Material Accountancy for Molten Salt Reactors: Challenges and Opportunities

Prepared for US Department of Energy

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NNSA National Nuclear Security Administration

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ABSTRACT

The large upfront capital costs associated with new nuclear facilities is a substantial barrier to future growth. Facility retrofits to accommodate effective nuclear security can be expensive, particularly in facilities with large inventories of bulk nuclear material. The recent and sustained interest in advanced nuclear power presents the opportunity to implement material control and accountancy (MC&A) measures in the design phase to lower the possibility of costly retrofits. Prior, related work has discussed other aspects of MC&A for liquid-fueled Molten Salt Reactors (MSRs), including item accounting strategies for front and back end material balance areas, material balance organization, and more. This work extends the current body of work for MC&A for MSRs by applying traditional nuclear material accountancy (NMA) principles employed at bulk nuclear facilities to several liquid-fueled MSR designs with varied fuel materials and neutron spectra. Although there are no substantial statistical barriers for applying traditional NMA techniques used in bulk facilities to liquid-fueled MSRs, challenges arise from several design properties not present in current bulk facilities. Specifically, the large inventory fissible inventory present during operation leads to high uncertainities in the material unaccounted for (MUF), thereby, reducing the probability of detection for material loss. Contemporary light water reactors (LWRs) also have large fissible inventories present during operation, but employ robust containment and surveillance (C/S) methods. Liquid-fueled MSRs have properties of both current light water reactors and bulk facilities, but are not directly analagous to either. Consequently, robust MC&A for liquid-fueled MSRs will require a blend of strategies used for LWRs and bulk facilities. Such a system will material accountancy that is complemented with robus C/S and process monitoring.

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NOMENCLATURE

ARS Advanced Reactor Safeguards

C/S Containment and Surveillance

CFR Code of Federal Regulations

DA Destructive Assay

FAP False Alarm Probability

KMP Key Measurement Point

LEU Low Enriched Uranium

LWR Light Water Reactor

MBA Material Balance Area

MBP Material Balance Period

MCSFR Molten Chloride Salt Fast Reactor

ML Machine Learning

MOSART MOlten Salt Actinide Recycler & Transmuter

MSDR Molten Salt Demonstration Reactor

MSFR Molten Salt Fast Reactor

MSR Molten Salt Reactor

MTHM Metric Ton Heavy Metal

MTIHM Metric Ton Initial Heavy Metal

MUF Material Unaccounted For

NDA Non-destructive Assay

NMA Nuclear Material Accountancy

NRC Nuclear Regulatory Commission

PD Probability of Detection

SCALE Standardized Computer-Analysis for Licensing Evaluation

SEID Standard Error of Inventory Difference

SITMUF Standardized Independent Material Unaccounted For

SQ Significant Quantity

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1. EXECUTIVE SUMMARY

The recent and sustained interest in advanced nuclear projects encompasses a wide range of reactor designs and fuel cycle facilities. Molten Salt Reactors (MSRs) are one such design. As with all nuclear projects, MSRs will require regulatory oversight, review, and analysis. One important component is the Material Control & Accountancy (MC&A) system. It is currently unclear what specific regulations and requirements will be applied to MSRs, however, material accountancy will be required. This work considers the statistical performance of existing accountancy approaches for bulk facilities as applied to liquid-fueled MSRs with consideration to their unique design features (e.g., continuous feeds and removals, flowing fuel material, constant depletion and decay). Other MSR design types, such as integrated core designs (e.g., designs where the entire core is replaced at end-of-life) or solid fueled liquid cooled designs, are outside the scope of the analysis discussed here.

Statistical tests used for bulk processing facilities (e.g., reprocessing and enrichment) are adapted for use with liquid-fueled MSRs. Using the best available computational tools, this work establishes a performance ceiling for existing statistical methods to perform material accountancy tasks, as many complex phenomena are not accounted for here (e.g., chemical interactions, precipitation of materials, holdup, etc.). This work considers five different reactor designs that span a variety of fuel cycles to draw broad conclusions regarding material accountancy for liquid-fueled MSRs. As these designs considered are largely initial concepts, many operational parameters needed to simulate reasonable inventories are not provided. Only a minimal set of assumptions were made:

- Salt lifetimes are 30 years
- Feed and removal rates are optimized to maintain a constant fissile or fertile inventory
- Only removals of fission product gasses and noble metals during normal operation
- Simulated material loss were modeled as substitutions of feed material

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NOTE: Results presented here should be taken as best estimates given the current availability and fidelity of underlying tools used to perform the analysis. This work bounds performance of a MC&A system for liquid-fueled MSR designs, but does not necessarily reflect actual expected performance of a specific design.

Analysis provided here shows that, regardless of design and fuel cycle choice, detection of material loss using traditional statistical tools alone is very challenging. Detection of the most obvious abrupt material losses using commonly employed statistical tests is projected to require at or above current state-of-the-art destructive analysis levels of precision. This largely arises from the large fissile inventory present in most designs considered here (see Figure 1-1 below for one such example). It is estimated that the lower bound for expected computational uncertainty is around 4% for thermal spectrum designs and 0.25% for fast designs, which when taken in the context of the large fissile inventory, can lead to large contributions to material balance uncertainty (See Figure 1-2 which shows standard error of the inventory difference for at 1% measurement uncertainty for the designs considered in this work).

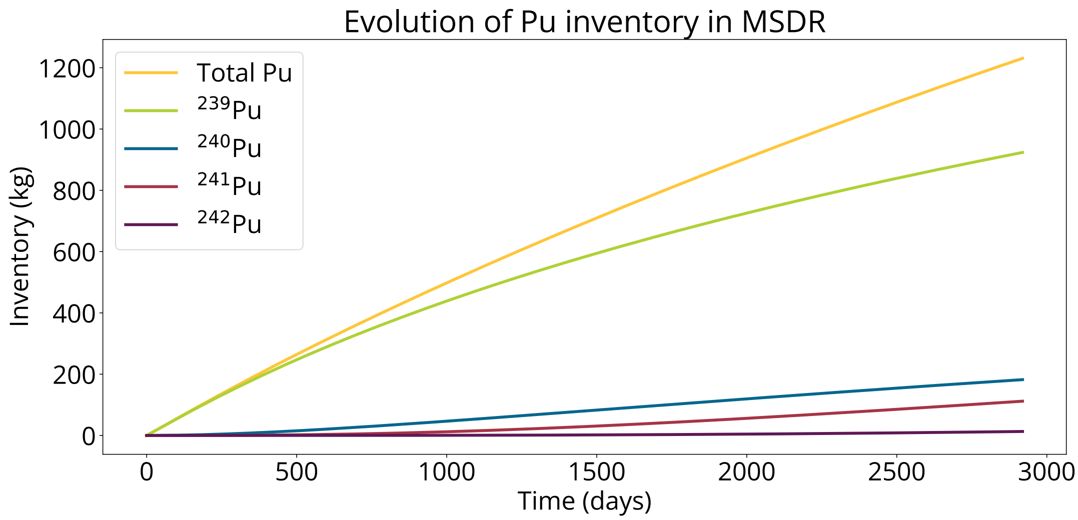


Figure 1-1. Total Pu as a function of time for Molten Salt Demonstration Reactor (thermal LEU/Pu cycle) inventory

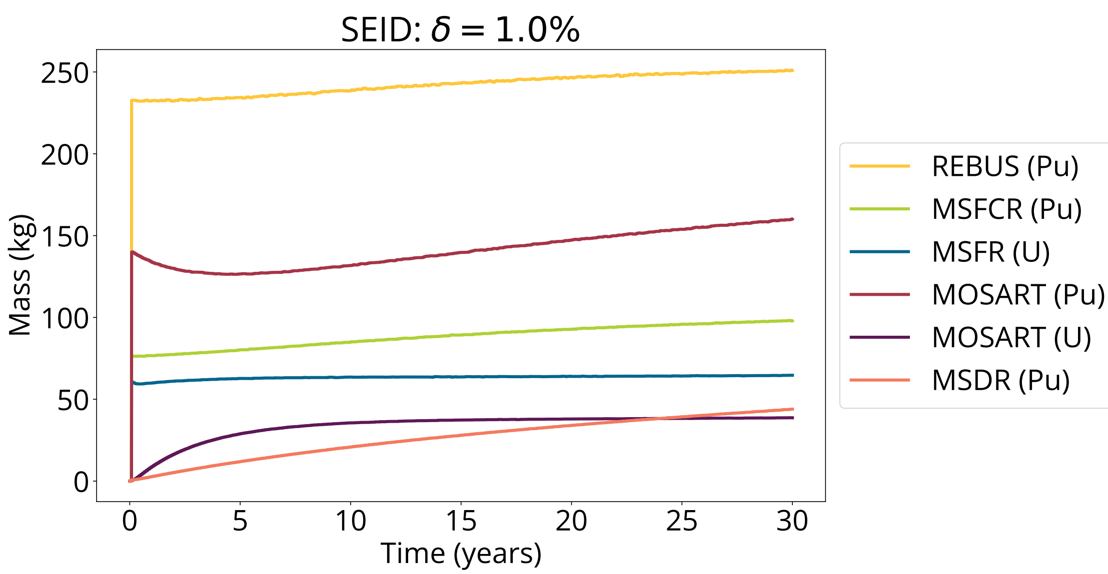


Figure 1-2. SEID as a function of time for several designs at 1% measurement uncertainty

This work also notes that although larger thermal power levels generally have larger fissile inventories, which reduces performance of a MC&A system, fissile inventory per unit design power varies across designs. This is described concretely in Tabe 1-1 where the nominal material balanced uncertainty (SEID) at 1% measurement uncertainty is shown for several designs. The MSCFR is the largest design considered in this work with a design power of 6000 MW_{th}, yet it has a smaller nominal SEID than both REBUS and MOSART, both of which have substantially smaller design powers. This shows the possibility of a security-conscious core design wherein equilibrium fissile inventory is minimized to improve overall MC&A system performance.

Design	Design Power (MW_{th})	Material	Average nominal SEID at 1.0% uncertainty (kg)
MSDR	750	Total Pu	26.11
MOSART	2400	^{233}U	33.60
MOSART	2400	Total Pu	141.164
MSFR	3000	^{233}U	63.20
REBUS	3700	Total Pu	242.08
MSCFR	6000	Total Pu	87.97

Table 1-1. Lower limits of detection based on current SEID values

The large quantity of fissile inventory present in liquid-fueled MSRs is not unlike conventional LWRs. For LWRs, this material is often secured using containment and surveillance (C/S) methods since specialized equipment (e.g., cranes) are required to access material in a LWR's core. Security for LWRs is further complemented by random sampling of discrete fuel assemblies with support from validated computational tools. Taken together, these measures provide assurances that material is present and accounted for. Liquid-fueled MSRs contrast with LWRs because fissile materials are in a bulk form. This is further complicated by other unique potential features of MSRs such as inventories that change with time, which impact performance of commonly used statistical tests.

