

Exploring Molten-Salt Reactor Source Terms

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INTRODUCTION

Molten-salt reactors (MSR) present an innovative way to generate nuclear power while addressing several safety concerns presented by current and previous generation light-water reactors. As the name implies, MSRs operate by dissolving fissile material in suitable eutectic salt mixtures that are melted and circulated through the core and heat exchanger. Solid-fuel reactors keep actinides and fission products (FP) contained in the fuel, whereas in MSRs, they are in a homogeneous mixture in continuous transit in the primary circuit. Additionally, the behavior of FP elements depends on the chemical characteristics of the carrier salt. As such, the source terms for MSRs can vary depending on the particulars of the chosen reactor design and fuel cycle.

The best current means of understanding the behavior of FPs in MSRs comes from the work carried out during the operation of the Molten-Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) from 1960 to 1969 and its subsequent decommissioning [1]. MSRE was an 8 MW(th) graphite-moderated thermal-spectrum reactor that was operated on both U-235 and U-233 fuel. There was no on-line fuel reprocessing to separate FPs, however, an off-gas system was used to strip fission product gases from the

salt. The experience gained at MSRE provides valuable insight for fluoride salt reactors while serving as a technical basis for studying fission product behavior in other MSR designs.

Fission product behavior in the MSRE fell into four distinct chemical groups: gaseous, always soluble, sometimes soluble, and insoluble. While this categorization of FPs applies to all MSR designs, the specifics might differ due to the carrier salt chemistry and operating conditions. The first of these groups consists of noble-gas FPs such as krypton and xenon which are removed from the fuel-salt by stripping in the off-gas system. Next are the noble-metals such as molybdenum, technetium, ruthenium, silver, and others which were not expected to form stable fluorides under the redox conditions of the MSRE. These metals tended to deposit on various surfaces as well as salt-gas interfaces during reactor operation. Finally, there are those elements that form stable fluorides and remain in the fuel-salt. These include rubidium, cesium, strontium, barium, and the lanthanides [2]. In case of salt spillage, these isotopes would remain in the salt and form the major constituent of the source term [3]. Moreover, all actinides were expected to form stable fluorides and remain dissolved in the salt under normal operating conditions. Figure 1

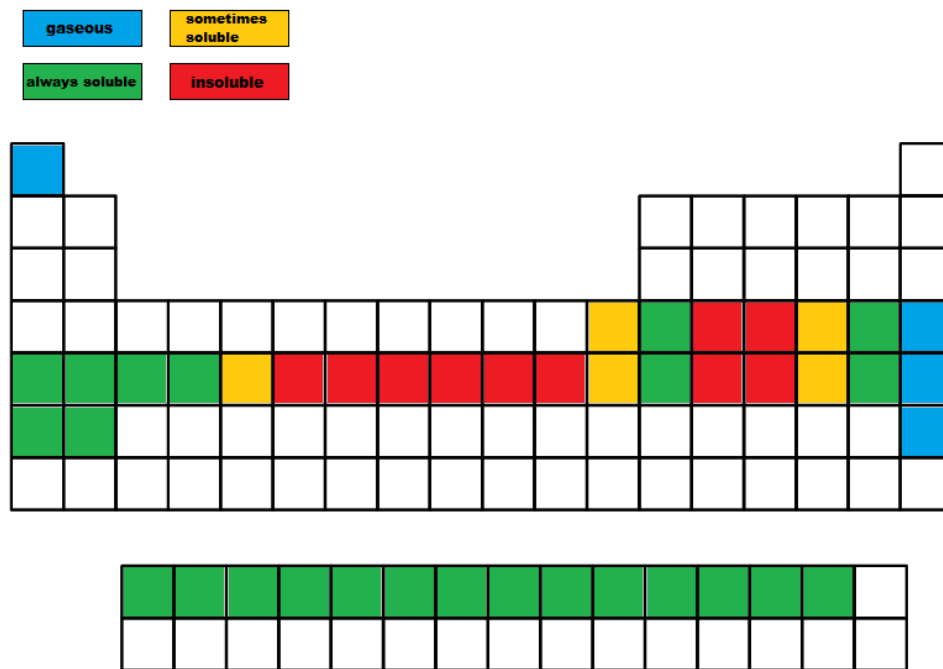


Fig. 1. Fission product classification in the MSRE (Source: ORNL-4865).

shows the chemical behavior of the important FP elements in MSRE.

