the step where SCALE is run. Most of this process is in ChemTriton, an object-oriented Python script using a semi-continuous batch process in order to simulate a continuous process [76]. It models changing isotopic composition of an irradiated fuel salt using SCALE for neutron transport and depletion

calculations [75]. The primary function of ChemTriton is chemical mixture management with depletion and neutron transport aspects performed by SCALE. Some features added to ChemTriton for MSR analyses are use of a generic input file for SCALE/TRITON, SCALE standard composition reading and writing, post-processing tools to list isotopic, flux, and four factor formula information during operation, multiple irradiation zone capabilities, time-dependent fuel feed and isotopic separations, and critical concentration search capabilities [76].

ChemTriton simulates continuous removals and feeds by maintaining an iterative approach, with each material stream being treated as an object. The material stream main characteristics consist of volume, isotopic composition, temperature, and heavy metal mass. The available actions include reading and writing stream isotopic information, separation of specific isotopes, addition of specific isotopes, and combining and splitting streams [76]. Currently, ChemTriton does not take delayed neutron precursor drift into consideration; in order to account for this, a correction factor or addition of a convective term must be added to the neutron transport equation to understand the effects on both neutronics and depletion [93].

5.8.1 Gaps in Modeling and Simulation

Precursor drift needs to be modeled to understand its effects on neutronics and depletion. Modern modeling and simulation tools and data are needed for integral benchmarks for reactor physics, thermal hydraulics, material properties and response models, and coolant/fuel/structure chemistry/corrosion [75]. Taking burnup into account is essential to accurately determine the composition of the fuel salt in order to model MSRs, current tools still need to improve in this aspect. Thermal hydraulics and neutronics calculations must be performed together in an MSR since there are interactions between the salt chemistry, neutronics, and thermal hydraulics that are complex. In most other reactors, thermal hydraulics and neutronics can be analyzed separately. The models to be developed can be made up of empirical relationships from data, analytical equations, and/or fitting to a

mathematical form.

Chapter 6

Operational Fuel Management

Fuel management is a unique aspect of [MSRs](#) since the composition of the salt can be changed by employing fission product removal or fissile material feed either in a continuous or batch process. Some of the important radionuclide groups and radionuclides can be seen in [Table 6.1](#). These radionuclides and some of the reprocessing methods employed will be discussed in this chapter.

Table 6.1: Important Radionuclides and Their Groupings

Group	Radioisotopes
Noble Gas	Xe, Kr
Noble Metals	Nb, Mo, Tc, Ru, Rh, Ag, Cd, In, Sn, Zn, Ga, Ge, As
Halogens	I, Br
Tellurium Group	Te, Sb, Se
Barium, Strontium	Ba, Sr
Rare Earth/Alkaline Metals	Y, La, Ce, Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er, Zr, Sm, Eu, Sr, Ba, Rb, Cs
Actinides	U, Pu, Np
Corrosion Products	Ni, Fe, Cr

6.1 Fuel Qualification

The [NRC](#) defines fuel qualification as "a process which provides high confidence that physical and chemical behavior of fuel is sufficiently understood so that it can be adequately modeled for both normal and accident conditions, reflecting the role of the fuel design in the overall safety of the facility. Uncertainties are understood such that any calculated fission product releases include appropriate margin to ensure conservative calculation of radiological dose consequences." [94]. For [MSRs](#), unique aspects of the reactor need to be considered by the [NRC](#) to ensure it meets the advanced reactor fuel qualification. These unique features include the fuel also acting as the coolant, the fuel being molten, fuel salts being highly corrosive, no cladding for fission product containment, fuel outside of the core, and freezing points above ambient temperatures. The flowing fuel salt in an [MSR](#) removes decay heat as well as being the principal heat source. The fuel salt composition will change due to fission products, which will affect the associated thermophysical properties. Understanding the impact this causes will be vital to the analyses on heat removal ability. Because the fuel is a liquid, the necessity to understand the mechanical behavior of the cladding, fuel, subassembly and core structures is restricted to the solid elements since there are no permanent mechanical stressors in liquids [95].

All of the fuel salts currently in consideration can become highly corrosive; in addition, some fuel salts can contain hazardous materials (such as beryllium) [95]. Some fission products may evolve out of the fuel salt which is discussed later, but this will change the salt composition. Some of these may also decay into other isotopes also effecting the fuel composition. The chemical composition changes of the fuel are because of refueling, chemistry control additions, corrosion buildup, and fission products. Since there is no cladding for containment of fission products, the fuel salt must be chemically compatible with the materials in the reactor. The fuel salt behavior when it freezes needs to be understood as well including plate-out, densification, stratification, decomposition and possible dimensional changes during freezing [95].

Another aspect to consider is the radiolysis behavior of the salt. Fluoride salt fuels for [MSRs](#) were determined to be radiation-tolerant while molten according to experiments to support the [MSRE](#) concerning irradiations of the fuel [40], [96]. Due to high energies produced from fission, ionic bonds may be broken, but recombination happens rapidly since the fuel salt is in a liquid state; due to this, very little fuel salt constituents will remain in a disassociated state [95]. However since this is temperature dependent, if the salt is frozen, recombination will happen much less based on the [MSRE](#) experience [97], [98]. Chloride salt is expected to act similarly to fluoride fuel salt although there is not nearly as much data for chloride salts.

Thermophysical properties will need to be taken into account in safety analysis codes, as well as how they change with time and as a result of a postulated accident or anticipated operational occurrence. Changes in the melting point, specific heat, and viscosity are of great concern since the ability to remove heat from the core could be hindered and fuel freezing could occur [95]. Fission product transport, retention, and location could be affected with changing chemical properties. Removal and build up of fission products will change the composition and thermophysical properties, as will addition of fuel. Plate-out of some fission products occurs on surfaces in contact with the fuel causing increased localized shielding requirements and increased heat deposition in certain locations; this may have criticality safety implications [95]. Plate-out of noble metals on the salt-wetted barrier is generally considered to have a protective effect [99].

6.2 Front End Processing

For [MSRE](#), all the fluoride starting materials were obtained from either commercial sources or the [USAEC](#) [61]. The lithium fluoride needs to be at least 99.99% Li-7 due to Li-6's high neutron absorption cross section. Lithium is commercially available as a hydroxide; the hydroxide is then converted to a fluoride. The uranium fluoride obtained from [USAEC](#) sources was highly enriched (93% U-235) even though only 32% enrichment was needed; this was balanced with the depleted fuel charge [61]. Zirconium tetrafluoride and beryllium fluoride were available through commercial sources [61].

Table 6.2: Salt Treatment vs. Salt Processing [100] (3 day depletion time steps)

Processing Type	Processing Group	Elements	Cycle Time	Removal Fraction ($t_{depl}=3$ days)
Salt Treatment	Volatile Gases	Xe, Kr	20 sec	1.3×10^4
	Noble Metals	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te	20 sec	1.3×10^4
	Seminoble Metals	Zr, Cd, In, Sn	200 days	0.015
	Volatile Fluorides	Br, I	60 days	0.05
Salt Processing	Protactinium	Pa	3 days	1.0
	Rare Earth Elements	Y, La, Ce, Pr, Nd, Pm, Sm, Gd, Eu	50 days	0.06
			500 days	0.006
	Discard	Rb, Sr, Cs, Ba	3435 days	8.74×10^{-4}

6.3 During Operations

Passive and active salt reprocessing can take place. In Table 6.2, salt treatments are passive processes, whereas salt processing are active processes. Salt treatment refers to controlling the species that come out of the solution of their own volition. Salt processing is primarily employed to improve performance by removing undesirable species.

6.3.1 Important Radioisotopes

The species within the salt due to fission include [101]:

- TRU: Pu, Np, Cm, Am
- Gaseous FPs: Xe, Kr, H-3
- Non-soluble FPs in the salt: As, Nb, Mo, Te, Ru, Rh, Pd, Ag
- Soluble FPs in the salt
 - Rare Earths: Sm, Eu, La, Ce, Nd, Pr, Pm

- Alkaline-earths: Rb, Cs, Sr, Ba
- Semi-noble metals: Ga, Ge, Cd, In, Sn, Sb
- Halogens: I, Br
- Protactinium-more important for thorium versus U-235.

Krypton and xenon, both noble, volatile gases, separate naturally out of the salt to the cover gas since they have almost no solubility in the salt. Noble and semi-noble metals will exist in the elemental state in the reductive fuel salts [102]. They are expected to primarily plate-out on the metal surfaces. Rb, Cs, Sr, Ba, Y, Zr, and the lathanides all form soluble, stable fluorides and are expected to stay in the fuel salt with some found in the off-gas system due to noble gas precursors.

Below are the assumptions made for the fission product behavior for the [MSBR](#):

- Elements that are present as gases and assumed to be partly absorbed by graphite and partly removed by gas stripping are Kr and Xe [103].
- Elements that plate-out on metal surfaces and are assumed to be removed instantaneously are Rh, Pd, Ag, and In [103].
- Elements that form volatile fluorides and are assumed to be removed by the fluoride volatility process are Se, Br, I, Nb, Mo, Ru, Tc, and Te [103].
- Elements that form stable fluorides that are less volatile than LiF and are assumed to be removed by vacuum distillation are Sr, Y, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, and Tb [103].
- Elements that are not separated from the carrier salt and are assumed to be removed only by salt discard are Rb, Cd, Sn, Cs, and Zr [103].

The amount of fission products produced is proportional to burnup. As can be seen in [Figure 6.1](#), the fission products with the highest yields are isotopes with mass numbers between 85-105 and 130-150. The radionuclides assessed in this thesis are in [Table 6.3](#).

Table 6.3: Nuclides of Interest [65], [104]

Nuclide	Significance	Cumulative Thermal Fission yield for ^{235}U (%)	Half -life (days)
^3H	Fission Product	0.0108 ± 0.0004	12.3 y
^{85}Kr	Fission Product	0.286 ± 0.021	10.739 y
^{89}Sr	Fission Product	$0.046896\pm 5.6798\text{E-}4$	50.563 d
^{90}Sr	Fission Product	5.73 ± 0.13	28.79 y
^{90}Y	Fission Product	0.05729 ± 0.0013192	64.00 h
^{91}Y	Fission Product	$0.058488\pm 5.2681\text{E-}4$	58.51 d
^{95}Zr	Fission Product	6.502 ± 0.072	64.032 d
^{95}Nb	Fission Product	6.498 ± 0.072	34.991 d
^{99}Mo	Fission Product	6.132 ± 0.092	65.924 h
^{99}Tc	Fission Product	6.132 ± 0.092	2.111×10^5 y
^{103}Ru	Fission Product	3.103 ± 0.084	39.247 d
^{106}Ru	Fission Product	0.41 ± 0.011	371.8 d
^{111}Ag	Fission Product	$1.9557\text{E-}4\pm 6.065\text{E-}6$	7.45 d
^{125}I	Fission Product	$2.9041\text{E-}12\pm 1.0677\text{E-}12$	59.407 d
^{129}I	Fission Product	0.706 ± 0.032	1.57×10^7 y
^{129m}Te	Fission Product	$0.0027707\pm 1.4992\text{E-}4$	33.6 d
^{131}I	Fission Product	2.878 ± 0.032	8.0252 d
^{132}Te	Fission Product	$0.042757\pm 4.2818\text{E-}4$	3.204 d

Table 6.3: Nuclides of Interest [65], [104] (continued)

Nuclide	Significance	Cumulative Thermal Fission yield for ^{235}U (%)	Half -life (days)
^{133}I	Fission Product	6.59 ± 0.11	20.83 h
^{133}Xe	Fission Product	6.6 ± 0.11	5.2475 d
^{134}Cs	Fission Product	0.0000121 ± 0.0000032	2.0652 y
^{137}Cs	Fission Product	6.221 ± 0.069	30.08 y
^{137m}Ba	Fission Product	$0.058725\pm 8.4065\text{E-}4$	2.552 min
^{140}Ba	Fission Product	6.314 ± 0.095	12.7527 d
^{140}La	Fission Product	6.315 ± 0.095	1.67855 d
^{141}Ce	Fission Product	5.86 ± 0.15	32.511 d
^{144}Ce	Fission Product	5.474 ± 0.055	284.91 d
^{147}Nd	Fission Product	$0.02232\pm 4.0176\text{E-}4$	10.98 days
^7Be	Tritium Production		53.12 d
^{51}Cr	Corrosion Product		27.7025 d
^{55}Fe	Corrosion Product		2.73 y
^{59}Fe	Corrosion Product		44.6 d
^{60}Co	Corrosion Product		5.2714 y
^{63}Ni	Corrosion Product		100 y
^{65}Zn	Corrosion Product		244 d

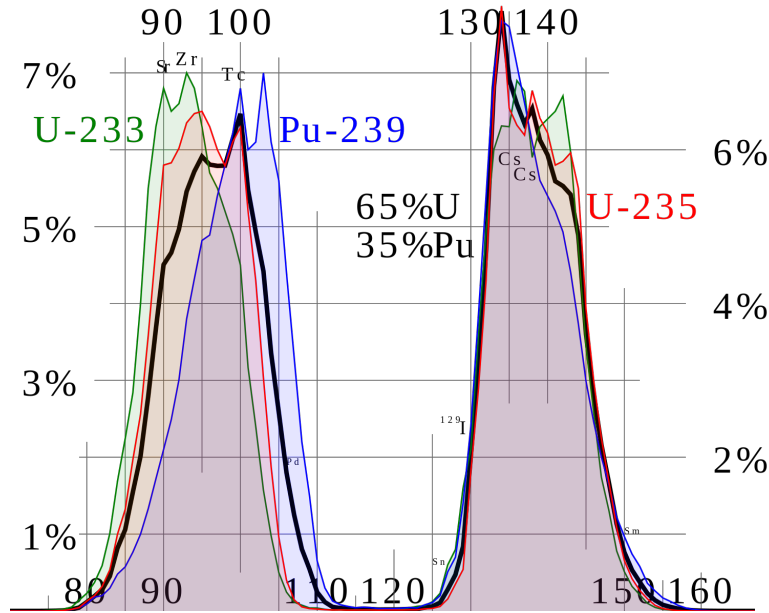


Figure 6.1: Thermal fission yield curve [105]

Some of the main radioisotopes of great concern are the fission products, Sr-90/Y-90, Cs-134, and Cs-137/Ba-137m, the gaseous fission products, Kr-85 and Xe-133, and Pu-239. The main corrosion products of concern are as follows: Chromium-51, Iron 55, Iron-59, Nickel-63, and Zn-65.