Additional security measures must complement material accountancy for liquid-fueled MSRs to reach acceptable levels of performance for the larger MC&A system. Although not explored in this work potential opportunities for MSRs to complement MC&A systems could include the following:

- **Opportunity**: Enhanced C/S for entry points into shielded environment housing primary fuel salt loop(s)
  - Discussed in work conducted by ORNL [1]
- **Opportunity**: Process monitoring of primarily loop systems (e.g., pump speeds, temperatures) may change in response to material loss
  - Considered in forthcoming work conducted during FY23
- **Opportunity**: Data-driven analytics could uncover novel monitoring strategies
- **Opportunity**: Core optimization during early design stages can reduce equilibrium fissile inventories leading to improved MC&A system performance

2. INTRODUCTION

Nuclear facilities are continuing to see high levels of interest around the world with at least 60 ongoing nuclear projects domestically and even more internationally [2]. This represents a significantly increased burden on regulatory stakeholders, particularly for advanced facilities with less historical operational experience. It is therefore important to develop new regulations and associated systems that result in safe and cost-effective nuclear operation. This work focuses specifically on material accountancy of liquid-fueled molten salt reactors (MSRs) which have several important features that differentiate them from traditional Light Water Reactors (LWRs). Material control and accountancy (MC&A) for current LWRs are relatively straightforward given that the core is sealed during operation, requires special equipment to open, and the fuel is contained within discrete fuel elements. However, liquid-fueled MSRs[1] often have several design specific features that prohibit the direct application of existing LWR-based material accountancy approaches.

Security for MSRs, and material accountancy in specific, are complicated by the heterogeneous design landscape. Conventional LWRs are primarily dominated by two different designs; pressurized water reactors (PWRs) and boiling water reactors (BWRs). MSRs, by contrast, have a wide range of potential designs, fuel cycles, neutron spectra, and features. This makes it difficult to develop a one-size-fits-all material accountancy strategy as unique design features will impact system design. This diverse design landscape emphasizes the need to consider MC&A during the design phase.

Although the landscape of liquid-fueled MSRs is heterogeneous, there are several common features common to most designs that will impact MC&A design. These features can be used to specify reference designs which can be used to perform preliminary material balance evaluations.

Contributions: The goal of this work is to quantify MC&A relevant performance metrics for several different MSR designs, focusing specifically on the core itself, using traditional material accountancy techniques that have been adapted to the unique properties of MSRs (e.g., inclusion of salt burnup). This preliminary work relies on the best available tools as of writing and may be updated as underlying computational software improves. Findings from this work should not be taken as high fidelity results indicative of actual MC&A performance of future designs, rather, findings should be taken as estimates to inform future R&D activity. This work complements other existing work that considers a broader view of MC&A for MSRs at the facility level [1, 3]. Contributions to existing literature are as follows:

- **Establish reference baseline inventories (Appendix B):** Leveraging existing tools such as SCALE and models from Betzler [4–7] and Rykhlevskii [8, 9], baseline inventories for several liquid-fueled MSR designs are established. These inventories serve as reference points for statistical analysis of material loss and can be used for future R&D as all of our SCALE input decks are available upon request.
- **Develop modified material balance for liquid-fueled MSRs (Section 5.1, Appendix D.1):** The traditional material balance works well for most contemporary bulk facilities. However, the fissile material in a liquid-fueled MSR is undergoing constant nuclear transmutation (i.e., depletion and decay), which must be accounted for. Consequently, accountancy techniques used

[1] This document will refer to liquid-fueled MSRs as simply MSRs for brevity, while noting in general, not all MSRs are liquid fueled.

with bulk facilities are not directly applicable. This work proposes two different formulations for such a material balance, compares them on the basis of uncertainty, and establishes a suitable formulation.

- **Estimate uncertainty from nuclear data (Section 5.3.1, Appendix G):** Proper quantification of uncertainty terms in a material balance is critical for accurate estimate performance of a MC&A system. Liquid fueled MSRs will likely have many contributions from modeling error (i.e., unknown system conditions not accounted for in modeling) and measurement uncertainty. This work builds upon parallel efforts [10] to estimate nuclear data uncertainty to establish a lower bound standard error of the inventory difference (SEID) for several liquid-fueled MSR designs (Section 7).
- **Determine baseline performance of traditional accountancy statistics for loss detection in liquid fueled MSRs (Section 6.3):** Traditional safeguards statistical analysis is applied to several different liquid-fueled MSR designs for several different material loss scenarios and levels of measurement error. Large fissile inventories for most designs result in large SEIDs and low probability of detection using statistical methods on the MSR inventory alone.

3. BACKGROUND

Security for LWRs is achieved using a variety of containment and surveillance (C/S) and MC&A measures. Implementations that ensure material is present and unmodified are relatively straightforward for LWRs given that fuel is in item form and difficult to move without special equipment (e.g., cranes). However, MSRs present challenges as the primary nuclear fuel is dissolved in a flowing molten salt. This can be further complicated by the presence of multiple loops (in the case of some fast reactors) that contain fissile material.

MSRs have some resemblance to existing bulk facilities such as reprocessing or enrichment plants. Bulk facilities already have established and effective material accountancy systems, however, MSRs are complicated by the fact that fuel material is undergoing constant nuclear transmutation (i.e., depletion, decay, etc.). This creates the need for an explicit inclusion of nuclear transmutation into the material balance. If not included, it would be impossible to determine if a change in actinide inventory was caused by material loss or depletion caused by normal reactor operation (further discussion provided in Appendix B and D).

Many MSR designs also have continuous feeds and removals of materials (i.e., fuel salt, fission product gases and noble metals) to improve reactor performance, but these operations complicate material balances as they must also be accounted for. Contemporary bulk facilities, such as enrichment facilities, also often have input and output material flows. These are often accounted for by counting and sampling input material containers (e.g., for enrichment these are UF_6 cylinders). A similar strategy may prove effective for continuous feeds and removals of MSR flows [1], with some modification. For example, a MSR design utilizing a constant feed will also have a complementary constant removal to maintain a constant system volume. These removal tanks could be subjected to increased C/S until the tank is filled and could be verified, at which point the tank could be treated as an item. Further descriptions of front-end and back-end MSR MC&A strategies can be found in [1].

Additionally, measurements used to calculate a material balance itself could be difficult to conduct. The MSR fuel salt, like all irradiated fuel materials, have high levels of activity which creates a challenging measurement environment. MSRs also lack the decades of research and experience that exist for current LWR designs. There are currently many unknowns for MSR salts and fuels. For example, basic salt properties and chemical interactions are unresolved for some materials. This could lead to unexpected holdups and material plate out that could complicate calculation of an accurate material balance. This work neglects these potential error contributions in order to establish a statistical ceiling for MC&A of several reference MSR core designs. Under real-world conditions, additional phenomena (e.g., uncertain reactor core conditions) will add additional uncertainty thereby further degrading MC&A performance.

4. RELATED WORK

Material accountancy of bulk facilities has been considered since at least the 1980s. The majority of this literature focuses on the development and application of "near-real-time" accounting [11, 12] wherein statistical evaluations are made in regular intervals throughout the year rather than a single yearly evaluation. The most common approach to material accountancy for bulk facilities involves the calculation of Material Unaccounted For (MUF) [13, 14] and the associated uncertainty. Simple control charts and thresholds can be used to detect abnormal operation for smaller facilities. However, larger facilities often employ more complex statistical tests such as a combination of the standardized independent transformed material unaccounted for (SITMUF) [15], Page's trend test [16–21] and GEMUF [22].

MSRs have some notable design differences from facilities considered in the traditional safeguards literature mentioned above, which necessitates the inclusion of additional tools and techniques. Reactor physics tools such as SERPENT [23–25] and SCALE [4–7] have been used to model MSR fissile material evolution and consider the contribution of nuclear data uncertainty to MC&A performance. Other tools such as TRANSFORM [26] and NERTHUS [27] have been used to model the thermophysical behavior of MSRs.

Literature focused on material accountancy strategy for MSRs dates back to 2020 and is relatively small. Initial work has been focused on exploring potential signatures for facility misuse, as these may vary from existing bulk facilities, and determining larger facility material accountancy strategy. Several key findings in the initial literature include the following:

- Representative MSR designs considered in literature often have large inventories leading to challenges for detecting material loss when relying solely on material accountancy and statistics [28, 29]
  - This work aims to summarize and consolidate findings related to this point
- Nuclear data uncertainty has a contribution to material accountancy performance. This contribution is large compared to the relative isotopic change due to loss (i.e., nuclear data uncertainty increases difficulty of potential process monitoring) [10].
- Nuclear data uncertainty is largely insignificant compared to inventory measurement error (i.e., has little contribution to material balance performance) [28, 29]
- Cumulative changes in fission product inventories, even for large material losses, could be difficult to detect [30]
- Facility-level material accountancy approach can be designed to rely more on input and output transfers to lessen the impact of large MSR core inventory uncertainties [1, 3]
- Consideration should be given to the MSR fuel salt activity and fissile concentration, which could have significant impacts on overall security strategy [1]

5. METHODOLOGY

This work primarily relies on existing tools and methodologies and only develops extensions where necessary. SCALE is primarily used to carry out reactor physics calculations and nuclear data uncertainty estimates whereas established literature on material balance analysis is applied. A general outline of analysis conducted in this work is as follows:

- SCALE is used to estimate time-dependent fissile inventories
- Various levels of measurement and computational uncertainty are applied and propagated
  - A "measured" data set is based on a range of measurement errors as experimental performance of measurement technology is unknown
  - A "calculated" data set is based on combined nuclear data uncertainty obtained from SCALE
  - Other potential sources of uncertainty (e.g., chemical phenomena) are not considered
- Page's trend test on SITMUF is calculated for the relevant fissile inventory based on the applicable fuel cycle

The procedure above is conducted for several different scenarios to obtain estimated performance statistics.

5.1. Analytical approach

The material balance (MB), which is sometimes referred to as material unaccounted for (MUF) or inventory difference (ID), is a statistical quantity that is calculated at regular intervals defined by the material balance period (MBP) [13]. The MB is calculated using measured data from key measurement Points (KMPs) within a material balance area (MBA). The MBP, KMPs, and MBA are all values that are determined by a subject matter expert and is often a balance between accountancy goals and measurement constraints (e.g., difficulty of measurement, required measurement frequency, etc.). As NMA can be thought of as an audit of facility records, the MB would be a methodology to validate reported book values. This is achieved by accounting for all material in a given area for a specified time period [14].

This work generates performance metrics in terms of probability of detection by considering Page's trend test (also called cusum test) on SITMUF. Specifically, this trend test looks for changes in the SITMUF sequence, which is a modified MUF sequence that incorporates knowledge of measurement system performance. Further details about the material balance, SITMUF, and Page's trend test can be found in Appendix D.1.

5.2. Data scope

A total of five different reference MSR designs are considered in this work. Material accountancy for MSRs will likely have to account for unique design features, so the exemplar designs discussed in this work are chosen to cover a variety of neutron spectrums and fuel cycles. These designs included the MSDR [31], MOSART [32], REBUS [33], MSFR [34], and MCSFR [35]. The designs are summarized in Table 5-1 below with additional details in Appendix C.