This summary presents representative calculations from SCALE ORIGEN¹ for fuel burnup for a once-through thermal-fluoride MSR (TFMSR) for both U-233 and U-235 fuel [4], and a once-through molten-chloride salt fast reactor (MCFR) using DU/Pu fuel [5]. The effect of the chemical behavior of FPs on the resulting source release for postulated accident scenarios is discussed and the source terms presented. An additional goal of this work is to articulate the gaps in knowledge when it comes to understanding the behavior of source terms in MSRs.

DESCRIPTION OF INVESTIGATED MSR DESIGNS

This section provides a brief overview of the MSR design space investigated and the design characteristics most important to determining source terms.

MSRs are Generation-IV nuclear reactor concepts that promise many benefits including inherent safety, fuel cycle flexibility, improved resource utilization, high-temperature heat generation, and consumption of legacy actinides [5]. Of late, there has been a significant revival of interest in this technology by U.S. and international nuclear energy programs as well as private companies. The result has been a wide array of reactor designs proposed by these organizations which are summarized in Figure 2.

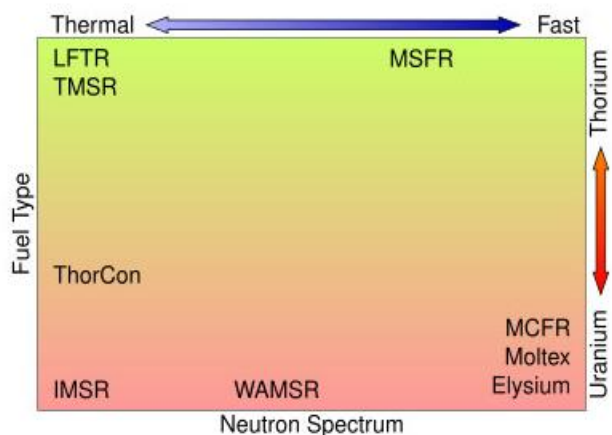


Fig. 2. MSR design space (Source: Kirk Sorenson).

The source term is the amount of radioactive material that can be dispersed into the environment in case of an accident. For this summary, only the reactor core, heat exchanger, drain tanks and associated primary circuit piping containing the fuel inventory as well as the off-gas system are considered. Any on-site or off-site reprocessing or waste storage is not considered and will have separate associated source terms. Given the variety of fuel types, neutron spectrum, and chemical characteristics of the carrier salts, there is potential for considerable divergence in the source

terms when comparing reactor designs. For example, long-lived transuranic isotopes which form a significant portion of the source term are lower for designs that employ the thorium fuel cycle. Moreover, the chemical behavior of FPs can be significantly different given the composition of the carrier salt used. While specific design choices will reflect the specifics of the means for source release, the qualitative source terms should not differ significantly, thus providing a means for evaluating radioactive release for postulated accidents.

The reactor designs considered in this paper were chosen to cover as much of the parameter space without getting into explicit design minutiae. The thermal-fluoride MSR is assumed to operate in a “sealed core” configuration for the entire lifetime of the core. This means that in the event of a breach in the containment, highly radioactive FPs have the potential to be released and form the dominant source term of concern. The MCFR shares many of the same advantages of thermal-spectrum fluoride MSRs while realizing a U/Pu breeding cycle due to improved capture-to-fission ratio in plutonium and other actinides at higher neutron energies. A representative salt composition, coupled with the knowledge that these designs employ a fast neutron spectrum, can be used to make reasonable estimates of the actinide and fission product concentrations and thus the resulting source terms.