6.3.2 Reprocessing

In solid-fueled reactors, all materials, including the fission products and actinides, stay in the initial fuel during operation, but for liquid-fueled reactors, continuous or batch chemical processing can be used to remove or add material. The neutronic benefit of reprocessing comes from the removal of fission products with high absorption cross sections and/or a high concentration; the downside to this comes from the issue with storage and disposal of these materials that have been separated. An additional benefit is eliminating the need for excess reactivity. This fuel processing can take place either onsite or offsite. If only basic reconditioning of the fuel is done onsite, there will be a reduction in the regulatory challenges since it will minimize the storage and disposal issue. If fission products are actively stripped from the salt, this can lower the in-core source term in an accident situation. The processing

facility has possible accidents and a separate source term. The source term of an MSR with fuel processing is reduced by the ratio of processing time to refueling time; the reduction in source term compared to solid fueled reactors is due to the long-lived fission products [106].

Both the noble gases and metals are insoluble in the fuel salt and typically move towards the fuel salt-cover gas interface. Some of the worst poisons are the noble gases, so it is attractive to separate them from the fuel salt, especially for breeder MSRs. The noble gases inherently leave the salt since the solubility for them is very low in the fuel salt. This allows them to be stripped from the salt which enhances the removal rate. Noble gases are removed via helium sparging and then passed through charcoal traps in the off-gas system [107]. The MSRE used a fuel pump bowl for gas sparging. Aggressive sparging could result in up to 40% of the fission products in the cover gas, which is almost all of the fission products that have gaseous precursors [33]. This will cause the short term fission product trap to have a substantial heat load, but there will be lower activity in the long term fission product traps [33]. Noble metals also have low solubility towards the fuel salt, and for the MSRE, these noble metals did not form stable fluorides [108].

Since processing strategies based on combining fluorination and distillation alone are very limited in their abilities to provide efficient separations, reductive extraction is used to separate rare earth metals/elements [42]. Reductive extraction refers to the exchange chemistries that occur between molten fluoride salts and liquid bismuth alloys [42]. These reactions typically involve simultaneous exchange of two or more different metals across the interface between the salt and metal phases [42].

Rare earth metals/elements tended to stay in the fuel salt and form soluble, stable fluorides. They can be separated out using reductive extraction. Equation 6.1 from [108] shows the relationship between the metal in the molten salt and the liquid metal phases for reductive extraction.

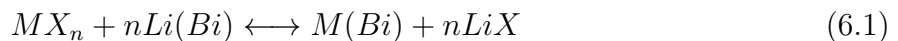
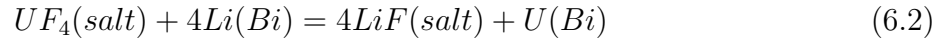


Table 6.4: Fission Product Removal Methods and Cycle Times Adapted from [109]

Method	Component	Removal time
Helium Sparging	Kr, Xe (noble gases)	50 sec
Plate-Out	Zn, Ga, Ge, As, Se, Nb, Mo, Ru, Rh, Pd, Ag, Tc, Cd, In, Sn, Sb, Te (noble and semimoble metals)	2.4 hr
Volatilization	U-233-237	10 days
	Br, I (halogens)	10 days
	Zr, Pa-233	10/200 days
Reductive extraction	Ni, Fe, Cr (corrosion products)	10/200 days
	Y, La, Ce,Pr, Nd, Pm, Gd, Tb, Dy, Ho, Er (trivalent RE's)	25/200 days
	Sm, Eu (divalent RE's), Sr, Ba	25/200 days
	Rb, Cs	10-200 days
Salt discard	Li, Be, other salt components	8-30 years

In this process, the metal halide (MX_n) reacts with lithium bismuth from the liquid phase to produce M in the liquid phase and lithium halide in the salt phase.

A specific example of this can be seen in Equation 6.2 from [42]



In this reaction there is an exchange between uranium in the salt and the dissolved lithium in the bismuth. The uranium is reduced, while the lithium is oxidized serving as the reductant [42].

Reprocessing of different elements is summed up in Table 6.4 along with their typical removal times. Also, Figure 6.2 shows a schematic of the typical separation processes.

Table 6.5: The Effect of Removal of Different Groups of Elements on Core Lifetime [76]

Categories Removed	Core Lifetime	
	Time (years)	Additional (+%)
None	2.73	-
Discard	2.73	0.2
Seminoble Metals	2.74	0.3
Volatile Fluorides	2.74	0.4
Noble Metals	2.92	7.1
Volatile Gases	2.93	7.5
Rare Earth Elements	3.12	14.4
Gases, Noble Metals	3.14	15.1
Gases, Noble Metals, Discard	3.14	15.1
Gases, Noble Metals, Seminoble Metals	3.14	15.2
Gases, Noble Metals, Fluorides	3.14	15.2
Gases, Noble Metals, Rare Earth Elements	3.63	32.9

6.5 Tritium

Tritium is of concern mostly due to the amount of it produced by lithium-bearing salts. If the system has lithium as a salt component, tritium can be produced at order of magnitudes higher amounts than [LWRs](#) due to Li-6 contamination [\[75\]](#). For a 1000MW_e reactor, tritium production for [MSRs](#) is estimated to be 2400 Ci/day; for [PWRs](#), 2 Ci/day; and for [PHWRs](#) like [CANDU](#), 2700 Ci/day [\[112\]](#). Some type of tritium recovery will most likely be needed to control environmental releases and worker exposure in all cases. In the [MSRE](#), tritium production was minimized by using fuel salt that was enriched to 99.993% Li-7. It was found that tritium migrated from the fuel salt, through the heat exchanger into the coolant salt, where it went through the radiator into the air. It also migrated through the fuel salt vessel and piping into the cover gas system.

Chapter 7

Regulatory Considerations

7.1 NUREG-1537

NUREG-1537 Parts 1 and 2 were published in 1996; it is titled Guidelines for Preparing and Reviewing Applications for the Licensing of Non-Power Reactors [113]. This is relevant to MSR development since a test reactor will need to be constructed and operated to ensure safety and demonstrate the concepts of MSRs. Part 1 is the format and content guide, which suggests a uniform format for presenting information in nonpower reactor applications acceptable to NRC [113]. Part 2 ensures the quality and uniformity of the staff review of an application [113]. While these are helpful, they do not provide adequate guidance for all of the advanced non-LWR technologies [114]. In 1996, Interim Staff Guidance (ISG) was added to address aqueous heterogeneous reactors, and in 2012, an ISG was developed to address licensing criteria for AHRs, updated criteria for aqueous heterogeneous nonpower reactors, and licensing criteria for a Part 50-licensed isotope production facility. Another ISG is needed to preparation and review of other nonpower MSRs [114]. NUREG-1537 covers all aspects of non-power reactor licensing with no General Design Criterias (GDCs) or Probability Risk Assessment (PRA) [115]. Licensing of advanced reactors is focused on adapting the current NRC LWR-based process [116]. NRC has recognized some of the unique passively safe aspects of advanced non-LWR reactors and has introduced an advanced reactor strategic plan that will address the policy issues needed to accommodate advanced

reactor concepts within the existing regulatory framework [116]. Applying performance-based, risk-informed regulatory processes is vital to substantially decreasing the cost, time, and uncertainty of commercial MSR licensing while providing for the health and safety of the public and protecting the environment [116]. The following sections contain some changes that will need to be made to different chapters of NUREG-1537.

7.1.1 Chapter 4: "Reactor Description"

Chapter 4's most significant change is references to heterogeneous fuel elements with fuel cladding acting as the initial fission product barrier since a MSR has homogeneous fuel with no cladding [114]. For a MSR, the initial fission product barrier is the fuel system boundary [114].

7.1.2 Chapter 5: "Reactor Coolant Systems"

Since the fuel salt dissipates heat through a heat exchanger to cooling systems void of fuel, the LWR based primary coolant system does not exist [114].

7.1.3 Chapter 6: "Engineered Safety Features"

The discussion in this chapter in regards to confinement and containment is generally applicable to all nonpower reactor designs [114]. There is a proposed adaptation to account for the multiple confinement or containment boundaries that might be included for MSRs depending on the relative location of the gas management and other cleanup systems to the fuel system boundary [114].

7.1.4 Chapter 9: "Auxiliary Systems"

Chapter 9 must include the homogenous fuel handling and storage of Spent Nuclear Material (SNM) used for reactor fuels, including systems, processes, criticality monitoring, vaults, shielding, and contamination control [114]. The form of the fuel during handling and storage should be addressed in a revision to this section [114].

7.1.5 Chapter 11: "Radiation Protection Program and Waste Management"

Chapter 11 will need to be changed significantly for a MSR. In a LWR, fission products get released to the fuel rod gap space where it is contained by the fuel clad, but in a MSR, fission products are released into the fuel salt solution and contained by the fuel barrier [114]. The gaseous fission products from MSRs will go directly into the gas space in the top of the vessel and enter the gas management system to be processed within the fission product barrier [114].

7.1.6 Chapter 13 "Accident Analyses"

A preliminary gap analysis will be necessary to comply with the requirements within chapter 13. These analyses are available for the MSRE, but this is not applicable to all the different designs and much has changed since then. Design information for the current reactor concepts are being held close to the belt by the vendors and companies developing them.

7.2 NUREG-0800

NUREG-0800, Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants, provides Standard Review Plan (SRP) guidance for large LWRs. This is relevant since the ultimate goal of all the MSR R&D and the test reactor is commercial deployment. This guidance laid out in NUREG-0800 when applied to nonpower reactors is very cumbersome according to NRC staff; this is because of the great differences in complexity and hazards between nonpower reactors and nuclear power plants [114]. The SRP is cumbersome to apply to liquid-fueled MSR technologies making a thoughtful licensing approach necessary for MSRs [116]. In [116], a regulatory gap analysis is performed for select chapters from the LWR SRP for a generic nonpower MSR program [116]. A successful nonpower MSR test program should provide the information needed for a review for commercial MSR eventually. The LWR SRP chapters dealing with the reactor, reactor

coolant, engineered safety feature, auxiliary system, and waste management systems were reviewed:

- Chapter 4, "Reactor"
- Chapter 5, "Reactor Coolant System and Connected Systems"
- Chapter 6, "Engineered Safety Features"
- Chapter 9, "Auxiliary Systems"
- Chapter 11, "Radioactive Waste Management" [116].

The remaining chapters, besides Chapter 15, "Transient and Accident Analysis," were not expected to differ significantly in their applicability to an MSR design and were not reviewed [116]. A specific reactor design will be needed for this chapter.

7.2.1 Chapter 4: "Reactor"

Chapter 4 provides the review criteria for the neutronics, the thermal-hydraulics, and the fuel [116]. Due to the fundamental differences between LWRs and MSRs, the current review criteria will be hard to apply to MSRs. LWRs retain all the fission products within the fuel rods, whereas MSR fuel is majorly different and is expected to move into areas outside the core. Section 4.2 focuses on accident analyses that should be evaluated on the thermal, mechanical, and materials design for the fuel system of LWRs and will need to be changed significantly for MSRs [116]. For MSRs, the review switches to chemical evaluation of fuel salt versus the current focus on structural components. There is a focus on specified acceptable fuel design limits in order to demonstrate that LWR fuel is not damaged, whereas this will be a chemistry exercise for MSR fuel [116]. This is why knowing the chemical parameters, as discussed in chapter 4 of this thesis, is vital to MSR licensing. Section 4.4 deals with thermal and hydraulic design and focus will need to be put on acceptable analytical methods to confirm that the core and reactor coolant system are safe.