MSR parameter summary					
Parameter	**MSDR**	**MOSART**	**REBUS**	**MSFR**	**MCSFR**
Thermal Power (MWth)	750	2400	3700	3000	6000
Fuel Salt Composition (mol%)	$LiF\text{-}BeF_2\text{-}ThF_4\text{-}UF_4$ (71.5-16-12-0.5)	$LiF\text{-}BeF_2\text{-}ThF_4\text{-}TRUF_3$ (69.72-27.1.28)	NaCl + (0.711% ^{235}U + 16.7 at.% TRU)Cl_3 (55-45)	$LiF\text{-}ThF_4\text{-}^{233}UF_4$ (77.5-19.9-2.6)	$NaCl\text{-}UCl_3\text{-}^{239}PuCl_3$ (60-36-4)
Fuel Salt Feed Material	3.08%^{235}U	0.711%^{235}U + TRU	0.711%^{235}U	^{233}U + ^{232}Th	0.711% ^{235}U + Pu
Fuel Salt (MTIHM)	121.0	28.83507	114.62944	43.33535	67.78803
Blanket Salt Composition	-	-	-	$LiF\text{-}ThF_4$ (77.5-22.5)	$NaCl\text{-}UCl_3$ (60-40)
Blanket Feed Material	-	-	-	^{232}Th	0.711% ^{235}U
Blanket Salt (MTIHM)	-	-	-	17.57098	133.76272
Fuel Cycle	U/Pu	U/Pu+Th/U	U/Pu	Th/U	U/Pu
Neutron Spectrum	Thermal	Fast	Fast	Fast	Fast

Table 5-1. Reference design parameters used to generate baseline performance metrics.

5.3. Data generation

All data used for this analysis is generated using computational means given the lack of real-world data from MSR facilities. Specifically, the SCALE code system (SCALE 6.3.b15) is used to generate results for this work. It should be noted that SCALE 6.3.b15 is still in beta, and as beta features are used, these results should be taken as the best possible estimate given the current development status of available tools for MSR modeling. Results presented in this work may require revision in the future as computational tools for MSR analysis mature, but current results should be accurate on the order of magnitude, which should be sufficient for an initial material balance analysis.

5.3.1. Uncertainty estimation

One important component in NMA is quantification of measurement uncertainty. For MSRs, there will also be a contribution to uncertainty from the reactor physics calculations that estimate the behavior of relevant fissile inventories. These uncertainties can arise from several sources including nuclear data uncertainty and uncertainty in model-relevant reactor conditions (e.g., chemistry, current fuel composition, etc.). As there are currently no real-world dataset available to characterize uncertainty in reactor conditions, this work focuses on the contribution of nuclear data uncertainty.

NOTE: Computational uncertainties discussed in this work represent the expected lower bound. There will be additional uncertainties that cannot be currently quantified that will contribute to the overall material balance uncertainty. These uncertainties will arise from imperfect knowledge of the system (e.g., chemical states, holdup, plate out, current salt conditions, etc.) will only further increase the overall material balance uncertainty.

Estimates for key fissile isotopes are obtained for each design considered by using the SCALE/SAMPLER sequence. The SCALE/SAMPLER sequence performs general uncertainty analysis by sampling a given input file and analyzing the output response distribution. Several different properties can be perturbed, including; nuclear data, resonance self-shielding, and a variety of model parameters.

This work focuses exclusively on nuclear data uncertainty data which includes cross-sections, yield, and decay data. SCALE/SAMPLER was run 500 times for each reactor type to determine the uncertainty in the actinide inventory as a function of time under baseline conditions [2]. Terms in this work such as "computational" or "calculated" uncertainty will therefore correspond to the nuclear data uncertainty (combined decay, cross-section, and yield uncertainty).

5.3.2. Baseline calculations

"Baseline calculation" are calculations that model the nominal operating behavior. This work considers the time-dependent inventory and associated MC&A implications of five different reactors (described in section 5.2 and 6.2). Developing and testing each individual model would require a significant level of effort. Instead, this work utilizes models generated in previous work by Betzler, Bae, and Rykhlevskii [7–9]. Further details can be found in Appendix E.1

5.3.3. Material loss calculations

It is important to simulate the impact of material loss using reactor physics tools due to the presence of potential non-linear feedbacks. For example, a removal of fissile material would reduce k_{eff} and alter the homogenized reactor cross-section, which could alter the actinide inventory rate of change as well as reactor operation. Consequently, the act of removing material could have security-relevant consequences beyond the immediate loss of fissile material. The evolution of fissile inventory under

[2] SCALE/SAMPLER was *not* run with the loss scenarios as it was assumed that there would be no significant differences in nuclear data uncertainty due to the loss itself.

material loss conditions described in this work all included feedback from relevant reactor physics phenomena. More details are provided in Appendix E.2.

6. EXPERIMENTAL RESULTS

This work is aimed at evaluating the MC&A implications of several different MSR designs. For each reactor, several different material loss scenarios are considered. The majority of the analysis presented here focuses on the fuel salt and does not consider loss from the blanket salt (if applicable) in significant detail. Generally, the blanket salts have low equilibrium inventories of fissile material, and as such, it is difficult to devise loss scenarios wherein at least one significant quantity is removed (see Table 6-1 for significant quantity values).

Material losses discussed in this work are modeled as substitution losses. That is, the removed actinide bearing salt is replaced with an equal mass of a surrogate material. Often, this makeup material is assumed to have the composition as the salt makeup feed. Substitution losses are simulated as it is assumed that direct losses would be easier to detect through use of process monitoring and bulk measurements.

A baseline salt lifetime(the time at which the entire inventory of salt is replaced) is assumed to be 30 years. This value is picked to serve as a common reference across the different evaluated reactors and may not represent a reasonable commercial operational lifetime for each design, though some designs are proposing longer salt lifetimes. Reference feed and removal rates for each design are adjusted to provide somewhat constant inventories of a target material and sufficient criticality, which varies from design to design.

NOTE: Discussion pertaining to MC&A analyses will focus on average inventory values and ignore the evolution over time during normal operation, which may vary based on flow optimization targets.

6.1. Uncertainty analysis

Effective NMA relies on accurate estimates of uncertainty for individual terms in the MB calculation. Errors in traditional measurement systems are well quantified and often expressed by a multiplicative error model, as discussed in Reference [36]. The material balance for MSRs will require a calculated component that also contributes to the overall material balance uncertainty. Analysis using the SCALE code system shows that the lower bound for expected computational uncertainty is around 4% for thermal spectrum designs and 0.25% for fast spectrum designs that are considered. Further details are provided in Appendix G.

6.2. Baseline operation

It is possible that material losses from liquid-fueled MSRs could impact reactor dynamics and the time-dependent evolution of fissile inventory. Consequently, material losses simulated in this work were integrated into the SCALE input deck in order to capture these effects. A baseline case consisting of nominal behavior was first considered as a point of reference. These nominal cases describe the expected evolution of fissile inventory as a function of time. Generally, the designs considered in this work all consist of large fissile inventories that evolve with time. Specific time dependent behavior changes from design to design and is described in further detail in Appendix F.

6.3. Loss scenario analysis

The metrics by which to benchmark traditional MC&A for MSRs is unclear. For example, International safeguards administered by the International Atomic Energy Agency (IAEA) are commonly negotiated between the IAEA and an individual state. Bulk facilities, which are the perhaps the closest contemporary nuclear facility type to MSRs, often rely on effective Material Control and Accounting (MC&A) systems. Statistical tests are used to evaluate material balances to test against loss, however, performance targets are often facility specific.

Domestic MC&A regulations administered by the Nuclear Regulatory Commission (NRC) are similarly based on facility type. It is unclear how a MSR would be categorized under existing NRC regulations. For example, many bulk facilities could be categorized as Category I facility. 10 CFR §74.41 defines these to be facilities that "exceed one effective kilogram of strategic nuclear material in irradiated fuel processing one effective kilogram of strategic special nuclear material in irradiated fuel reprocessing operations other than as sealed sources and to use this material at any site other than a nuclear reactor licensed pursuant to part 50 of this chapter". Note that there is a special provision wherein nuclear power reactors licensed under 10 CFR §50 are exempt from this regulation as they are defined as Category II facilities. Specifically, 10 CFR §73 defined Category II as special nuclear material of moderate strategic significant or irradiated fuel. This distinction, which remains unresolved for MSRs, is important as the MC&A goals defined in 10 CFR §74 vary based on these categories. Dion and Hogue discuss potential applicable regulations in greater detail in [1].

A wide range of different material loss are considered for each of the five designs considered in this work; at least nine cases for each. Developing MC&A insights for MSRs can be challenging as there can be multiple material streams, multiple fissile species, changes in inventory over time, and changes to reactor dynamics. Capturing these phenomena requires consideration of a wide range of material loss. Although not exhaustive, the loss scenarios presented in the following sections are designed to study general trends and responses when considering the MSR-specific challenges described above. All scenarios were based on removals of SQs, which is summarized in Table 6-1 below based on IAEA values [37]. Note that for all scenarios described in the following subsections, Scenario 0 refers to the no-loss, baseline case. Further details regarding the statistical assumptions made in the following analysis can be found in Appendix E.3.

Significant Quantity	
Material	**Quantity (kg)**
Total Pu	8
^{233}U	8
^{235}U (HEU)	25
^{235}U (LEU)	75

Table 6-1. Significant quantity values for several materials

6.3.1. MSDR

All loss scenarios considered here removed one SQ of material while simultaneously increasing the feed such that the total system mass remained constant. This evaluation considered losses over different intervals of time (i.e. both protracted and abrupt) at various points in the operational lifetime of the salt. Note that for loss scenarios with equal material loss duration, there is a lower removal stream required for losses that occur at the end of salt lifetime because of the larger plutonium inventory. Scenarios are summarized in Table 6-2 below where the material loss duration is generally reported as a multiple of the MBP, t. Scenarios are categorized as either 'Early', 'Middle', or 'Late', corresponding to the time in the salt lifetime where the material loss is initiated. This is important as the relevant fissile content (i.e., Pu for the MSDR) changes over the salt lifetime.

Material Loss Scenarios			
Early			
Scenario	Target quantity (kg)	Duration (xMBP)	Start Time (years)
1	8	0.03	1
2	8	0.5	1
3	8	1	1
4	8	2	1
Middle			
5	8	0.03	3
6	8	0.5	3
7	8	1	3
8	8	2	3
Late			
9	8	0.03	7
10	8	0.5	7
11	8	1	7
12	8	2	7

Table 6-2. MSDR Loss scenarios

Additionally, four sets of measurement uncertainties, as described below in Table 6-3, were considered.

Measurement Uncertainties (%)				
Type	Scenario 1	Scenario 2	Scenario 3 (Baseline)	Scenario 4
Bulk Random	0.1	0.5	1.0	5.0
Measured Random	0.1	0.5	1.0	5.0
Bulk Systematic	0.1	0.5	1.0	5.0
Measured Systematic	0.1	0.5	1.0	5.0
Calculated Systematic	0.1	0.5	4.5[3]	5.0

Table 6-3. Measurement uncertainty cases

The probabilities of detection are shown in Figure 6-1. Note that Scenario 0 in Figure 6-1 is the baseline normal case which indicates the false alarm probability whereas the other cases are described in Table 6-2. For convenience, additional labels have been added (Late, Middle, Early, None) to describe when the material loss started within the salt lifecycle. Note that the threshold statistics (i.e., (h, k)) are adjusted for each set of measurement errors such that the false alarm probability remains roughly constant. Regardless of measurement error, the probability of detecting a loss decreases with salt lifetime. This is due to the increase in overall inventory resulting from plutonium buildup that leads to a higher material balance uncertainty. In agreement with conventional material accountancy knowledge, more protracted losses lead to lower detection probabilities. Unsurprisingly, higher measurement errors also lead to lower probabilities of detection.