GENERAL CONSIDERATIONS FOR MSR SOURCE TERMS

MSRs allow for on-line fuel processing to remove FPs products. In the case when the fuel is processed, after some start-up period the reactors operate at an equilibrium state where fissioned fuel is continuously replenished either by feeding new fuel or breeding in the fertile material. Furthermore, the fuel is continuously mixed in solution. Therefore, the term “fuel burnup” does not apply to these reactors as it does to current light water reactors as no specific amount of energy release can be associated with a sample of the fuel. Additionally, in all MSR designs the xenon and krypton bubble out of solution. Thus, there is no need for excess reactivity to compensate for fuel burnup and poisoning from Xe-135 [3]. Moreover, there is no fuel meltdown possible in MSRs as these reactors are designed for molten fuel. Similarly, since the fuel is the primary coolant, these designs do not have a “Loss-of-Coolant” accident in the traditional sense. In the case of reactors where the fuel is the primary coolant, a loss-of-coolant would entail a primary pump-trip or a breach in the primary system. Thus, terminology familiar to current generation nuclear reactors do not directly apply to MSRs presenting the need for an updated technical vocabulary applicable to these designs.

¹ SCALE Code System, ORNL/TM-2005/39, Version 6.2.1 (August 2016).

Apart from a knowledge of the number of atoms of an isotope present at any time, it is also important to consider the state of the isotopes to understand their location following a release event. Evidently noble gases stored in the off-gas system will remain in the same state following release until they decay. The soluble elements form stable fluorides in the salt and remain in solution. The insoluble and sometimes-soluble elements tend to plate out in different surfaces of the primary circuit during operation and remain there in the event of salt drainage [2]. For the chloride-salt system, the chemistry is similar with some differences adopted from [6].

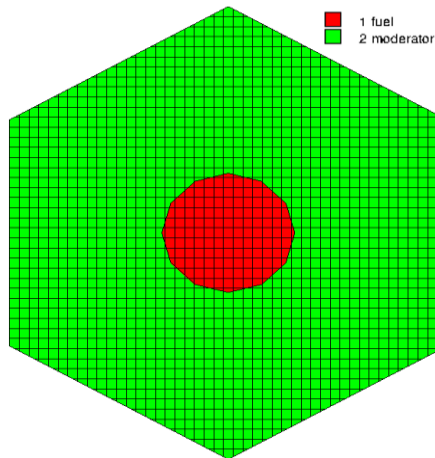


Fig. 3. ORIGIN 2-D fuel cell for depletion calculation.

CALCULATION OF ACTINIDE AND FISSION PRODUCT CONCENTRATIONS

Depletion calculations have been carried out in SCALE using the ORIGIN package and 238 group ENDF/B-VII.0 cross section data library. Given the salt composition and basic lattice geometry, the isotopic concentrations can be determined quite easily. However, ORIGIN is limited in that the fuel is treated as a solid lump and consequently there is no bubbling of gaseous FPs or reprocessing capability. This can be problematic for certain elements such as Xe-135. In the presented calculations, Xe-135 remains in core and undergoes neutron capture to Xe-136. However, in an MSR, the Xe-135 would bubble out and decay to Cs-135 in an off-gas storage tank. Barring these limitations, ORIGIN provides a quick, easy, and reliable means of calculation that is representative of the fission product distribution in a depleting mass of fuel.

For the TFMSR, calculations were carried out for low-enriched U-235 and 90 wt% U-233 (similar to MSRE) fueled systems. U-233 fuel is included to study the source terms in a thorium cycle MSR. A representative fuel cell for this design is shown in Figure 3. For the simulations presented, a hexagonal lattice with a pitch distance of 15 cm and a fuel salt channel of radius 2.7 cm is chosen. Mirror boundary conditions are imposed on the edges to simulate

an infinite lattice. This infinite lattice of fuel is burnt at a rate of 20 MW/MTU. For the MCFR, a 30 mole% depleted-uranium DU-Cl₃ and 30 mole% reactor-grade PuCl₃ mixture in 40 mole% NaCl is used. Since there is no moderator, for the MCFR calculation both regions in Figure 3 are set to fuel. The salt is then burned at the same rate of 20 MW/MTiHM. These input properties are summarized in Table I.

Table I. ORIGIN inputs for depletion calculations

	TFMSR U-235	TFMSR U-233	MCFR
Pitch	15 cm	15 cm	-
Moderator	Graphite (1.8 g/cc)	Graphite (1.8 g/cc)	-
Carrier salt	LiF-BeF ₂	LiF-BeF ₂	NaCl
Fuel	LEU	U-233 (90wt%)	Reactor grade Pu
Fuel density	3.353 g/cc	3.353 g/cc	3.022 g/cc

Given the nature of the “infinite universe” calculations, the resulting output is in concentration per unit mass. This output can be used to define specific activity which is independent of inventory size given appropriate half-lives and mass numbers. The resulting specific activities from each of the FP chemical groups and actinides after seven years of operation (arbitrary) is presented in Table II.