7.2.2 Chapter 5: "Reactor Coolant System and Connected Systems"

Chapter 5 has two main focuses. The first is to ensure that the reactor coolant pressure boundary will stay intact for continuous core heat removal. The second focus is on evaluation of the interaction of interconnected systems to ensure that the reactor coolant pressure boundary will stay intact [116]. Both are directly concerned with the reactor coolant pressure boundary. Section 5.2.2 deals with overpressure protection. Since [MSRs](#) operate at low pressure, the overpressure concern is not important. in [LWRs](#) however, it must be mitigated to prevent a catastrophic failure of the reactor coolant pressure boundary [116]. Section 5.4, which deals with the components of the reactor coolant system, will need to have some new components added, such as the gas management system, fuel salt drain tank, fuel salt pump, piping in the system containing fuel salt, fuel salt makeup and sampling system, and the primary heat exchanger [116].

7.2.3 Chapter 6: "Engineered Safety Features"

The Chapter 6 sections and subsections have many that do not apply for [MSRs](#). The focus of Chapter 6 is to make sure that the plant safety features are provided to mitigate design-basis accident consequences [116]. Section 6.3, which deals with the emergency core cooling system will need to be different for [MSRs](#) since [MSRs](#) might not have an entirely separate system dedicated to this task. For [LWRs](#) the focus is on preventing fuel and clad damage that could lead to insufficient cooling, whereas for [MSRs](#), it is the integrity of the fuel salt system boundary [116]. For Section 6.4, the control room habitability system, it is largely applicable for [MSRs](#) as well [116]. Section 6.5, which deals with containment supports systems, will largely not be applicable to [MSRs](#) as it stands right now. The atmospheric cleanup systems section is [LWR](#) specific, and will need to be adapted since the source terms will differ for [MSRs](#) which will dictate containment leakage rate limits [116].

7.2.4 Chapter 9: "Auxiliary Systems"

A lot of Section 9.2 is not applicable since the ways water are used is significantly different between [MSRs](#) and [LWRs](#). Section 9.1 refers to fuel handling of heterogeneous fuel and will need substantial modification to be applicable to criticality safety and fuel storage of homogeneous fuel [116]. A couple new subsections focusing on safeguards and fuel salt loading handling system should be added [116]. Section 9.2 focuses on cooling water and water-based systems for [LWRs](#) and will most likely not be applicable to [MSRs](#) since water is unlikely to be used as a cooling fluid [116]. Some new subsections will be needed for [MSR](#) cooling fluid for cooling systems in the reactor auxiliary, fuel salt storage, gas management, fuel salt drain tank, and primary coolant drain tank [116]. Section 9.3 focuses on sampling which will need to add sampling requirements for [MSRs](#) since the current section is [LWR](#)-focused [116]. Some new subsections should be added including ones for the [MSR](#) fuel salt heating system and the cover gas cleanup and purification system [116]. Section 9.4 is focused on ventilation systems and is applicable to [MSRs](#), but may need more source term information included. Section 9.5, fire, communications, lighting, and diesel support systems are technology neutral and sufficient for [MSRs](#) [116].

7.2.5 Chapter 11: "Radioactive Waste Management"

Much of Chapter 11 is generally applicable to [MSRs](#) in all areas [116]. In [LWRs](#), the coolant source term must be evaluated so that the appropriate waste system design features to process, treat, release, or recycle liquid and gaseous effluents can be developed [116]. There will need to be a substantial rewrite for section 11.1 of NUREG-0800, since the [LWR](#) and [MSR](#) coolant source term will be much different. The liquid, gaseous, and solid waste management systems review text indicates that the system used to manage and treat process and effluent streams must be evaluated to ensure compliance with regulatory limits for liquid effluent discharges and associated doses to members of the public [116]. For a [MSR](#), a liquid waste management system may not be necessary since it is likely the fuel salt will stay onsite for reuse. Section 11.2 needs to be retained for a review of individual [MSR](#) technologies even though it may not be applicable to most [MSR](#) designs [116]. For a [LWR](#),

the gaseous waste management system is categorized as nonsafety, whereas in a [MSR](#), it will be categorized as a safety-related system. In an [MSR](#), the gaseous waste management system will be what governs the regulations for removing fission products that are gases, such as Xe and Kr, and will need to take place inside containment. The section that deals with gaseous waste management, section 11.3, will require a rewrite [116]. [MSR](#) and [LWR](#) solid waste management system will be similar in review aspects. The solid waste management system section, with the exclusion of dewatering solid waste, will be applicable to [MSRs](#) as written. In [LWRs](#), the Process and Effluent Radiological Monitoring Instrumentation and Sampling System ([PERMISS](#)) is used to monitor gaseous and liquid process streams and effluents from the liquid, gas, and solid waste management systems [116]. [PERMISS](#) for a [MSR](#) will have a similar waste management role, but some rewrite of section 11.5 may be needed.

Chapter 8

Source Term Analysis

8.1 Intro to Analysis

In this analysis, several different [MSR](#) models will be run in SCALE, specifically run in ORIGEN/ChemTriton. One model is similar to the [MSDR](#), one model is the [MSRE](#), and the last is a 17x17 Westinghouse [PWR](#) for comparison. The parameters of interest in this analysis are the concentrations of different nuclides in an [MSR](#) specifically with comparison to concentrations of these same nuclides in a [PWR](#). A comparison of actinide production for the [MSDR](#) and [MSRE](#) model was performed. In addition to this, the ratio of [MSR](#) nuclide concentrations to [PWR](#) nuclide concentrations was calculated. Burnup is typically different for [MSR](#) and [PWR](#), as well is the burnup can differ greatly between different [MSR](#) models. The burnup marker of Cs-137 was compared for [MSR](#) and [PWR](#) in order to see how much of it and other fission products are in each system with varying burnup. Some [MSR](#) designs generate more tritium than [PWRs](#) so it was given special attention. The radionuclide concentrations have very different magnitudes so multiple plots were required to accurately display them.

Putting all this information together results in being closer to determining a source term, but more research will still need to be done. The source term needs to take into account the chemistry and species in the salt under different accident scenarios.

8.2 Modeling and Simulation

The [MSR](#) model used in this simulation is similar to the [MSDR](#) design except it used 5% LEU instead of thorium as the fuel and only used lithium in the salt composition instead of lithium and beryllium. The initial salt composition consists of Li-7, F-19, U-235, U-238, and a small quantity of Li-6. This design is a thermal neutron spectrum [MSR](#) that employs a graphite moderator. There was minimal salt conditioning done; the gaseous fission products were removed and solid fission products were assumed to plate-out on the surfaces of the reactor; the specific elements can be seen in Table [8.1](#). There was also a continuous feed of 5% enriched uranium in the model. The temperature associated with the model is 625°C. It is assumed to be 750 MW_{th} with 138.54 metric tonnes of salt and 62.1 metric tonne of heavy metal. The power is 6.197 MW per metric tonne of heavy metal. This model was run in ORIGEN/ChemTriton. The [MSDR](#) model is compared against the [LWR](#) model used, which is a 17x17 Westinghouse [PWR](#). A model of the [MSRE](#) was also used for comparison in this simulation. Its initial fuel salt composition consists of Li-7, Be-9, F-19, Zr-90, U-235, and U-238. It is a thermal spectrum [MSR](#) with a graphite moderator. The elements that are removed are the same as the [MSDR](#) model seen in Table [8.1](#). 35% enriched uranium was continuously feed into the salt. The temperature associated with the [MSRE](#) model is about 650°C, and the power is 36.39 MW per metric tonne of heavy metal.

Table 8.1: Isotopes Associated with the Removal Processes in the Model

State of Fission Products	Isotopes
Gaseous Fission Products	Kr, Xe, Ar, H, N, O
Solid Fission Products	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te

Table 8.2: Chemical Specifications for MSRE Fuel Salt. Adapted from [61]

Impurity	Allowable Concentration (wt %)
Water	0.1
Cu	0.005
Fe	0.01
Ni	0.0025
S	0.025
Cr	0.0025
Al	0.015
Si	0.01
B	0.0005
Na	0.05
Ca	0.01
Mg	0.01
K	0.01
Li (natural)	0.005
Zr (natural)	0.025
Cd	0.001
Rare earths (total)	0.001

8.3 Time Effect on Source Term

8.3.1 Initial Loading

The components in the salt at the beginning of life are the elements of the salt, the fuel used (U, Pu, and/or Th), and trace amount of other materials. the amount of contaminants allowed for MSRE can be seen in Table 8.2. In the MSRE, zirconium was added as a constituent to prevent precipitation of UO_2 and resultant criticality hazards in the event that oxide contamination of the fuel occurs [61]. Specifically, the amount of excess oxygen needs to be as low as possible since salts containing excess oxygen are much more corrosive [33].

8.3.2 During Operations

In an [MSR](#), the source term can be everywhere: in the salt, in the processing system, lost to the containment area, and/or absorbed or adhered to surfaces in the reactor. Some of these areas include

- Reactor Core/Vessel
- Primary Heat Exchanger
- Off-gas System
- Piping
- Drain Tank
- Pump
- Fuel pump
- Reprocessing Area (if there is one)

The three main systems that contain source terms are typically the off-gas system, fuel salt system, and salt processing and handling. Also in [MSRs](#), trace fissile material accumulation could eventually become significant. In [MSRs](#), the source term is quite different due to delayed neutrons being emitted outside the core and due to the composition of the salt changing.

8.3.3 End of Life

The amount of processing performed on the system during operation will have a huge effect on the end of life source term since if a lot of reprocessing is done, there will not be as much worrisome material released in the event of an accident. An offgas sorbent, such as charcoal beds like what were used in the [MSRE](#), could be used as the end-of-life disposal medium for the fission gas [33].

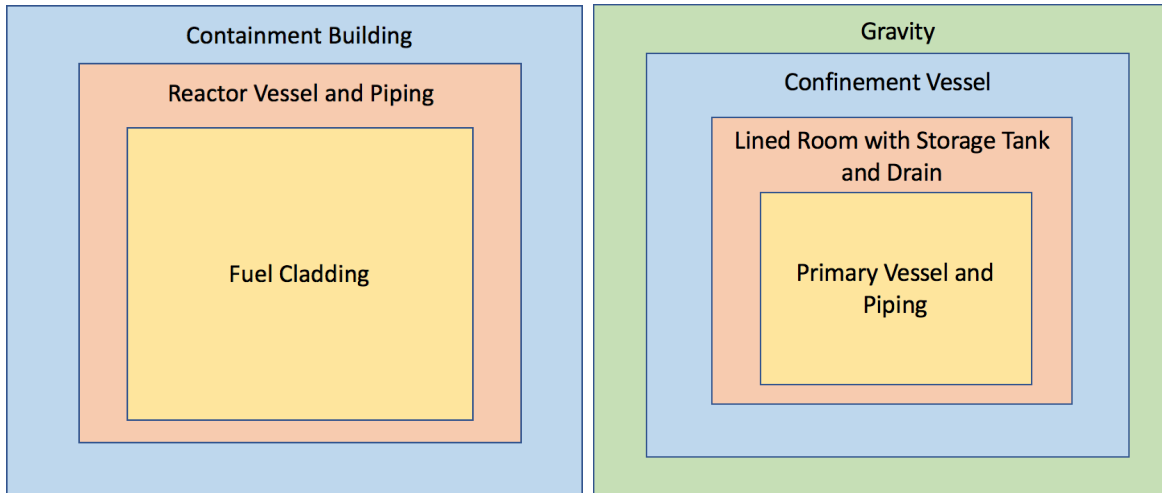


Figure 8.1: A view of the differences between barriers to release for [PWR](#) and [MSR](#). [PWR](#) is on the left and [MSR](#) is on the right.

8.4 Other Considerations for Source Term Analysis

The defense in depth is comparable for [LWRs](#) and [MSRs](#). For [LWRs](#), the first barrier is the fuel cladding, the second is the reactor vessel and the piping for the cooling water system, and the third is the containment building [11]. For [MSRs](#), the first barrier is the same as the [LWR](#) secondary barrier of the primary vessel and piping, the second is a lined room with a storage tank and drain, the third is a confinement vessel similar to the containment building for [LWRs](#), and the fourth is a gravity barrier that results from the reactor being underground; [MSRs](#) have an extra barrier for their defense in depth strategy, with gravity adding an extra step to protect against an environmental release [11]. Since there is no driving force due to gravity, the salt will be contained underground and is less likely to be released in the event of an accident since the radionuclides would have to move upward into the air, which is rarely possible. In [MSRs](#), the outermost containment layer acts as a radiation barrier and does not provide mitigation for high pressure. A schematic of the barriers can be seen in Figure 8.1.