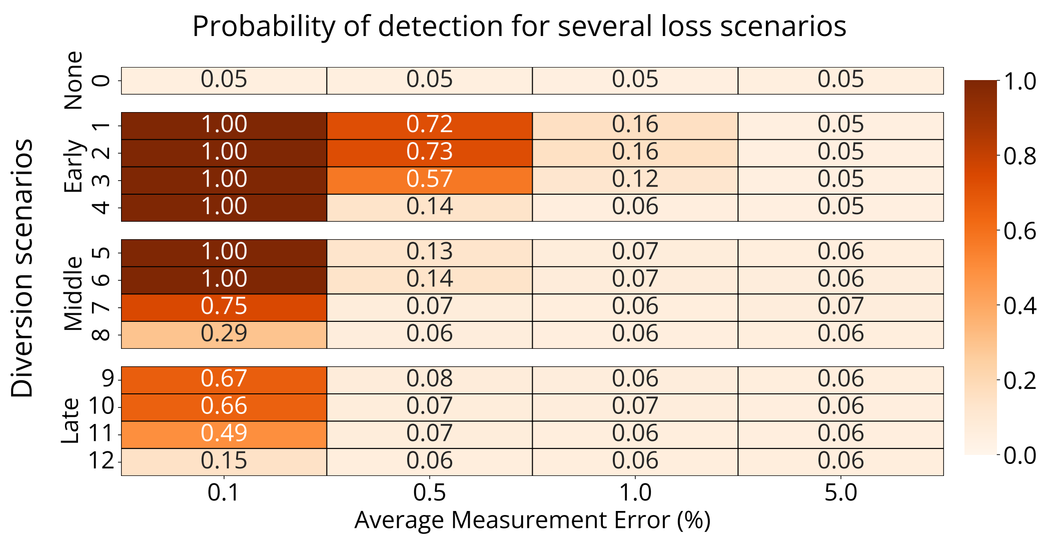


Figure 6-1. Probability of detection for several material losses. A value of 1.00 is a perfect probability of detection whereas 0.00 indicates the scenario cannot be detected.

6.3.2. MOSART

MOSART is somewhat unique from a MC&A perspective as it is designed for the explicit purpose of transmuting spent LWR fuel. As a consequence of this design focus, the single primary salt contains both ^{233}U (to drive criticality) and plutonium (a transmutation target from LWR fuel). Feed and removal rates for MOSART were tuned to produce roughly constant inventories of key actinides such as uranium and plutonium to reduce potential changes in MC&A requirements over the salt lifetime. However, there is a gradual build up and saturation of ^{233}U, which could impact MC&A performance. losses of both ^{233}U and plutonium were considered at various points in the salt lifetime, as specified by Table 6-4 below.

Results of the loss scenario, which are expressed as the probability of detection for the specified loss as detected by Page's trend test on SITMUF, are provided in Figure 6-2 and 6-3 below for plutonium and

[3]Derived from cross section uncertainty using SCALE/SAMPLER

^{233}U losses respectively. In both cases, ^{232}Th was used as a substitution material on the basis that it is already used as a feed material and has low proliferation attractiveness.

Several trends can be observed from the uranium loss cases. Note that the MOSART ^{233}U inventory grows over time before reaching a steady state value. Consequently, for a given material loss, detection probability decreases over the salt lifetime. This can be seen in comparing the performance of Scenarios 1, 2, and 3, which for an error of 0.1% have probabilities of detection of 1.00, 0.96, and 0.76 respectively. The decrease in performance is a result of increasing inventory, which increases the overall SEID, making it more difficult to detect material losses.

Next, note that, as expected, increasing measurement error magnitude is inversely proportional to probability of detection. While errors at the 0.1% level enable reasonable detection performance, most scenarios are undetectable at the 0.5% error level. Increasing levels of measurement error eventually converge to detection probabilities that are essentially the false alarm probability.

The plutonium content in the MOSART salt also increases over the salt lifetime, but not to the same extent as the growth in ^{233}U. The total plutonium inventory is significant even in fresh fuel salt owing to the presence of dissolved spent LWR fuel. This results in more difficult to detect losses than in the uranium case owing to a larger material balance uncertainty, as demonstrated in Figure 6-3. In contrast to the uranium losses, only a few of the scenarios are detected at higher than random levels. Scenarios 14-17 see some detection, albeit still relatively poor performance, due to the larger quantities of material that was removed (i.e., 4 SQ versus 1 SQ).

Scenario	Loss target[1]	Replacement material	Target quantity (kg)	Loss duration (xMBP)	Start time (yr since startup)
1	^{233}U	^{232}Th	8	1	1
2	^{233}U	^{232}Th	8	1	5
3	^{233}U	^{232}Th	8	1	10
4	^{233}U	^{232}Th	16	1	20
5	^{233}U	^{232}Th	24	1	10
6	^{233}U	^{232}Th	32	1	10
7	^{233}U	^{232}Th	32	2	10
8	^{233}U	^{232}Th	32	3	10
9	^{233}U	^{232}Th	32	5	10
10	Total Pu	^{232}Th	8	1	5
11	Total Pu	^{232}Th	8	1	10
12	Total Pu	^{232}Th	16	1	20
13	Total Pu	^{232}Th	24	1	10
14	Total Pu	^{232}Th	32	1	10
15	Total Pu	^{232}Th	32	2	10
16	Total Pu	^{232}Th	32	3	10
17	Total Pu	^{232}Th	32	4	10

Table 6-4. MOSART loss summary

[1] Quantities denoted (total) indicate that the target quantity was split over several species based on their proportions in the target inventory.

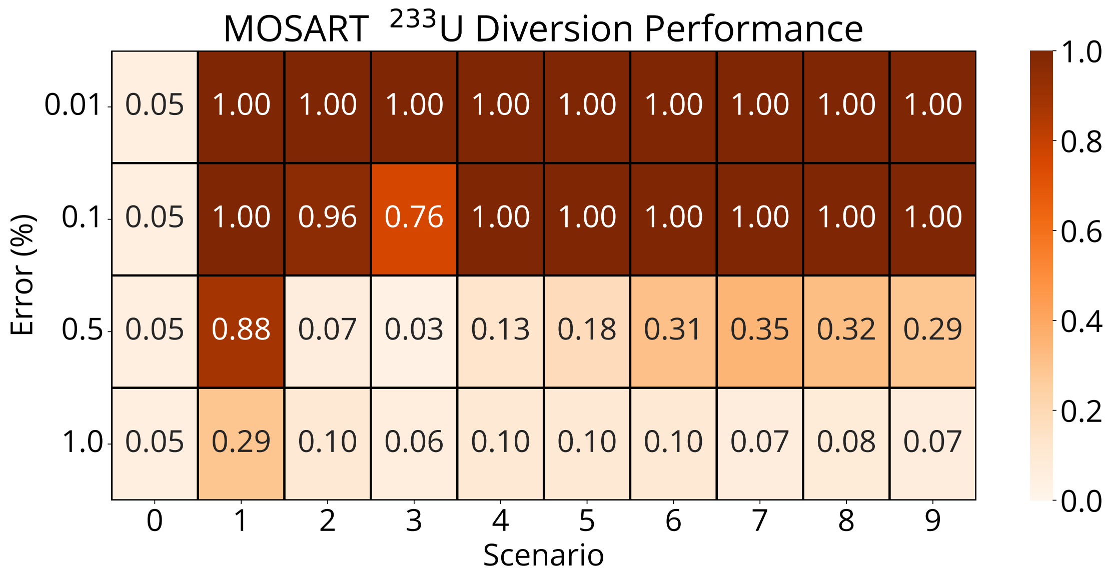


Figure 6-2. Probability of detection for several material loss cases conducted on the simulated MOSART primary salt

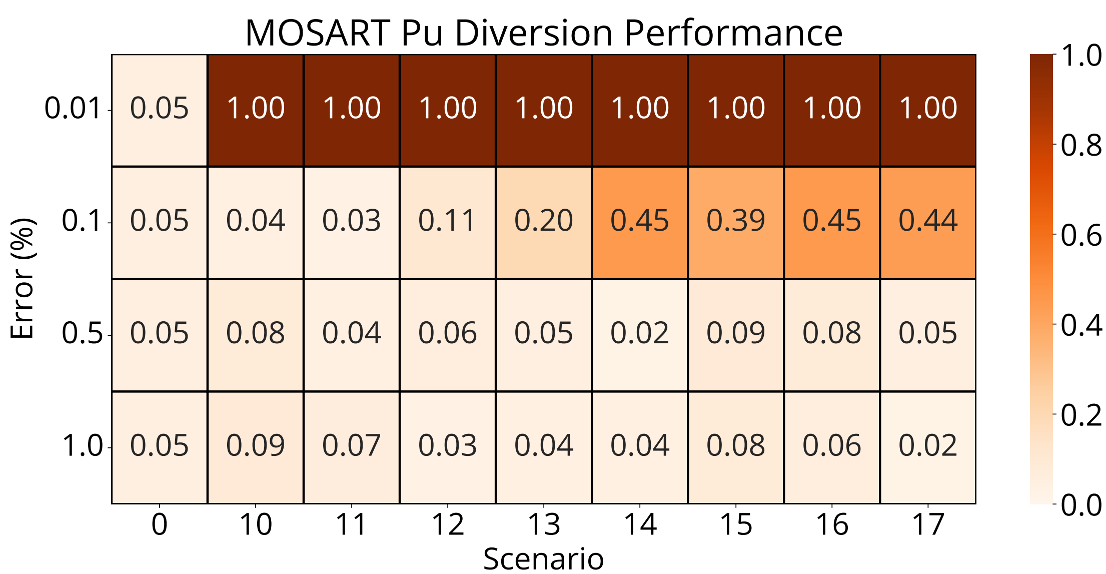


Figure 6-3. Probability of detection for several material loss cases conducted on the simulated MOSART primary salt

6.3.3. REBUS

The REBUS design is a large, single fluid U/Pu cycle reactor. Losses from the REBUS primary salt are focused on bulk removals of plutonium, as described in Table 6-5.

Probabilities of detection are shown in Figure 6-4 below based on Page's trend test on SITMUF. The performance for REBUS scenarios is generally poor for all feasible levels of measurement error and even struggle in a few cases for the extremely low measurement error. This poor performance can be attributed to the relatively large plutonium inventory (1000s of kgs) compared to removals of only a few significant quantities.