Table II. Calculated specific activity of interest in Ci/g

	TFMSR U-235	TFMSR U-233	MCFR
Gaseous	0.427	0.457	0.602
Semisoluble	0.665	0.868	-
Soluble	2.854	4.404	3.456
Insoluble	0.796	0.153	1.915
Actinides	0.209	0.004	0.129
Total	4.951	5.886	6.102

SOURCE TERMS FOR POSTULATED ACCIDENT SCENARIOS

In any facility where many curies worth of radioactive material is present, there are two containment barriers preventing dispersal into the environment. For MSRs, core vessel and piping form the primary barrier. The secondary barrier is generally an annulus made of concrete and other shielding material that houses all components of the primary system. For the presented study, two postulated release scenarios are considered - a spray or drip type leakage in the primary system with adequate operator response, and a

major rupture of the reactor vessel that unloads all the fuel salt into the secondary containment area which has experienced a breach. Ref. [7] describes such episodes for the MSRE.

Spray or Drip Leakage in Primary Circuit

For a spray or drip type leakage in primary piping at a few cubic centimeters a minute, it was assumed that the reactor would be shut down before any more than four liters of the fuel salt had escaped. It was further assumed that about 10% of the solid FPs dissolved in the salt, 10% of the iodine, and all the gaseous FPs would volatilize and be dispersed into the cell environment.

Table III. Source term (Ci) for postulated primary circuit leak

	TFMSR U-235	TFMSR U-233	MCFR
Total released (Ci)	50,000	70,802	43,335
Volatilized dissolved isotopes	4,720	7,075	4,178
Volatilized I₂	337	378	413

For the MSRE, it was predicted that ~20,000 Ci of activity would be released into the secondary cell of which ~1400 Ci of solid FPs, ~480 Ci of I₂, and ~3200 Ci of Xe and Kr would volatilize. The burnup time is not quoted for these results [7]. For a similar set of circumstances, the calculated activities at the end of 7 years of operation for the described reactor systems are presented in Table III. It is assumed that all insoluble FPs plate out in the system and all semi-soluble isotopes remain in the salt. Furthermore, no actinides volatilize which is a fair assumption.

Table IV. Source term (Ci) for worst possible scenario

	TFMSR – U235	TFMSR – U233	MCFR
Total	9.299x10 ⁸	1.279x10 ⁹	7.511x10 ⁸
Volatilized dissolved isotopes	7.875x10 ⁷	9.486x10 ⁷	6.200x10 ⁷
Volatilized I₂	5.617x10 ⁶	6.292x10 ⁶	6.151x10 ⁶
Gaseous FPs	9.556x10 ⁷	1.019x10 ⁸	1.08x10 ⁸

Major Rupture of Reactor Vessel and Breach of Secondary Containment

For the MSRE, this accident was considered the worst possible scenario where all primary inventory including the

gaseous FPs were released into the secondary containment area. In this sequence of events, a breach in the secondary containment would lead to source term release into the environment. This scenario results from only the most severe and unanticipated incidents such as a major earthquake or tsunami, intentional sabotage, or missile damage. With similar assumptions as before for percentage of FPs volatilizing, the MSRE study quotes a possible activity of ~6.8x10⁵ Ci of solid FPs, ~2.5x10⁵ Ci of I₂, and ~3.75x10⁵ Ci of noble gases being dispersed into the environment. For a similar set of circumstances, the source terms after 7 years of operation for the described reactor systems are presented in Table IV. It is assumed that the reactors have a power output of 2 GW(th).

CONCLUDING REMARKS

Representative source terms for MSRs have been calculated using SCALE for two accident scenarios originally postulated for MSRE. Although ORIGEN is meant for solid fuels, it provides a reliable estimate of the isotopic evolution and the resulting specific activities. To represent the vast MSR design space, a thermal-fluoride and a fast-chloride fueled once-through MSR were investigated. For these accidents the source terms are presented for the two reactor designs after seven years of full-power operation burning at 20 MW/MTiHM.

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