8.5 Possible Accidents

Many accident scenarios for traditional LWRs may not be applicable for MSRs. Some of the main differences between LWRs and MSRs are source terms outside of the core, passive safety systems in most MSRs, and the principal contributor to dose may not be reactor core accidents. The likelihood of energetic events and phase changes is reduced since it is a low pressure system [117]. Hazards analysis was done for the MSRE. From this, the most probable accident was found to be a small leak into the secondary container and the maximum credible accident was found to be a break in the drain line [117]. This was done using a barrier approach instead of a component failure approach typical for LWRs.

Some initiating events for MSRE according to Chisholm and Krahn include:

- Fuel salt pump failure
- Coolant salt pump failure
- Uncontrolled rod withdrawal
- Change in fuel salt concentration in core due to precipitation
- Leakage from freeze valve or freeze flange
- Increase or decrease in heat removal by coolant system
- Decrease in fuel salt flow rate
- Reactivity and power distribution anomalies
- Leakage of substance through the first barrier
- Decrease in fuel salt inventory
- Radioactive release from a subsystem or component [118].

8.6 Results

Burnup is an important consideration for reactors; this model of [MSR](#) has a lower power and thus lower burnup than the Westinghouse [PWR](#).

$$Burnup = \int_0^\tau \frac{Power(t)}{X_{initial}} dt = \int_0^\tau SP(t) dt \quad (8.1)$$

Equation 8.1 shows the typical traditional way to measure burnup where SP is specific power, but [MSRs](#) need to account for the feed rate and removal rate as well if material is being feed into and removed from the [MSR](#). Equation 8.2 shows how feed rate is included.

$$Burnup = \int_0^\tau SP(t) dt * \frac{X_{initial}}{X_{initial} + \int_0^\tau \dot{f}_{feed} dt - \int_0^\tau \dot{r}_{removed} dt} \quad (8.2)$$

An important marker for burnup is Cs-137; a plot of the comparison between [MSR](#) and [LWR](#) can be seen in Figure 8.2. Looking at 50 GWd/MTHM on Figure 8.2, which is the typical [PWR](#) burnup, it can be seen that employing removal of even a small number of elements results in a reduction of elements that are not removed, thus lowering the source term for an [MSR](#); note that Cs-137 was not removed from this system. As can be seen in Figure 8.3 and in Figure 8.4, most of the radionuclides of concern reach a steady state value at which they level out or slowly decrease. This is true for all but Sr-90, I-129, and Cs-137 which continue to build up over time in this [MSR](#) design, as can be seen in the top of Figure 8.3. In a [PWR](#), these same isotopes also build up over time as can be seen in Figure 8.5, as well as Sr-89, Ru-106 and Tc-99. Figure 8.6 shows isotopes with lower concentrations compared to Figure 8.5 for a [PWR](#); it can be seen that most of these isotopes level out quickly typically within the first 6 months of operation. When comparing the graphs for [PWR](#) and [MSR](#), it can be seen that the magnitude of the concentration is larger for a [PWR](#) even though it is only run for 5 years versus 15 years for the [MSR](#). The [PWR](#) does have a higher burnup than the [MSR](#), but as can be seen in Figure 8.2, which compares burnup, the inventory of the [MSR](#) after 15 years is only slightly higher than the inventory for the [PWR](#) after 5 years. Note that this result is without employing processing to remove Cs-137 from the salt. Figure 8.7 shows the ratio of the concentration of different isotopes for [MSR](#) versus [PWR](#) at the

same burnup. In all of the analyses, the ratio is always less than one, meaning that there will always be less of a specific isotope for this [MSR](#) design versus this [PWR](#) design. Both Kr-85 and Xe-133 are removed from the system reducing the concentration and source term of each found in the [MSDR](#) model at any given point in time. Figure 8.8 shows that the [MSDR](#) model has much less tritium in the system at any given point in time. This is due mostly to the [MSRE](#) fuel salt containing beryllium and also the amount of removal processes employed. Furthermore, in Figure 8.9, it can be seen that the [MSRE](#) produces more tritium than the [MSDR](#) further showing the difference adding beryllium to the fuel salt makes in terms of tritium production. In Figure 8.10, the amount of transuranics produced is shown; the [MSRE](#) produces more, but it is not a huge change and can mostly be attributed to the fuel being 30% enriched uranium versus 5% LEU.

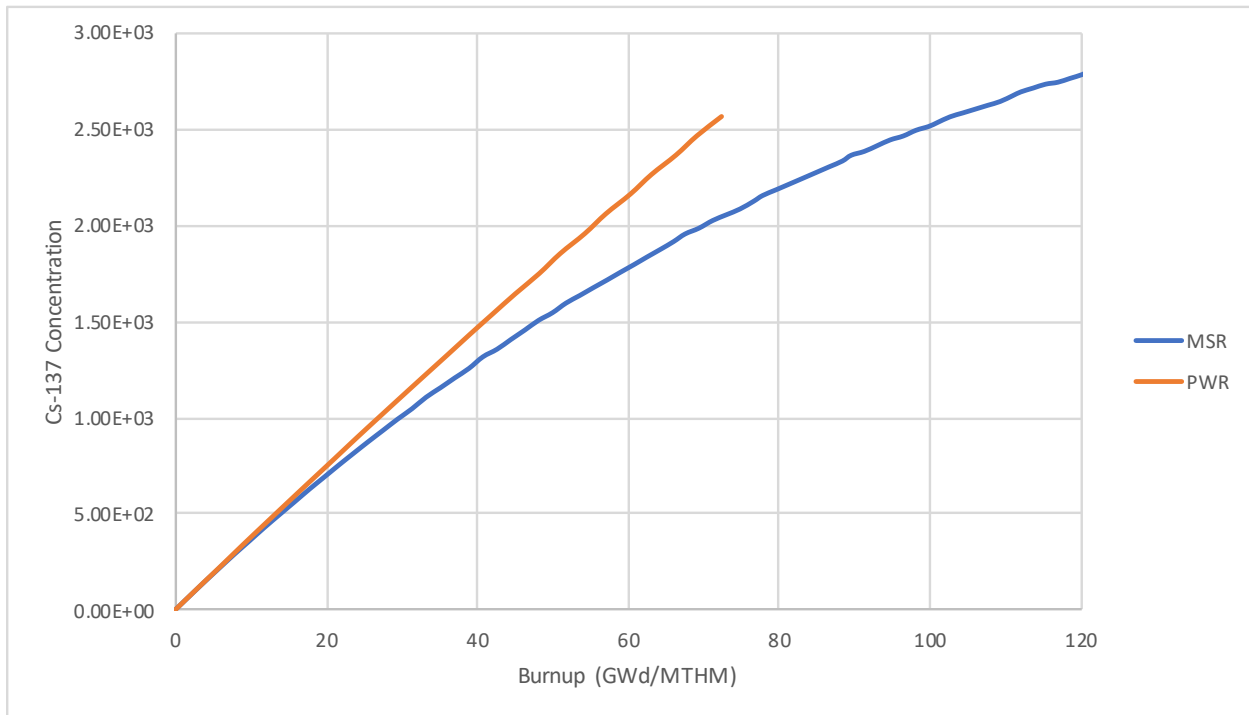


Figure 8.2: Comparison of burnup for [MSR](#) and [PWR](#) using the Cs-137 concentration, an important burnup marker

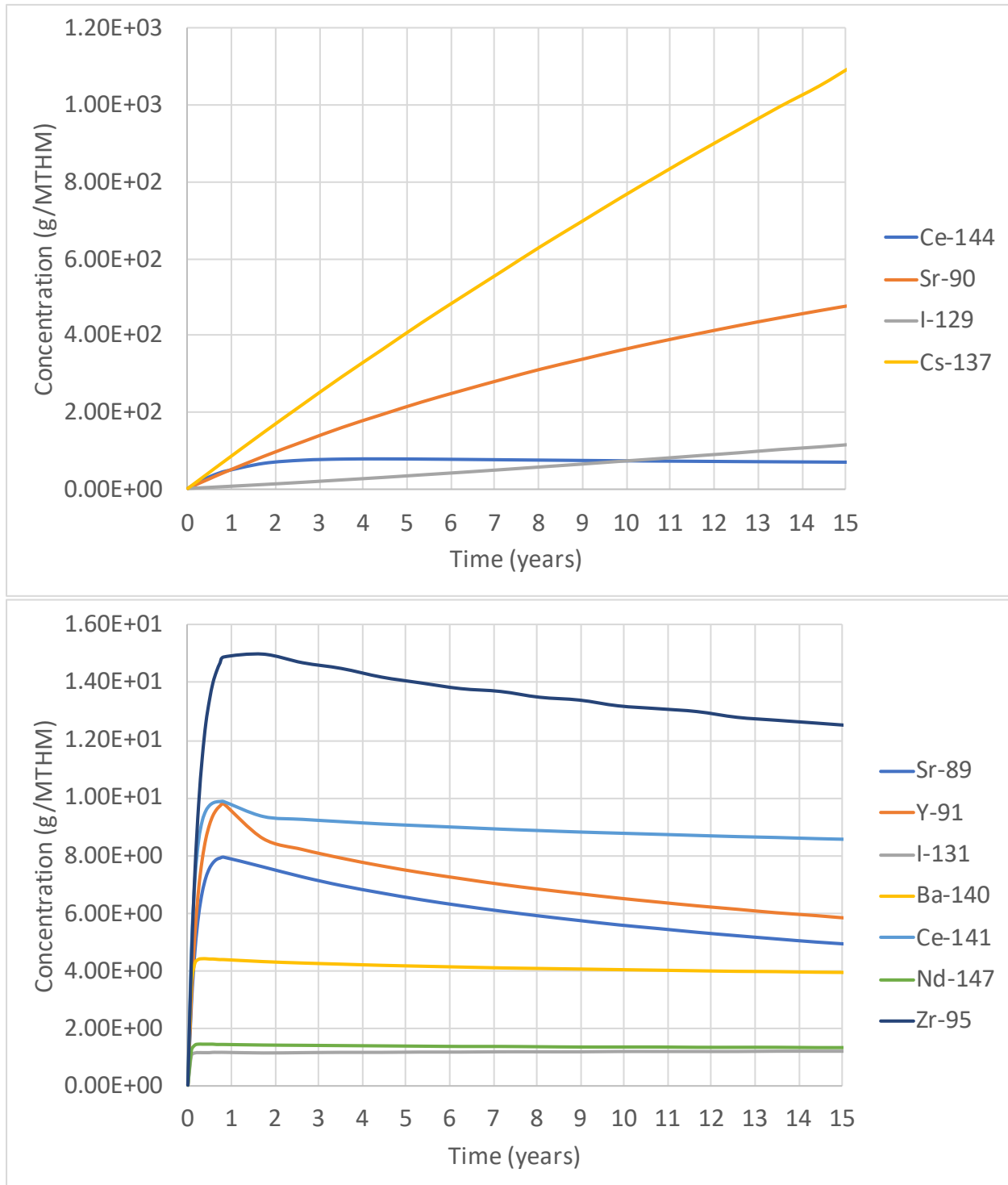


Figure 8.3: Buildup of nuclides with higher concentrations over a 15 year time span for MSR