Scenario	Loss target[1]	Replacement material	Target quantity (kg)	Loss duration (xMBP)	Start time (yr since startup)
1	Total Pu	^{238}U	8	1	1
2	Total Pu	^{238}U	8	1	5
3	Total Pu	^{238}U	8	1	10
4	Total Pu	^{238}U	16	1	20
5	Total Pu	^{238}U	24	1	10
6	Total Pu	^{238}U	32	1	10
7	Total Pu	^{238}U	32	2	10
8	Total Pu	^{238}U	32	3	10
9	Total Pu	^{238}U	32	4	10

Table 6-5. REBUS loss summary

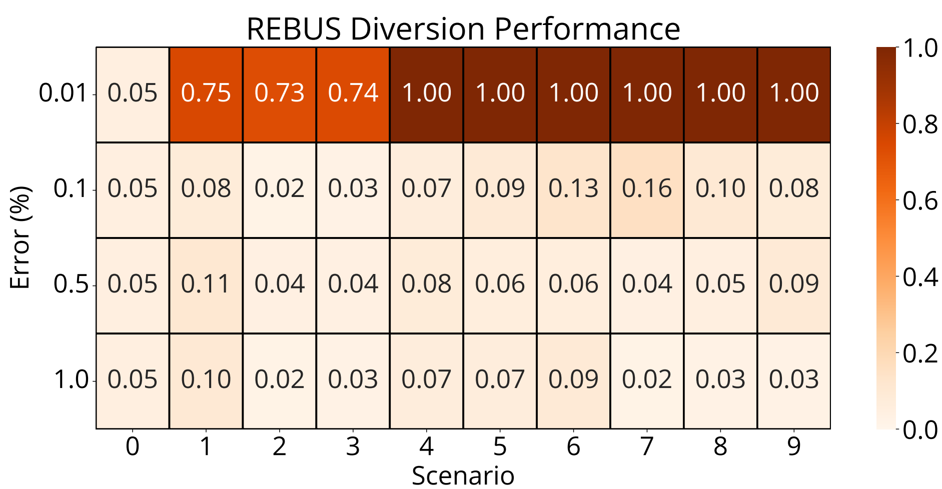


Figure 6-4. Probability of detection for several material loss cases conducted on the simulated REBUS primary salt

[1]Quantities denoted (total) indicate that the target quantity was split over several species based on their proportions in the target inventory.

6.3.4. MSFR

The MSFR is a two-fluid design with a Th/U fuel cycle. Normal operation consists of ^{233}U breeding in the blanket, which is then transferred to the fuel salt for power production. Here, ^{233}U is the fissile element of interest that is removed for loss scenario analysis. Losses from both the fuel and blanket salts were considered, however, due to the low equilibrium inventory levels of ^{233}U in the blanket, ^{233}Pa was diverted instead. Given the low fissile inventories in the blanket, the fuel salt was the main focus of this analysis. The scenarios considered in this analysis are summarized in Table 6-6.

The results of the losses are shown below in Figure 6-5. For convenience, the fuel salt results are highlighted with a yellow border whereas the blanket salt results are highlighted in a green border. Similar to other designs considered thus far in this work, the MSFR performs poorly (i.e., low probability of detection) for most of the realistic error cases.

The MSFR exhibits one of the interesting properties of multi-fluid MSR systems. In this work, it was decided that multi-fluid systems could comprise a single MBA as the salt loops are likely to reside in close physical proximity. However, considering both fluids together can conceal large abrupt losses due combining both inventories. For example, the blanket salt losses remove nearly all of the inventory for the loss target (i.e., almost all ^{233}Pa is removed from the blanket). However, this is concealed by also adding in the fuel salt inventory, which is much larger than the blanket inventory and simulated loss magnitude.

Scenario	Location	Loss target[1]	Replacement material	Target quantity (kg)	Loss duration (xMBP)	Start time (yr since startup)
1	Fuel salt	^{233}U	^{232}Th	8	1	1
2	Fuel salt	^{233}U	^{232}Th	8	1	5
3	Fuel salt	^{233}U	^{232}Th	8	1	10
4	Fuel salt	^{233}U	^{232}Th	16	1	20
5	Fuel salt	^{233}U	^{232}Th	24	1	10
6	Fuel salt	^{233}U	^{232}Th	32	1	10
7	Fuel salt	^{233}U	^{232}Th	32	2	10
8	Fuel salt	^{233}U	^{232}Th	32	2	10
9	Fuel salt	^{233}U	^{232}Th	32	3	10
10	Blanket salt	^{233}Pa	^{232}Th	4	1	1
11	Blanket salt	^{233}Pa	^{232}Th	4	1	5
12	Blanket salt	^{233}Pa	^{232}Th	4	1	10
13	Blanket salt	^{233}Pa	^{232}Th	4	2	10

Table 6-6. MSFR loss summary

[1] Quantities denoted (total) indicate that the target quantity was split over several species based on their proportions in the target inventory.

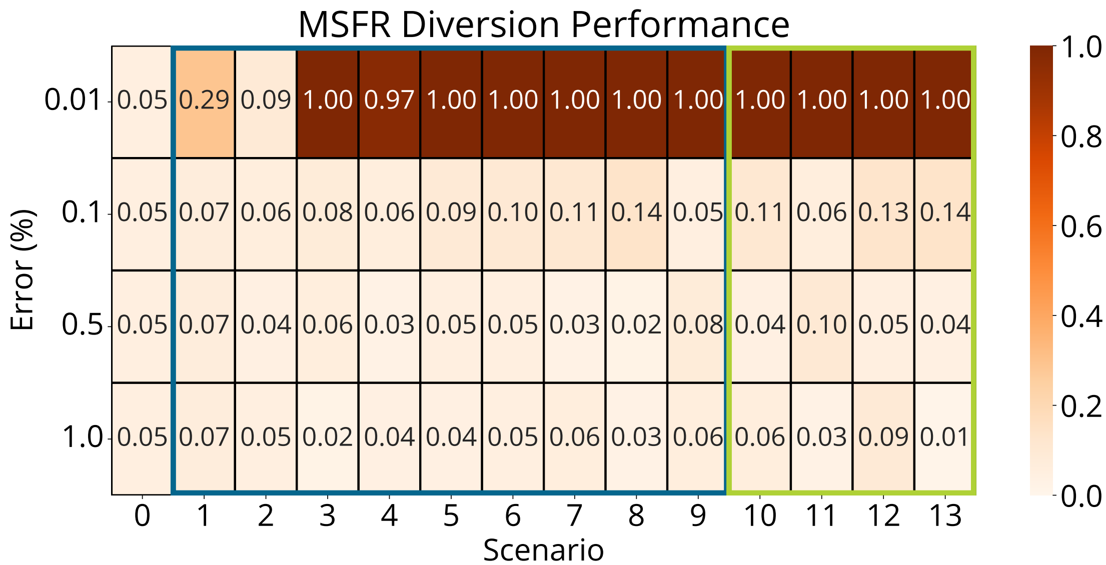


Figure 6-5. Probability of detection for several material loss cases conducted on the simulated MSFR fuel and blanket salts; fuel salt losses are highlighted in blue whereas blanket salt losses are highlighted in green

6.3.5. MSCFR

The MSCFR is the final design considered in this work and is a large, two-fluid design based on the U/Pu fuel cycle. Plutonium is largely bred in the blanket salt after which it is transferred to the primary fuel salt to drive the reactor. Similar to the MSFR, the MSCFR has low equilibrium inventories of actinides in the blanket salt. As such, only two blanket losses were considered for this analysis. A summary of loss scenarios is provided in Table 6-7 below.

Results are summarized in Figure 6-6. Again, fuel salt losses are highlighted with a blue border whereas blanket salt losses are highlighted with a green border. losses from both salts are undetectable using reasonable error rates, and even with the extremely low uncertainty error set (i.e., 0.01% errors), some cases are difficult to detect. This stems from the large inventory of plutonium in the MSCFR. Although plutonium has no significant inventory changes over the salt lifetime, the large inventory leads to a SEID that is much larger than the material losses considered.

Scenario	Location	Loss target[1]	Replacement material	Target quantity (kg)	Loss duration (xMBP)	Start time (yr since startup)
1	Fuel salt	Total Pu	^{238}U	8	1	1
2	Fuel salt	Total Pu	^{238}U	8	1	5
3	Fuel salt	Total Pu	^{238}U	8	1	10
4	Fuel salt	Total Pu	^{238}U	16	1	20
5	Fuel salt	Total Pu	^{238}U	24	1	10
6	Fuel salt	Total Pu	^{238}U	32	1	10
7	Fuel salt	Total Pu	^{238}U	32	2	10
8	Fuel salt	Total Pu	^{238}U	32	3	10
9	Fuel salt	Total Pu	^{238}U	4	32	10
10	Blanket salt	Total Pu	^{238}U	6	1	5
11	Blanket salt	Total Pu	^{238}U	6	1	10

Table 6-7. MSCFR loss summary

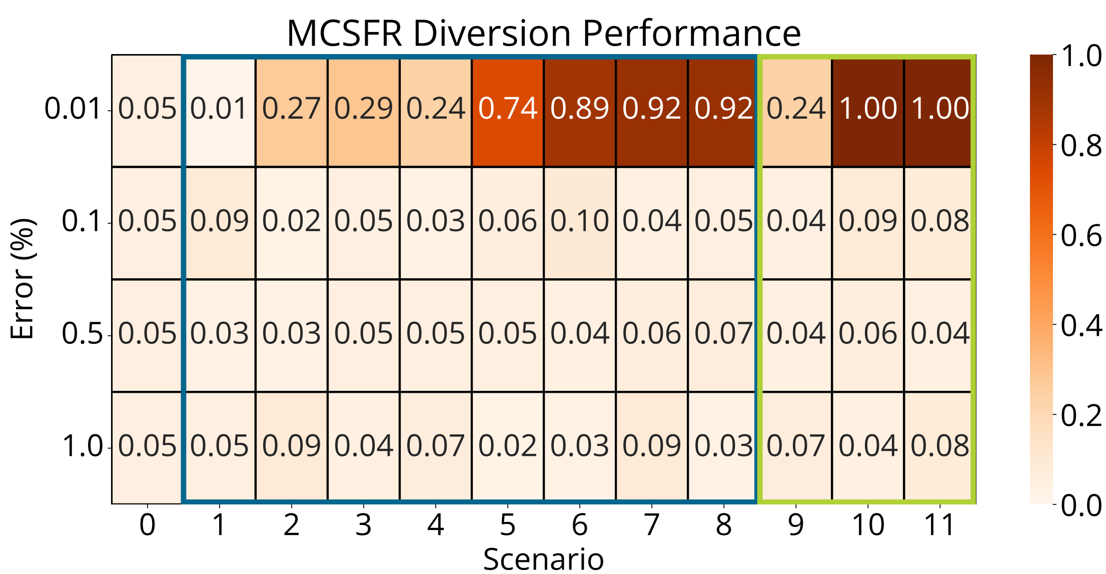
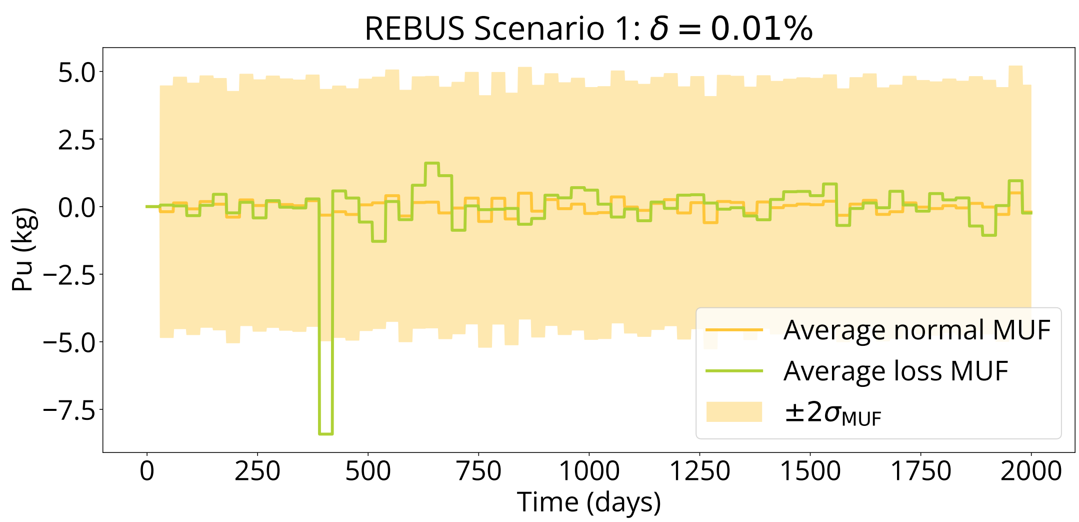