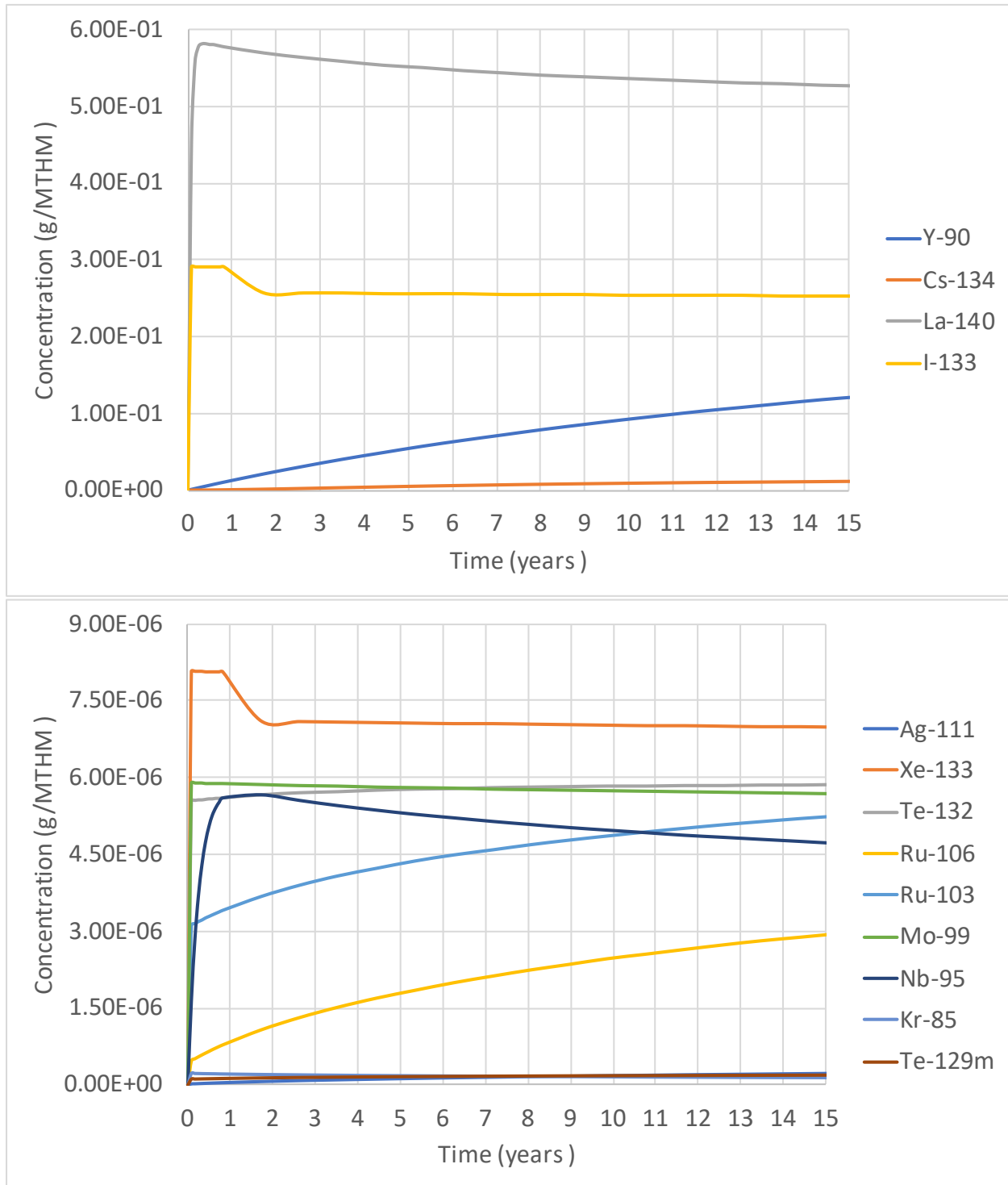


Figure 8.4: Buildup of nuclides with lower concentrations over a 15 year time span for MSR

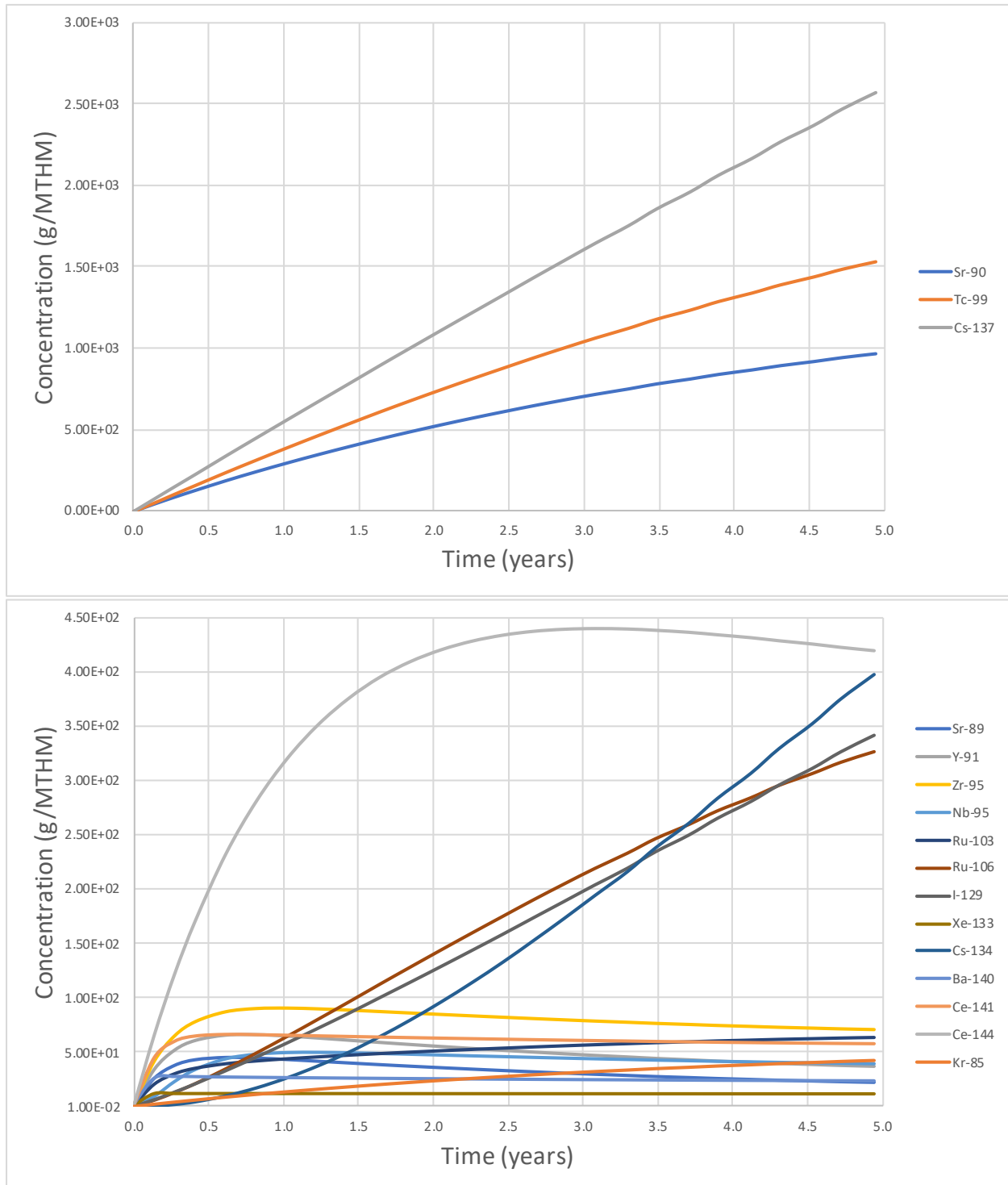


Figure 8.5: Buildup of nuclides with higher concentrations over a 5 year time span for PWR

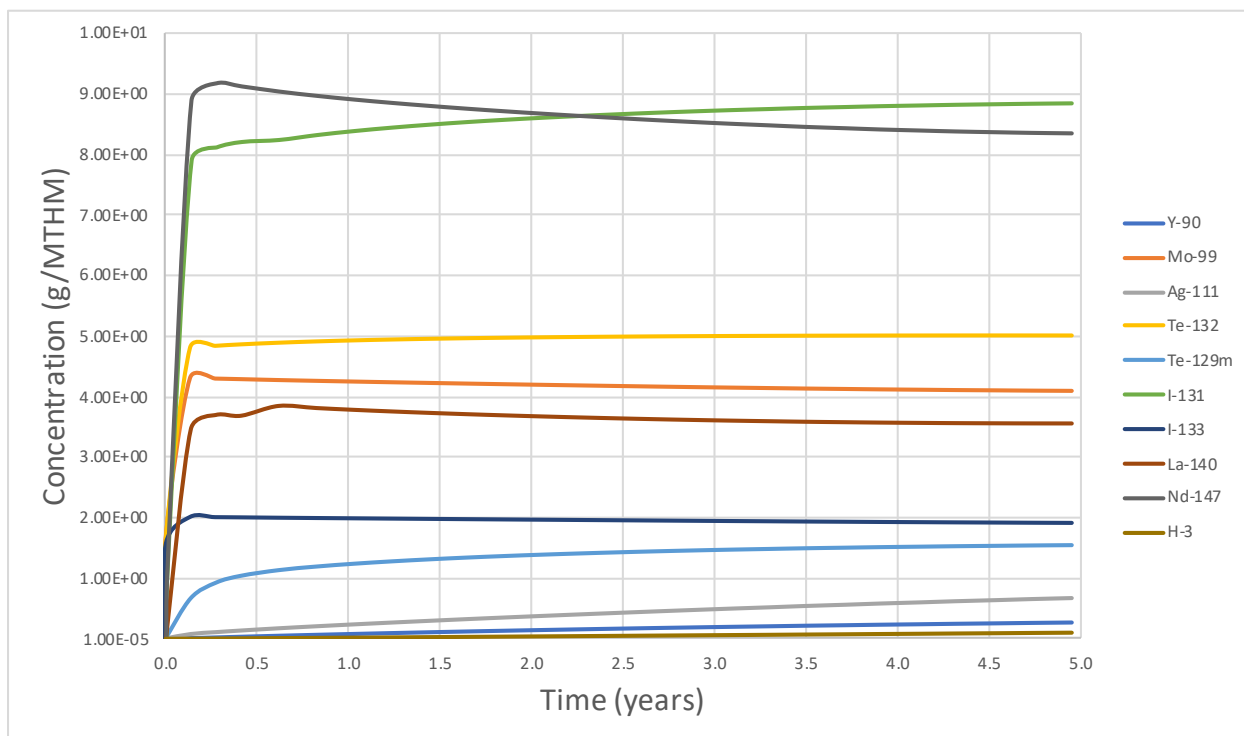


Figure 8.6: Buildup of nuclides with lower concentrations over a 5 year time span for [PWR](#)

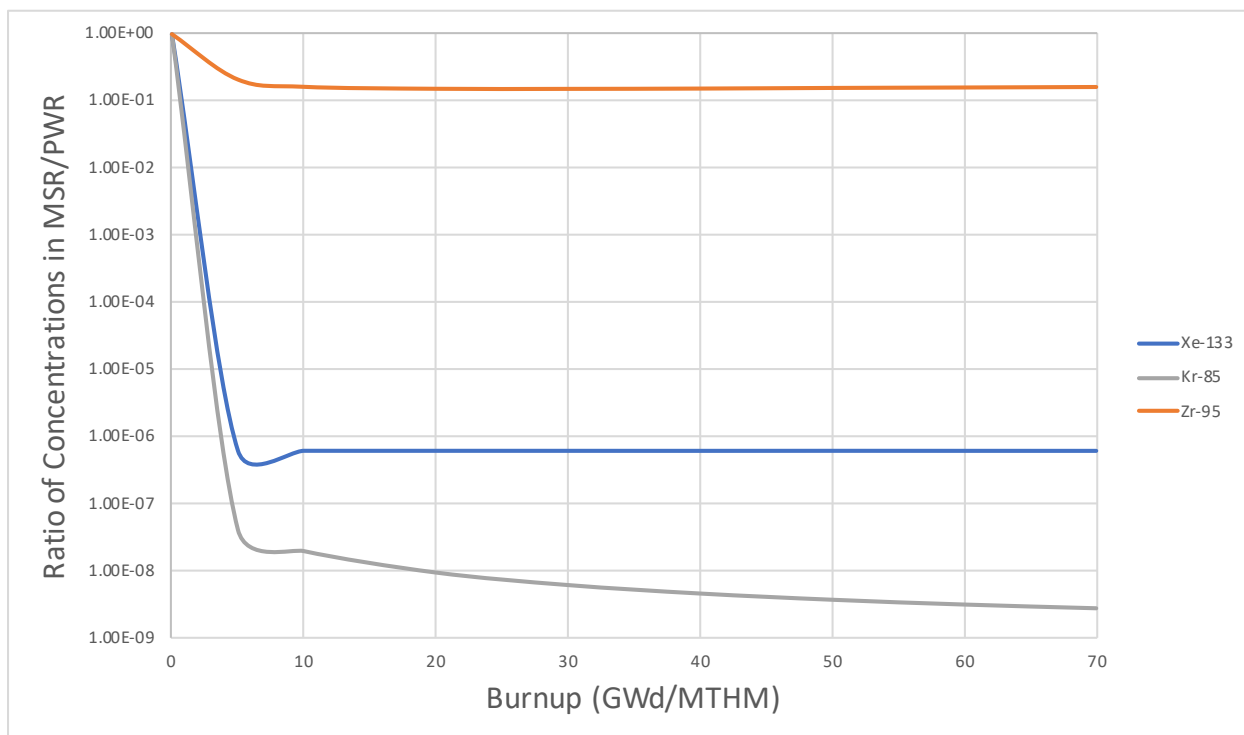


Figure 8.7: The ratio of concentrations of different nuclides for [MSR](#) versus [PWR](#) with the same burnup amount, which happens at a later time for [MSR](#) compared to [PWR](#)