Figure 6-6. Probability of detection for several material loss cases conducted on the simulated MCSFR fuel and blanket salts; fuel salt losses are highlighted in blue whereas blanket salt losses are highlighted in green

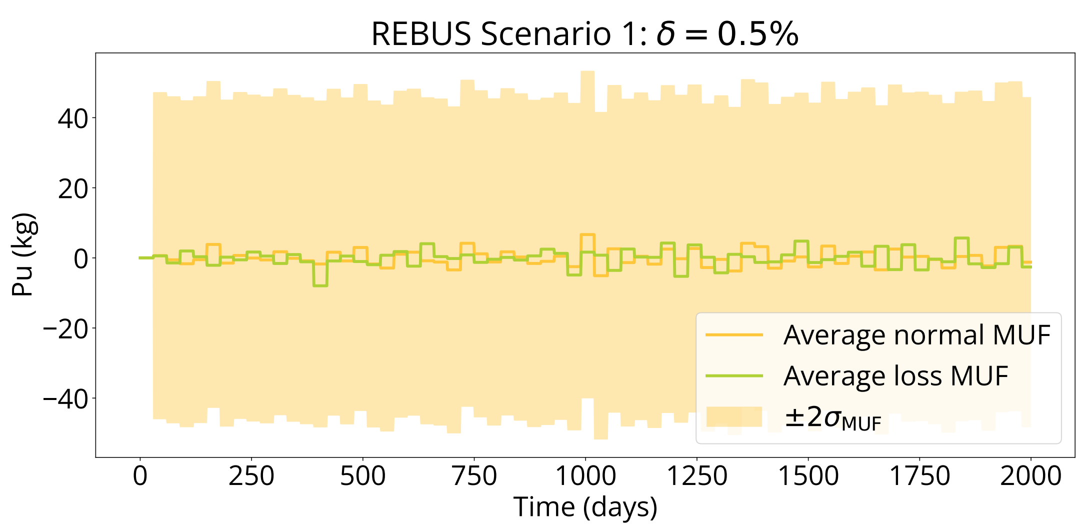
[1]Quantities denoted (total) indicate that the target quantity was split over several species based on their proportions in the target inventory.

7. DISCUSSION

Generally, detection of material loss on the order of multiple significant quantities is difficult in MSRs using obtainable measurement errors. The inability to detect these losses are not a reflection of any particular design feature of MSRs themselves. Rather, the inventories of fissile inventory required for these large reactors lead to significant uncertainty in the material balance, even at extraordinarily low measurement uncertainties. For example, consider comparisons of the average MUF sequence and associated SEID for the REBUS design in Figure 7-1 below.



(a)



(b)

Figure 7-1. Visual depiction of material loss in comparison to SEID for REBUS for two different relative standard deviations

NOTE: The material loss seen in the Figure 7-1a is undetectable in Figure 7-1b wherein the measurement uncertainty is much larger than the 8kg loss of material.

A comparison of SEID for all designs considered in this work is presented in Figure 7-2 below. Designs shown in Figure 7-2 have different nominal thermal power, so some difference in SEID between designs is expected given the variation in reactor size. Regardless of design, large fissile inventories create a challenging environment to detect material loss using statistical testing on material accountancy data alone.

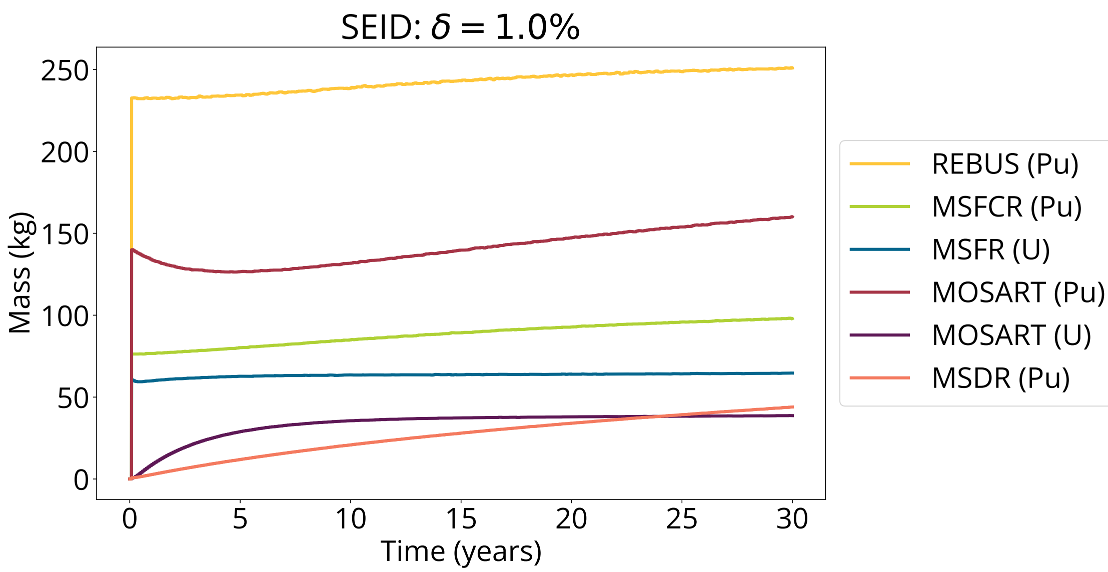


Figure 7-2. Nominal SEID liquid-fueled MSR designs.

The impact of a large fissile inventory is described more concretely in Table 7-1 where the average nominal SEID is reported for materials of interest for each of the designs considered. The lower limit of detection (LLD) (e.g., the required SEID for a 95% detection probability) is also provided (see Appendix A for further discussion). It is statistically impossible to detect even 10 SQs at high confidence levels in some scenarios when using DA-level measurement errors.

Design	Design Power (MW_{th})	Material	Average nominal SEID at 1.0% uncertainty (kg)	Lower limit of detection at 1.0% uncertainty (kg)
MSDR	750	Total Pu	26.11	85.65
MOSART	2400	^{233}U	33.60	110.22
MOSART	2400	Total Pu	141.164	463.02
MSFR	3000	^{233}U	63.20	207.29
REBUS	3700	Total Pu	242.08	794.05
MSCFR	6000	Total Pu	87.97	288.56

Table 7-1. Lower limits of detection based on current SEID values

Reactor design plays an important role in determining the fissile inventory. Figure 7-3 shows the power normalized SEID for the designs considered in this work. Fissile inventory generally scales with thermal design power, however, some designs have lower fissile inventory per unit power than others. The MSFCR has the largest thermal power design at 6000 MW_{th}, but has a SEID that is lower than both the 3700 MW_{th} REBUS and 2400 MW_{th} MOSART.

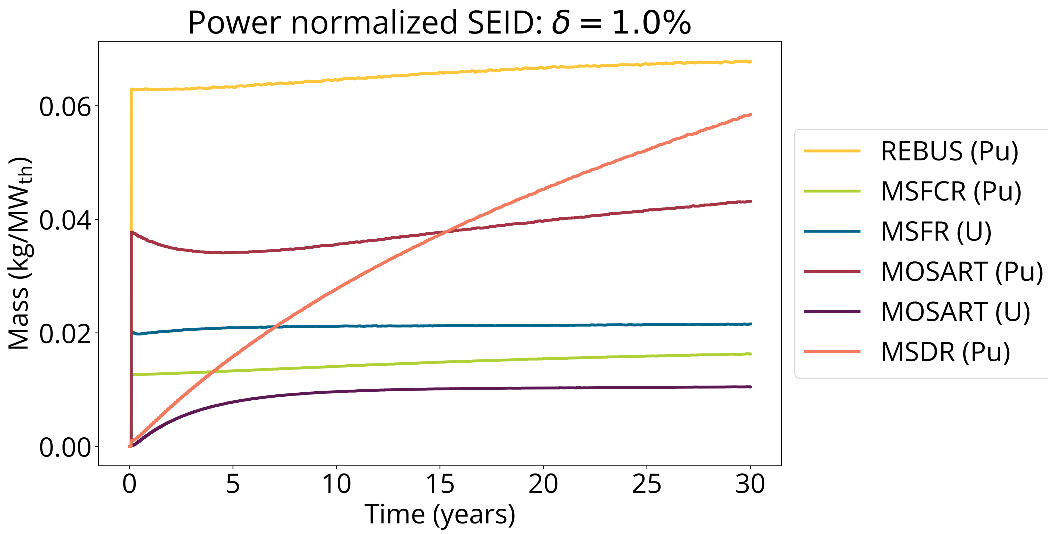


Figure 7-3. Power normalized SEID for various reactor designs.

The growth in fissile inventory for liquid-fueled MSRs also causes challenges for setting thresholds used for Page's trend test. A static set of h, k values are used to evaluate each material loss. These values are tuned to provide roughly a 5% false alarm probability per year regardless of the salt lifetime. This results in a somewhat uneven distribution of false alarms (i.e. more false alarms later in the salt lifetime) (shown in Figure 7-4 for the MSDR) which would also impact the probability of detection for later material losses.

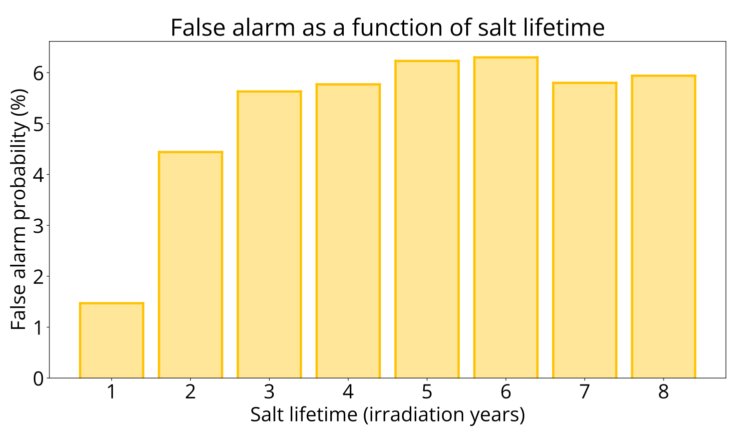


Figure 7-4. False alarm probability as a function of salt lifetime at the 1% uncertainty level for 8 operational years of the MSDR.

There are some potential statistical remedies that may be available, however, these are not explored in depth in this document. These include further optimization of threshold criteria (for Page's trend test), analysis of separate material balances for multiple salt streams, and optimization of material balance time. Given the large inventories, these are not anticipated to have a significant impact on the probability of detection or SEID relative to current values. In fact, these results are likely optimistic as they neglect the potentially significant modeling related errors that would occur during practice (e.g., imprecise knowledge of the MSR system leading to a suboptimal simulation or missing knowledge about chemical interactions).

This work does not discuss alternative, non-statistical solutions to the lack of statistical sensitivity to material loss. For example, the concentration of actinides compared to the total fuel salt (which could be quite low) and self-protecting nature of the salt are not considered. These results are similar to that of an entire LWR core as they often contain large inventories of security relevant materials. However, current MC&A for LWRs use a combination of C/S with periodic verification of some spent fuel to implement effective MC&A.

Although MSRs do have flowing fuels that are somewhat related to existing bulk facilities (e.g., reprocessing and enrichment), the most likely conventional MC&A scheme in the near future (i.e., excluding potential R&D advances like process monitoring) would involve consideration of features stated above while employing strong C/S methods.

8. CONCLUSIONS

This work described the application of traditional material accountancy statistics to several MSR designs. Detection of many loss scenarios were shown to be impossible to detect using traditional methods. This largely arose from the sizeable inventory of material proposed in these designs. Additional sources of uncertainty arising from simulation imperfections could also increase SEID in real world scenarios.

MSRs have features of both bulk facilities and LWRs. The most logical choice for a material accountancy system for the MSR core itself would be based on principles used for bulk facilities. However, bulk facilities do not have constant depletion occurring in inventory. LWRs do have constant depletion, but benefit from fuel that is contained in discrete items. The large bulk inventory of fissile inventory in the MSR is the largest contributor the relatively high material balance uncertainty which drives poor loss detection when relying exclusively on material accountancy of the core and traditional statistical tests.

Given the limitations of contemporary statistical methods for MC&A in the face of large inventory uncertainties, practical near-term solutions for safeguarding MSRs will require consideration of realities of material diversion (e.g., concentration of fissile isotopes and radiation intensity) while employing effective C/S strategies. Process monitoring could also play an important role in an effective MC&A system for liquid-fueled MSRs. Use of accountancy tanks and rigorous monitoring of input and output flows from the core could also improve the overall MC&A system.

REFERENCES

[1] Michael P. Dion and Karen K. Hogue. Domestic mc&a recommendations for liquid-fueled msrs. *ORNL/SPR-2022/2673*, September 2022.

[2] Third Way. 2019 advanced nuclear map.

[3] Michael P. Dion, M. Scott Greenwood, Karen K. Hogue, Sean E. O'Brien, Logan M. Scott, and Greg T. Westphal. Mc&a for msrs: Fy2021 report. *ORNL/SPR-2021/2305*, September 2021.

[4] Benjamin R. Betzler, Jeffery J. Powers, and Andrew Worrall. Fuel cycle and neutronic performance of a spectral shift molten salt reactor design. *Annals of Nuclear Energy*, 2017.

[5] Benjamin R. Betzler, Sean Robertson, Eva E. Davidson (nee Sunny), Jeffery J. Powers, Andrew Worrall, Leslie Drewan, and Mark Massie. Fuel cycle and neutronic performance of a spectral shift molten salt reactor design. *Annals of Nuclear Energy*, 2018.

[6] Pedro Jr Vicente Valdez, Benjamin R. Betzler, William Wieselquist, and Massimilliano Fratoni. Modeling molten salt reactor fission product removal with scale. 3 2020.

[7] Jin Whan Bae, Benjamin R. Betzler, and Andrew Worrall. Molten salt reactor neutronic and fuel cycle sensitivity and uncertainty analysis. 11 2019.

[8] Andrei Rykhlevskii, Benjamin R. Betzler, Andrew Worrall, and Kathryn D. Huff. Fuel cycle performance of fast spectrum molten salt reactor designs. 8 2019.

[9] Benjamin R. Betzler, Andrei Rykhlevskii, Andrew Worrall, and Katheryn Huff. Impacts of fast-spectrum molten salt reactor characteristics on fuel cycle performance. 9 2019.

[10] Andre Vidal Soares, Branko Kovacevic, Amanda Johnsen, Azaree Lintereur, Benjamin Betzler, Michael Dion, Nathan Shoman, and William Walters. The impact of nuclear data uncertainty on identifying plutonium diversion in liquid-fueled molten salt reactors. *Nuclear Physics B*, 2022 (in-press).

[11] TP Speed and D Culpin. The role of statistics in nuclear materials accounting: issues and problems. *Journal of the Royal Statistical Society: Series A (General)*, 149(4):281–300, 1986.

[12] Aaron S Goldman, Richard R Picard, and James P Shipley. Statistical methods for nuclear materials safeguards: an overview. *Technometrics*, 24(4):267–274, 1982.

[13] Aaron Goldman, Richard Picard, and James Shipley. Statistical methods for nuclear materials safeguards: An overview. *Technometrics*, 24(4):267–275, 1982.

[14] James Doyle. *Nuclear safeguards, security and nonproliferation*. Butterworth-Heinemann, Oxford, England, Jun 2008.

[15] Barry Jones. Near real time material accountancy using SITMUF and a joint page's test: comparison with MUF and CUMUF tests, 1988.

[16] B Jones. Calculation of diversion detection using the sitmuf sequence and page's test: application to evaluation of facility designs. In *Proceedings of the 7th ESARDA Symposium on Safeguards and Nuclear Material Management*, 1985.

[17] BJ Jones. Calculation of diversion detection using the sitmuf sequence and page's test. In *Nuclear safeguards technology 1986*. 1987.

[18] B Jones. Comparison of near real time materials accountancy using sitmuf and page's test with conventional accountancy. In *Proceedings of the 9th ESARDA Symposium on Safeguards and Nuclear Material Management*, 1987.

[19] B Jones. Near real time materials accountancy using sitmuf and a joint pages test: improvement of the test. *ESARDA Bulletin*, 16:13–19, 1989.

[20] E. S. Page. Continuous inspection schemes. *Biometrika*, June 1954.

[21] Tom Burr and M. S. Hamada. Revisiting statistical aspects of nuclear material accounting. *Science and Technology of Nuclear Installations*, March 2013.

[22] R Seifert. *The GEMUF test: A new sequential test for detecting loss of material in a sequence of accounting periods*. IAEA, 1987.

[23] Jaakko Leppänen. *Serpent: a Continuous-energy Monte CarloReactor Physics Burnup Calculation Code*, 18 edition, June 2015.

[24] Andrei Rykhlevskii, Jin Whan Bae, and Kathryn D. Huff. Modeling and simulation of online reprocessing in the thorium-fueled molten salt breeder reactor. *Annals of Nuclear Energy*, 128:366–379, 2019.

[25] Luke Seifert, Alexander Wheeler, and Ondrej Chvála. Material flows and on-line reprocessing in serpent, November 2021.

[26] M. Scott Greenwood, Benjamin R. Betzler, A. Lou Qualls, Junsoo Yoo, and Cristian Rabiti. Demonstration of the advanced dynamic system modeling tool transform in a molten salt reactor application via a model of the molten salt demonstration reactor. *Nuclear Technology*, 206(3):478–504, 2020.

[27] Nicholas J. Dunkle. Dynamic modeling of thermal spectrum molten salt reactors, 2022.

[28] Nathan Shoman and Michael Higgins. Fy21 final report on molten salt reactor safeguards modeling. *SAND Report*, SAND2021-10928R, 2021.

[29] Nathan Shoman and Michael Higgins. Fy22 final report on molten salt reactor safeguards modeling. *SAND Report*, SAND2022-11048O, 2022.

[30] Branko Kovacevi, Andre Vidal Soares, Azaree Lintereur, William Walters, and Amanda Johnsen. Gamma-ray signatures for identifying plutonium diversion in molten salt reactors. *Nuclear Physics B*, 2022 (in-press).

[31] E. S. Bettis, L.G. Alexander, and H. L. Watts. Design studies of a molten-salt reactor demonstration plant. *ORNL-TM-3832*, June 1972.

[32] V Ignatiev, O Feynberg, I Gnidoi, A Merzlyakov, V Smirnov, A Surenkov, I Tretiakov, R Zakirov, V Afonichkin, A Bovet, V Subbotin, A Panov, A Toropov, and A Zherebtsov. Progress in development of li,be,na/f molten salt actinide recycler and transmuter concept, Jul 2007.

[33] A. Mourogov and P.M. Bokov. Potentialities of the fast spectrum molten salt reactor concept: Rebus-3700. *Energy Conversion and Management*, 47(17):2761–2771, 2006. 12th International Conference on Emerging Nuclear Energy Systems.

[34] Centre National De La Recherche Scientifique (CNRS). Final report - evol (evaluation and viability of liquid fuel fast reactor system), 2010.

[35] R. C. Asher, G. Long, H. A. C. McKay, and D. L. Reed. An assessment of a 2500 mwe molten chloride salt fast reactor, 1974.

[36] K. Zhao, M. Penkin, C. Norman, and S. Balsley. *International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials*, December 2012.

[37] International Atomic Energy Agency. *IAEA Safeguards Glossary, 2022 Edition*.

[38] R.C. Robertson. MSRE design and operation report, part I, description of reactor design. *ORNL-TM-0728*, 1965.

[39] W. Wieselquist, M. Williams, D. Wiarda, M. Pigni, and U. Mertyurek. Overview of nuclear data uncertainty in scale and application to light water reactor uncertainty analysis, 2018.

APPENDIX A. LOWER LIMIT OF DETECTION

NOTE: The discussion below is focused on one-sided testing (i.e., testing for material loss only); however, the procedure for determining thresholds for two-sided (i.e., testing for material loss and gain) testing is similar.

Note that the material balance sequence itself is a distribution such that **MUF** $\sim \mathcal{N}(\boldsymbol{\mu}, \boldsymbol{\Sigma})$ where $\boldsymbol{\mu}$ is some mean and $\boldsymbol{\Sigma}$ is the covariance matrix that is determined by measurement error. It follows that a single material balance instance at a specified material balance is also defined by a distribution such that MUF $\sim \mathcal{N}(\mu, \sigma_{\text{MUF}})$. Under loss conditions, the mean of MUF will shift based on the magnitude of the loss such that $\text{MUF}_{\text{loss}} = \mathcal{N}(\mu^*, \sigma_{\text{MUF}})$. This is illustrated below in Figure A-1.