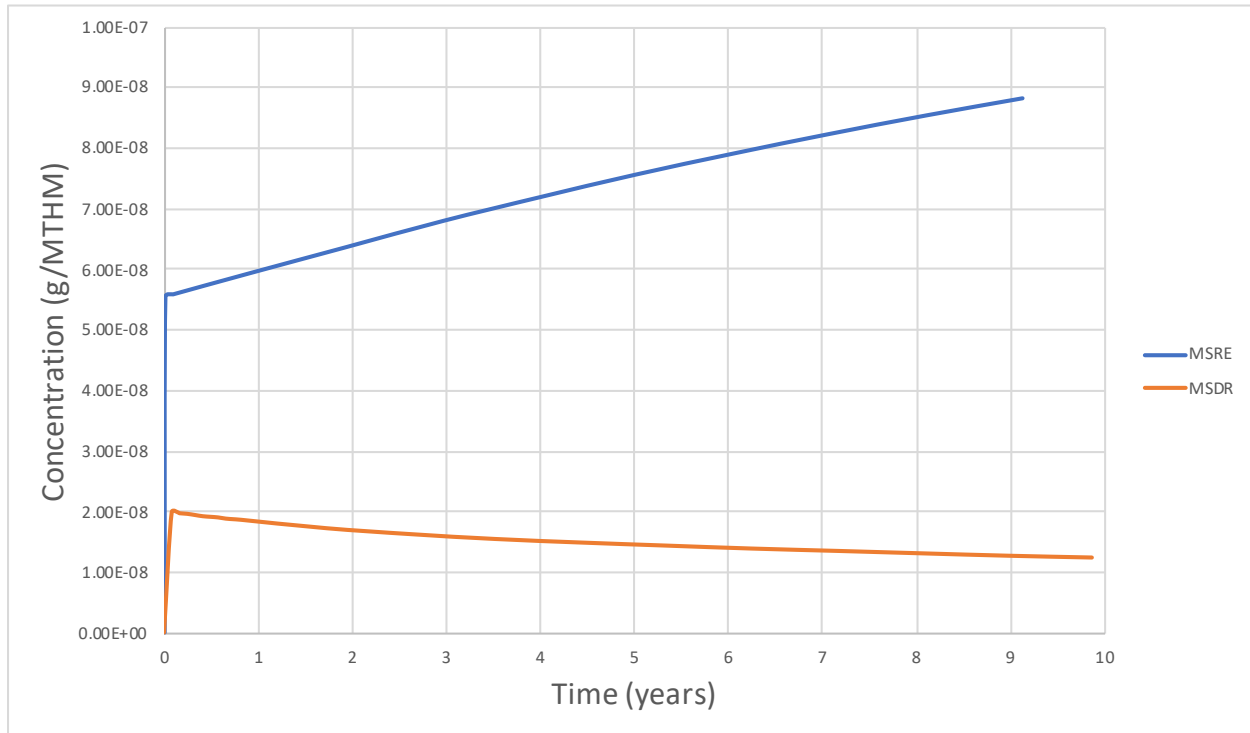


Figure 8.8: The amount of tritium in the system at any given point in time for the [MSDR](#) model and the [MSRE](#). Assume tritium removal is employed.

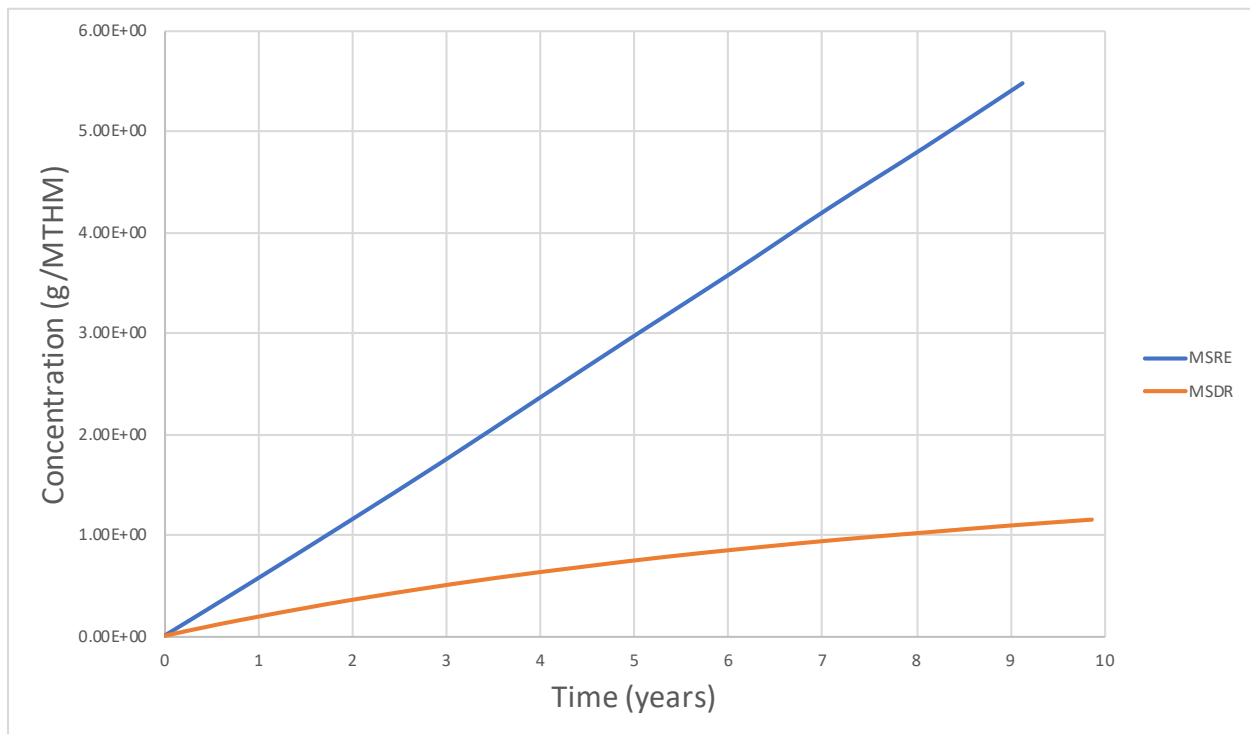


Figure 8.9: The total amount of tritium produced including tritium removed from the system for the [MSDR](#) model and the [MSRE](#)

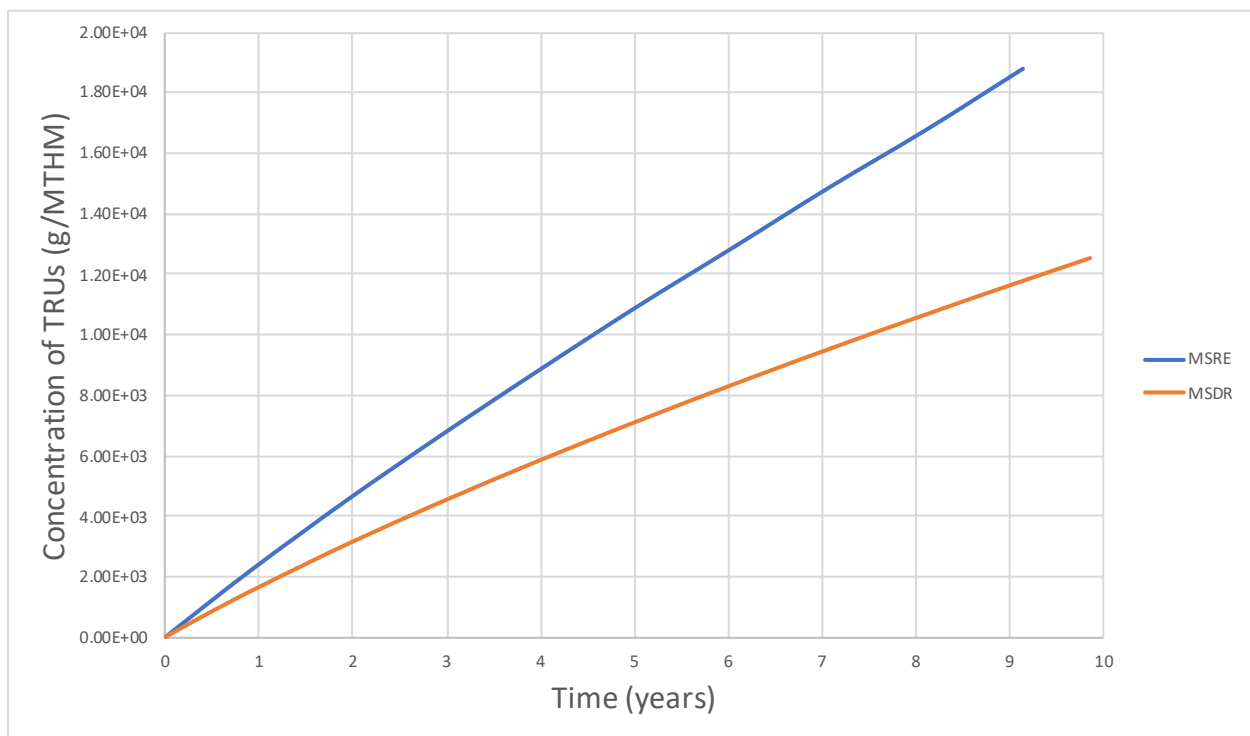


Figure 8.10: The total amount of transuranics produced in both the [MSRE](#) and [MSDR](#) model

Chapter 9

Conclusions & Future Work

[MSRs](#) have been of interest since the 1960s, and the resurgence of interest with the Gen IV Initiative is a great step towards potential commercial deployment. The [ARE](#) and [MSRE](#) provided proof of concept and vital information. A thermal spectrum, fluoride salt LEU burner using a simple salt such as LiF or LiF-BeF₂ would be the best option, in my opinion, due to there being the most info about them. A lot of the thermophysical and thermochemical properties are known for the fluoride salts, but more research must be done especially for thermal conductivity and viscosity since they are the ones with the least information. Modeling and simulation still needs some improvements mostly concerning the circulating fuel and its effects on delayed neutrons and delayed neutron precursors. Batch reprocessing with simple cleaning and polishing done between removals is the best step forward in my opinion since continuous reprocessing presents more regulatory challenges. Current regulations are still [LWR](#) centered, but they have slowly evolved and there are now regulations for some advanced reactors, such as the AP1000 and EPR. There are still a lot of changes necessary to regulatory and licensing concerns to make them relevant and encompass [MSR](#) technology and concepts. Source term first principles for [MSRs](#) shows absolute source term masses is less than 20% compared to [LWR](#) dependent of radionuclides with most actually being less than 10% even after burnup was accounted for. Any amount of reprocessing done on the fuel lowers the source term. [PWR](#) release fractions as a function of burnup are conservative compared to what is seen in the [MSR](#) analysis. This shows that [PWR](#) source term analysis is bounding and conservative; source term analysis for [MSRs](#)

and other advanced reactors and even [PWRs](#) can be further refined to be less conservative and more realistic. [MSRs](#) are a viable concept and I think a great path forward for nuclear energy.

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Appendix

Appendix A

Phase Diagrams

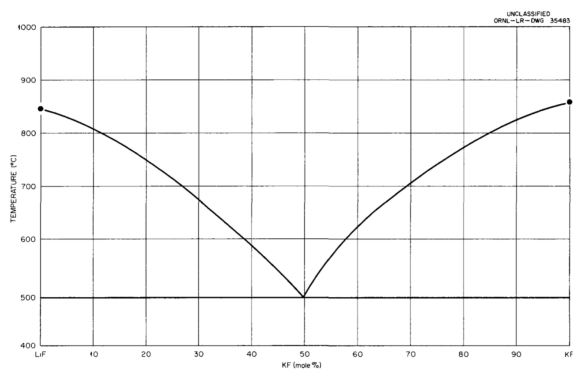


Figure A.1: Phase diagram of LiF-KF [46], [119]

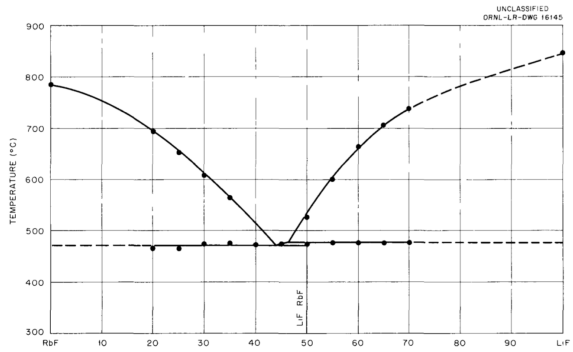


Figure A.2: Phase diagram of LiF-RbF [46]

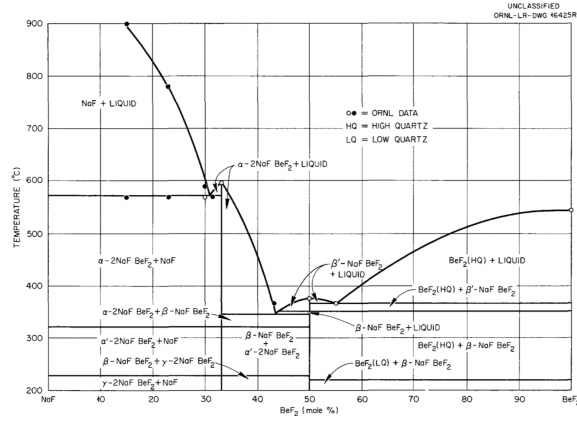


Figure A.3: Phase diagram of NaF-BeF₂ [46], [120]

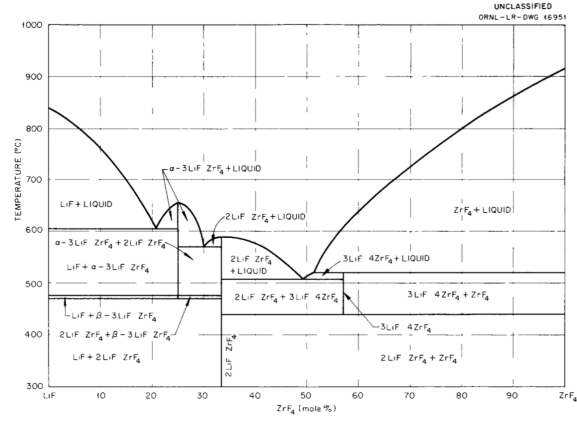


Figure A.4: Phase diagram of LiF-ZrF₄ [46]

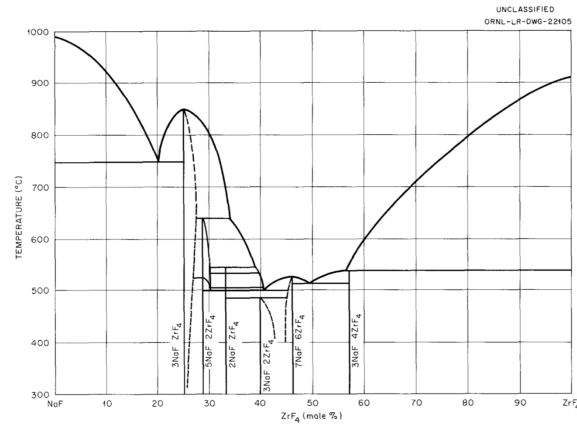


Figure A.5: Phase diagram of NaF-ZrF₄ [46], [121]

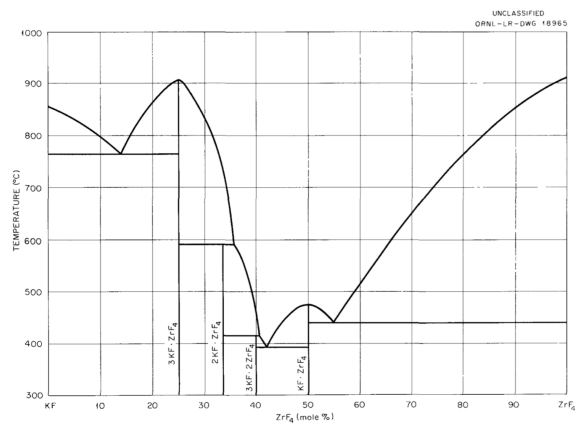


Figure A.6: Phase diagram of KF-ZrF₄ [46]

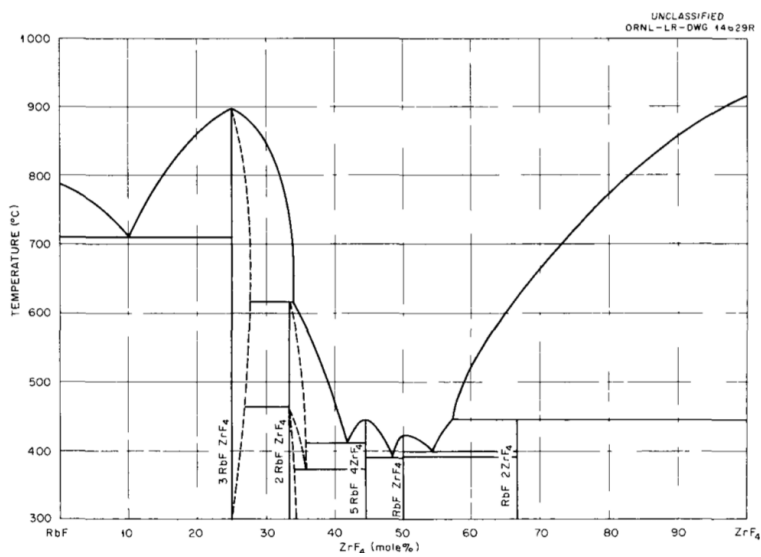


Figure A.7: Phase diagram of RbF-ZrF₄ [46]

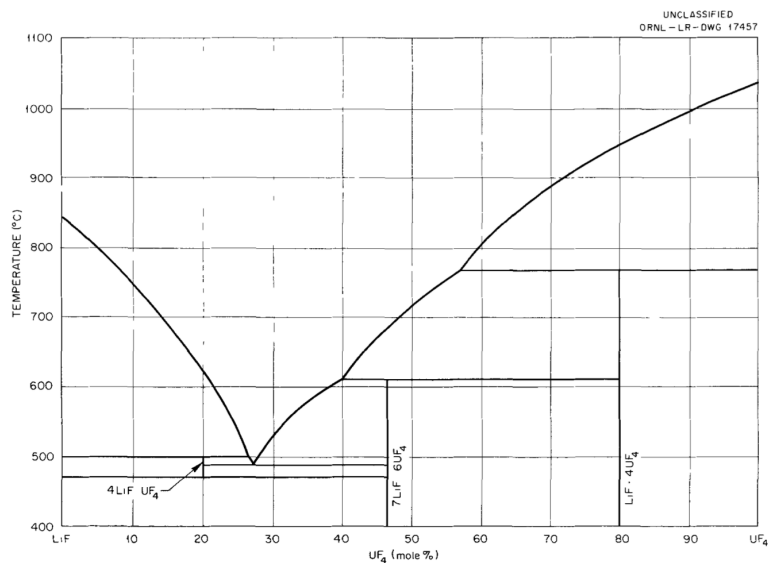


Figure A.8: Phase diagram of LiF-UF₄ [46]

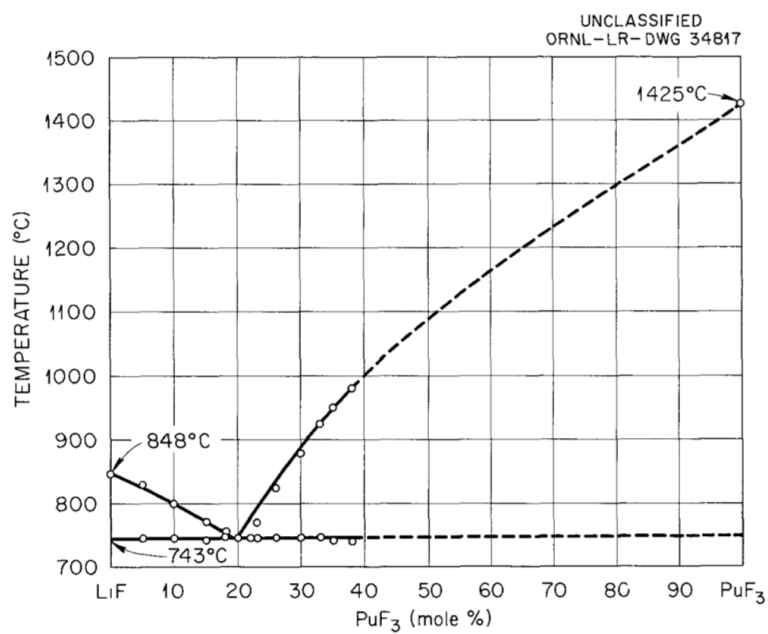


Figure A.9: Phase diagram of LiF-PuF₃ [46], [122]

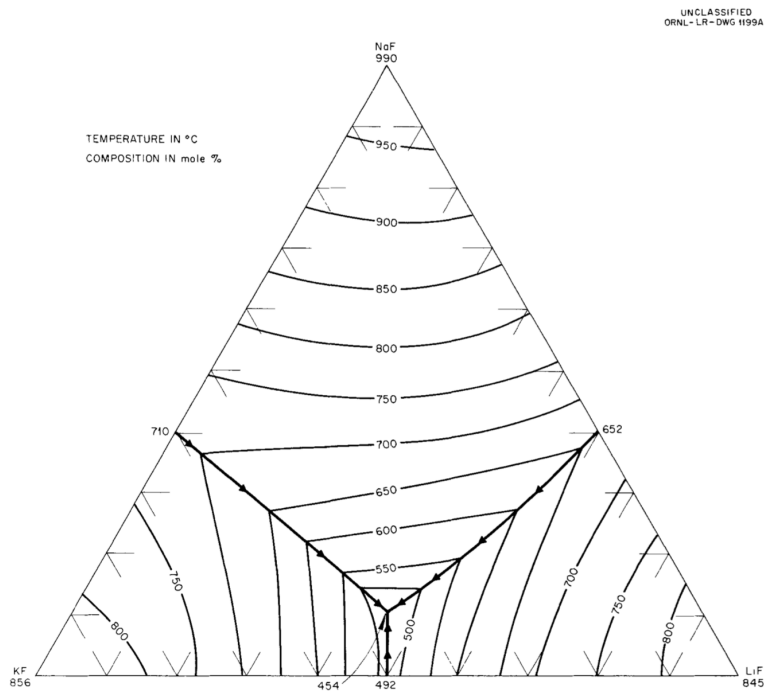


Figure A.10: Phase diagram of LiF-NaF-KF [46], [119]

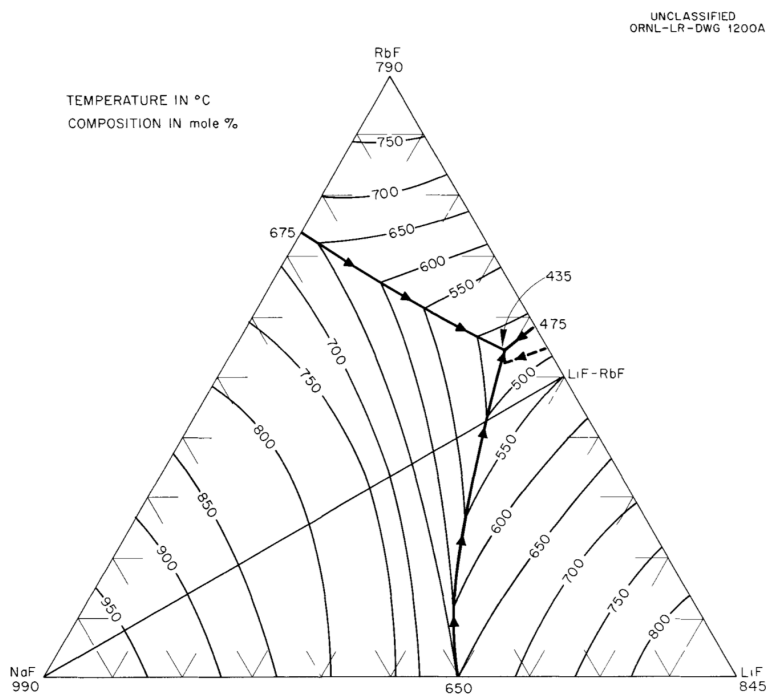


Figure A.11: Phase diagram of LiF-NaF-RbF [46]

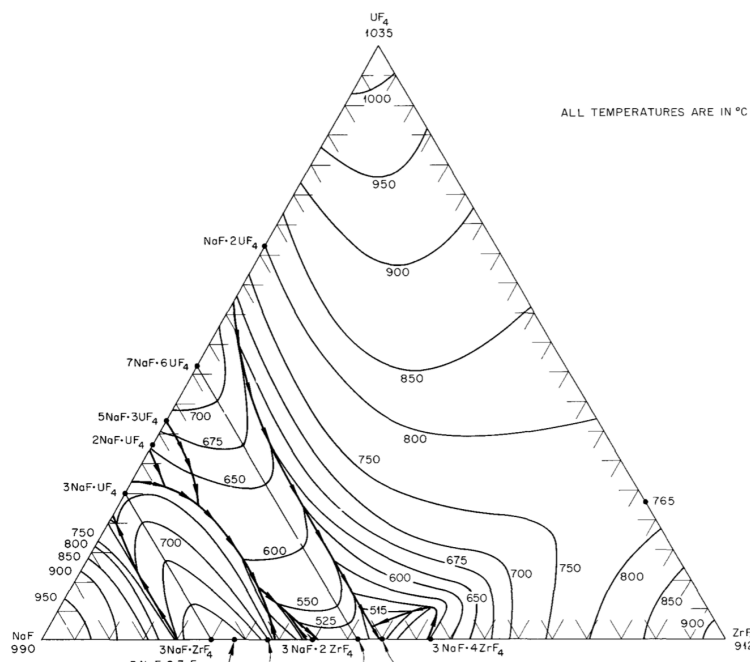


Figure A.16: Phase diagram of NaF-ZrF₄-UF₄ [46], [121]

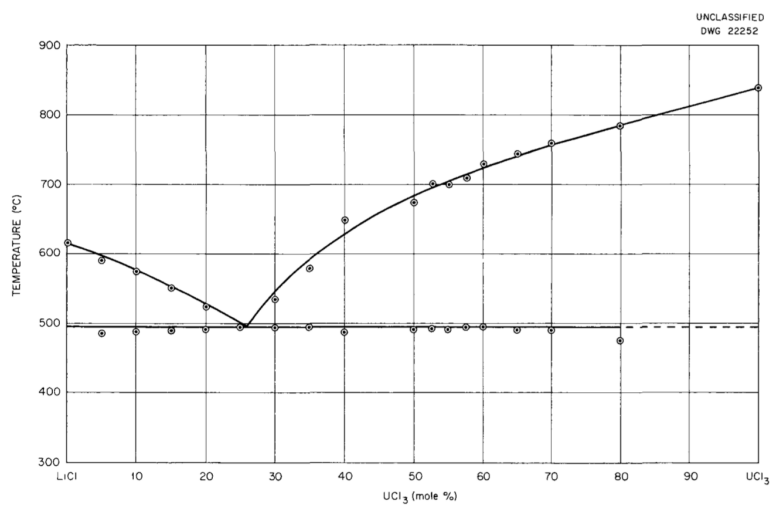


Figure A.17: Phase diagram of LiCl-UCl₃ [46]

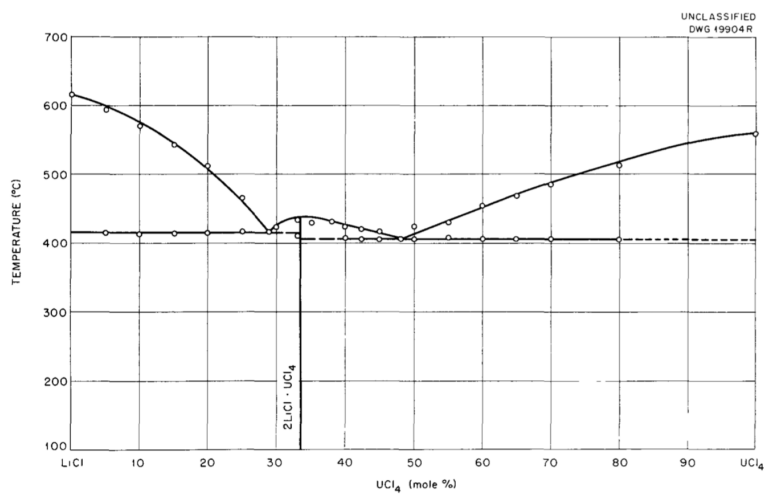


Figure A.18: Phase diagram of LiCl-UCl₄ [46]

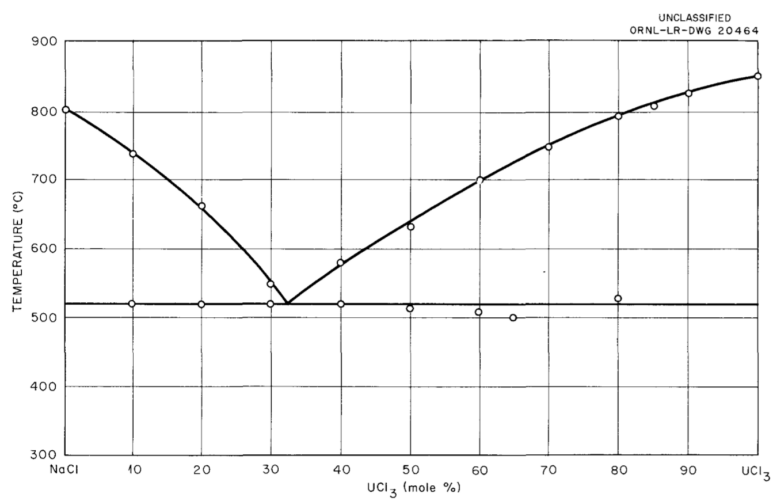


Figure A.19: Phase diagram of NaCl-UCl₃ [46], [124]

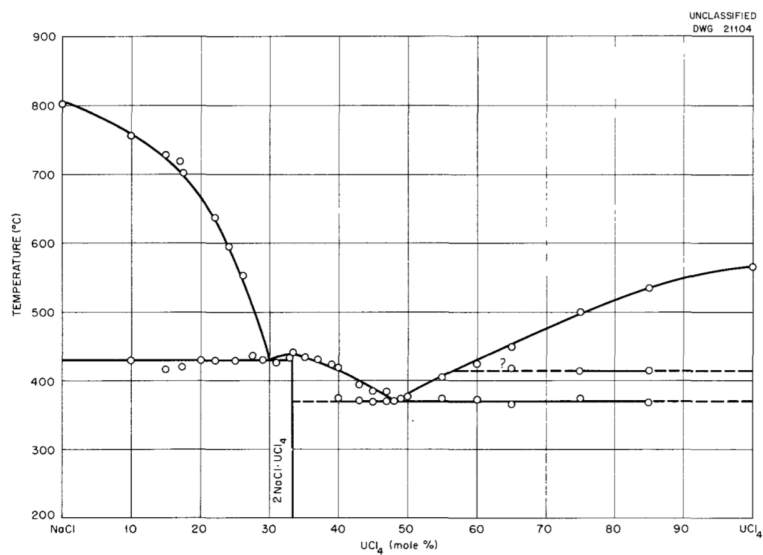


Figure A.20: Phase diagram of NaCl-UCl₄ [46]

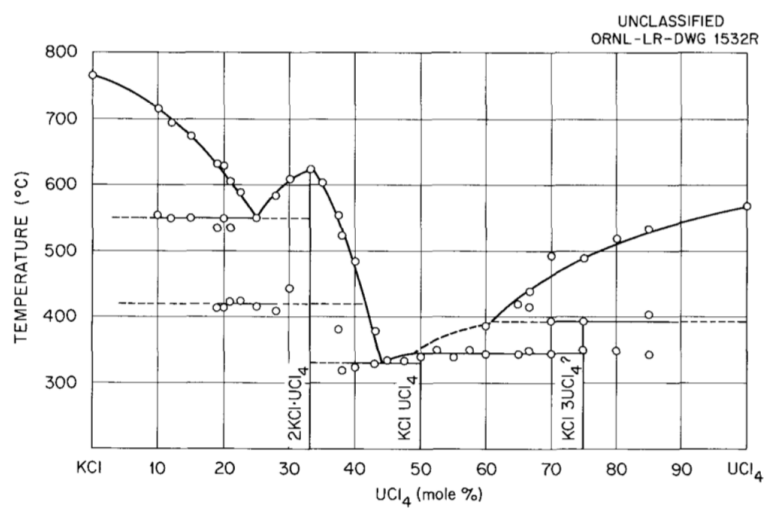


Figure A.21: Phase diagram of KCl-UCl₄ [46]

Appendix B

Modeling Inputs

A MSDR Input

=t-depl parm=(addnux=4)

MSDR assembly

v7-252

read comp

' materials at 625 degrees C

' fuel salt LiF with 5% LEU

li-6 1 1.13888E-05 898.15 end

li-7 1 2.27766E-01 898.15 end

u-235 1 1.46481E-01 898.15 end

u-238 1 2.78314E+00 898.15 end

f-19 1 1.55260E+00 898.15 end

' moderator

graphite 3 DEN=1.77666 1.0 898.15 end

' waste mixture(s)

xe-135 7 0 1.0e-20 898.15 end

' waste mixture(s)

xe-135 8 0 1.0e-20 898.15 end

```

end comp

read celldata
    latticecell symmslabcell pitch=4.29574 3 fuel=0.36068 1 end
end celldata

read depletion
    1 decayonly 7 8
end depletion

' 750 MWt for 138.54136 MT salt, 62.1 MT HM
read burndata
    power=6.196823919 burn=300 nlib=10 end
    power=6.196823919 burn=5610 nlib=17 end
    power=6.196823919 burn=5610 nlib=17 end
    power=6.196823919 burn=5610 nlib=17 end
    power=6.196823919 burn=5610 nlib=17 end
end burndata

READ opus
    matl=1 7 8 end
    title = 'Masses in fuel salt'
    units=gper nrank=2000 libtyp=all
    time=days typarms=nucl sort=no
END opus

read timetable
    ' gaseous fission product removal
    flow
    from 1

```

```

to 7
type continuous
units pers
nuclides Kr Xe Ar H N O end
constant 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 end
time 0.0 end
factor 1.0 end
end flow
' solid fission product removal
flow
from 1
to 8
type continuous
units pers
nuclides Se Nb Mo Tc Ru Rh Pd Ag Sb Te end
constant 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-
01 3.3012E-01 3.3012E-01 3.3012E-01 end
time 0.0 end
factor 1.0 end
end flow
' uranium feed
flow
to 1
type continuous
units gpers
nuclides u-238 u-235 end
constant 0.95 0.05 end
time 0.0 end
factor 8.223603303E-05 end
end flow

```

end timetable

read model

252 group solution

read parm

drawit=yes

prtmxtab=yes

end parm

read materials

mix=1 pn=1 com="fuel salt" end

mix=3 pn=2 com="graphite" end

end materials

read geom

global unit 1

polygon 10 0 0 0 11.91641 1.73101 11.91641 2.23901 11.4081275 2.23901 0 0 0

polygon 20 2.59969 0 2.59969 11.4081275 3.10769 11.91641 6.56971 11.91641 7.07771
11.4081275 7.07771 0 2.59969 0

polygon 30 7.43839 0 7.43839 11.4081275 7.94639 11.91641 11.21774607 11.91641
11.91641 11.21774607 11.91641 0 7.43839 0

polygon 40 0 0 0 12.27709 11.43381 12.27709 12.27709 11.43381 12.27709 0 0 0

cuboid 90 14.5161 0. 14.5161 0.

media 1 1 -10 -20 -30 40

media 3 1 10

media 3 1 20

media 3 1 30

media 3 1 -40 90

boundary 90 30 30

```

end geom

read bounds
    all=refl
end bounds

end model
end

```

B MSRE Input

```

=t-depl parm=(addnux=4)
MSRE Assembly
v7-56
', _____
' Mixture data
', _____

read comp
'Fuel - (2 to 1) (LEUF4+ThF4)+(2LiF+BeF2)
    u-238 1 0.069437189 922.039 end
    zr-90 1 0.227477489 922.039 end
    u-235 1 0.036917963 922.039 end
    li-7 1 0.226918919 922.039 end
    be-9 1 0.130615527 922.039 end
    f-19 1 1.391037912 922.039 end
'moderator
    graphite 26 DEN=1.843 1.0 900.0 end
' waste mixture(s)
    xe-135 7 0 1.0e-20 898.15 end

```

```

' waste mixture(s)
    xe-135 8 0 1.0e-20 898.15 end
end comp
read celldata
latticecell symmslabcell pitch=5.08 1 fuelr=2.21432066 26 end
end celldata

' _____

' Depletion data
' _____

read depletion
    1 decayonly 7 8
end depletion
' _____

' Burn data
' _____

read burndata
    power=36.3907476056 burn=3 end
    power=36.3907476056 burn=30 end
    power=36.3907476056 burn=300 end
    power=36.3907476056 burn=3000 end
end burndata
READ opus
matl=1 7 8 end
title = 'Masses in fuel salt'
units=gper nrank=2000 libtyp=all
time=days typarms=nucl sort=no
END opus

```

```

read timetable
' gaseous fission product removal
    flow
        from 1
        to 7
        type continuous
        units pers
        nuclides Kr Xe Ar H N O end
        constant 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 end
        time 0.0 end
        factor 1.0 end
    end flow
' solid fission product removal
    flow
        from 1
        to 8
        type continuous
        units pers
        nuclides Se Nb Mo Tc Ru Rh Pd Ag Sb Te end
        constant 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01
            3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 3.3012E-01 end
        time 0.0 end
        factor 1.0 end
    end flow
' thorium feed
    flow
        to 1
        type continuous
        units gpers
        nuclides u-238 u-235 end

```

```

        constant 0.65 0.35 end
        time 0.0 end
        factor 1.45829479E-03 end
    end flow
end timetable
', _____
'NEWT model data
', _____

read model
Single MSBR Cell
read parm
drawit=yes echo=yes
end parm
read materials
    1 1 ' Fuel Salt ' end
    26 1 ' Graphite ' end
end materials
read geom
global unit 1
    cuboid 10 2.21432066 0 2.21432066 0
    cuboid 30 2.54 0 2.54 0
    media 26 1 10
    media 1 1 30 -10
    boundary 30 4 4
end geom
read bounds
    all=refl
end bounds
end model

```

end

Vita

Sarah Elizabeth Creasman was born to Scotty and Cindy Creasman and raised in East Tennessee. She graduated as salutatorian from Meigs County High School in 2012. Upon graduation, she first attended Middle Tennessee State University until she settled on being a nuclear engineering major provoking a change of college to the University of Tennessee where she graduated with a Bachelor's of Science in 2017. Since January 2018, Sarah has been working on her Master of Science degree at Oak Ridge National Laboratory and the University of Tennessee. Following graduation, she will continue her education at the University of Tennessee as a Doctor of Philosophy student in Nuclear Engineering.