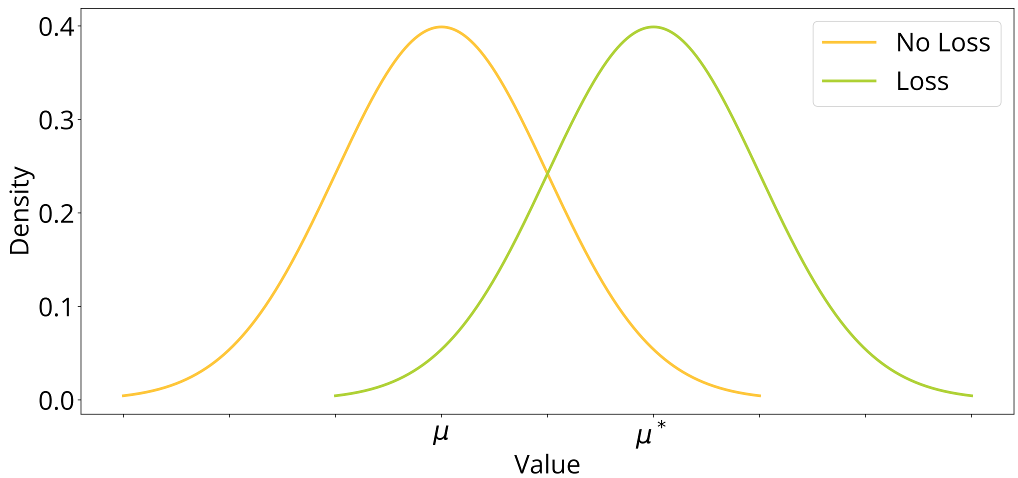


Figure A-1. Shift in MUF distribution due to material loss

One common goal by the IAEA is to set system requirements such that the probability of detection for a material loss be 95% with a false alarm probability of 5%. This is often calculated using Page's trend test on SITMUF. However, a lower limit of detection probability can be established that relates a mean shift due to a material loss to SEID. These constraints will be expressed as follows:

$$\begin{aligned} P(x > h \mid \mathcal{N}(\mu, \sigma_{\text{MUF}})) &\leq 0.05 \\ P(x > h \mid \mathcal{N}(\mu^*, \sigma_{\text{MUF}})) &\geq 0.95 \end{aligned} \tag{A.1}$$

Where h denotes some threshold, μ is the average MUF under normal conditions, and μ^* is the average MUF under loss conditions. For simplicity, assume that $\mu = 0$ and $\sigma_{\text{MUF}} = 1$. This leads to an updated set of constraints that can be used to develop a relationship between μ^* and SEID:

$$P(x > h \mid \mathcal{N}(0,1)) \leq 0.05 \tag{A.2}$$

$$P(x > h \mid \mathcal{N}(\mu^*, 1)) \geq 0.95 \tag{A.3}$$

Specifically note the normal cumulative distribution function and normal quantile function:

$$F(x) = \Phi\left(\frac{x-\mu}{\sigma}\right) = \frac{1}{2}\left[1 + \operatorname{erf}\left(\frac{x-\mu}{\sigma\sqrt{2}}\right)\right] \tag{A.4}$$

$$F^{-1}(p) = \mu + \sigma\Phi^{-1}(p) = \mu + \sigma\sqrt{2}\operatorname{erf}^{-1}(2p-1),\ p \in (0,1) \tag{A.5}$$

First, determine h by combining the constraint in Equation A.2 with the expression for the quantile function in Equation A.5 to find $h = F^{-1}(0.95) \approx 1.64$ for $\mathcal{N}(0,1)$.

Next, use the constraint from Equation A.4, the expression for the quantile function in Equation A.5, and the previously determined value for $h \approx 1.65$. Solving Equation A.5 as $F^{-1}(p = 0.05; \sigma_{\text{MUF}} = 1) = 1.65$ for μ^* leads to $\mu^* \approx 3.28$.

An expression for the relationship between μ^* and SEID subject to the general performance constraints often set by the IAEA is as follows:

$$\begin{aligned}
F^{-1}(p) = \mu + \sigma\Phi^{-1}(p) &= \mu + \sigma\sqrt{2}\operatorname{erf}^{-1}(2p-1),\ p \in (0,1) \\
F^{-1}(p = 0.95|\mathcal{N}(0,\sigma_{\text{MUF}})) &= F^{-1}(p = 0.05|\mathcal{N}(\mu^*,\sigma_{\text{MUF}})) \\
1.64\sigma_{\text{MUF}} &= \mu^* - 1.64\sigma_{\text{MUF}} \\
3.28\sigma_{\text{MUF}} &= \mu^* \\
\sigma_{\text{MUF}} &= \frac{\mu^*}{3.28}
\end{aligned} \tag{A.6}$$

Equation A.6 refers to the case of fixed probabilities, it can be expanded to a more general case of $\sigma_{\text{MUF}} \leq \frac{\mu^*}{3.28}$ by nothing that $F^{-1}(p|\mathcal{N}(0,\sigma_1) \leq F^{-1}(p|\mathcal{N}(0,\sigma_2)$ where $\sigma_1 < \sigma_2$.

APPENDIX B. MATERIAL BALANCE FORMULATION

There are two possible material balances as follows (subscript m denotes a measured inventory whereas subscript c denotes a calculated inventory that is estimated via computational means):

1. MUF = $I_{m,t} - I_{c,t}$
2. MUF = $(I_{m,t} - I_{m,t-1}) - (I_{c,t} - I_{c,t-1})$

Note that the second option was selected as the optimal expression. Although not described at length here, the equations governing the variance of the first material balance approach was also developed. A direct comparison using identical parameters (i.e. same errors) was performed in Figure B-1 below.

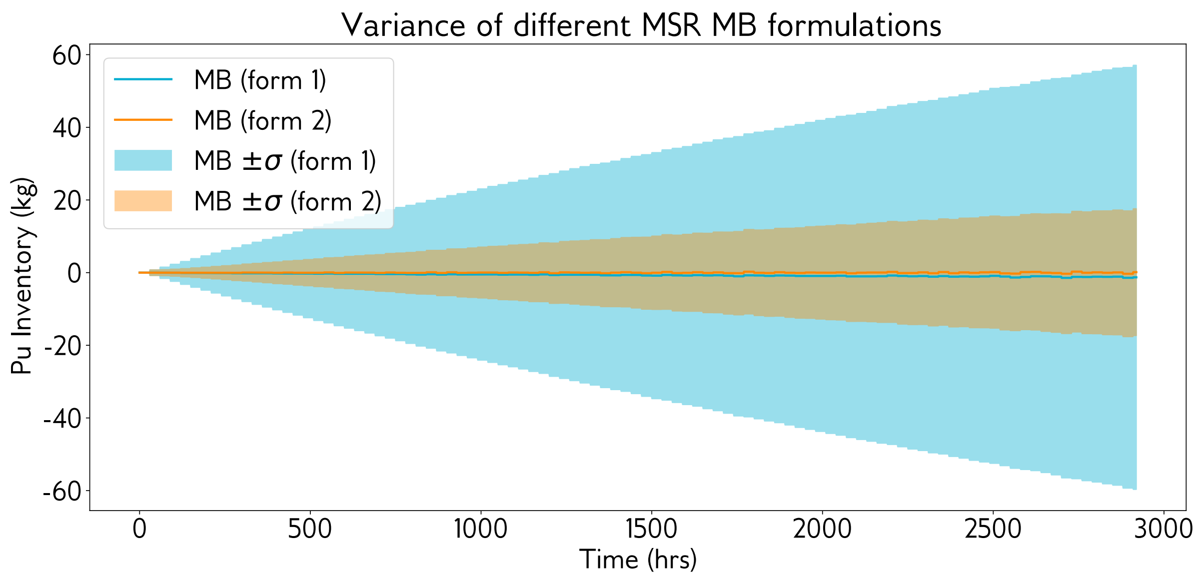


Figure B-1. Comparison between material balance formulations

Note that for the same errors the second formulation (i.e. $(I_{m,t} - I_{m,t-1}) - (I_{c,t} - I_{c,t-1})$) exhibits a much lower variance than the first formulation. This can be explained by considering the variance of each material balance. The first formulation is the simple difference between the measured and calculated inventories. The variance for an individual run could be large due as a result of the systematic errors. The second formulation minimizes the impact of the systematic error by considering *relative* differences. A given inventory from time t and $t-1$ are assumed to have a systematic bias which results in very little impact on the material balance variance.

APPENDIX C. EXEMPLAR DESIGN INFORMATION

C.1. MSDR

The MSDR [31] conceptual design by ORNL is semi-commercial in scale (750 MW_{th} / 350 MW_e) and was intended to demonstrate technologies developed during the molten salt reactor experiment [38]. The reactor featured a fluoride salt and was originally intended to be a thorium breeder. However, for the purpose of this work a LEU (5%) fluoride-based fuel salt is considered instead [5]. Note that the choice of fuel salt will impact some MC&A conclusions as the actinide inventory is directly tied to the neutron flux (i.e. thermal vs fast) and fuel choice.

The MSDR has several properties common to most MSRs such as continuous feed of fuel material and fission product removal. A constant stream of LEU (assumed to be in powder form so that system mass largely remains constant) is added during operation while noble metals are removed by a cold trap and gaseous fission products are sparged. Most of the more volatile fission products are left in the primary salt loop which was designed to be replaced every 8 years. The simulated MSDR unit cell is shown below in Figure C-1.

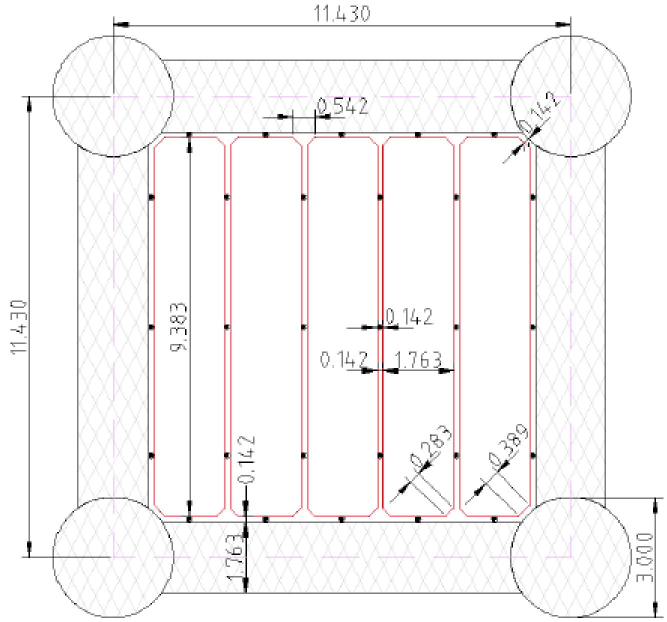


Figure C-1. MSDR unit cell as reproduced in [26]

MOSART

The primary goal of the MOlten Salt Actinide Recycler & Transmuter (MOSART) is to reduce long lived waste in LWR spent fuel without U-Th support materials and while also producing electricity. This 2400 MW_t design has a LiF-BeF_2 carrier salt and single (TRU/Th)F_4-BeF_2-LiF fuel salt that serves as both the fissile and fertile material [32]. The reactor core consists of a cylindrical core with no solid material present inside to serve as moderator. Fuel salt enters the core through a radial inlet in the bottom of the core and salt flows upward through natural circulation before leaving through an outlet pip on the top reflector. An example of the MOSART fuel loop is shown in figure C-2. The MOSART three key transient operations that must be modeled in SCALE: (1) ^{232}Th is fed as feed material, (2) LWR TRU as feed material, and (3) continuous salt treatment to remove fission product gasses and noble metals.

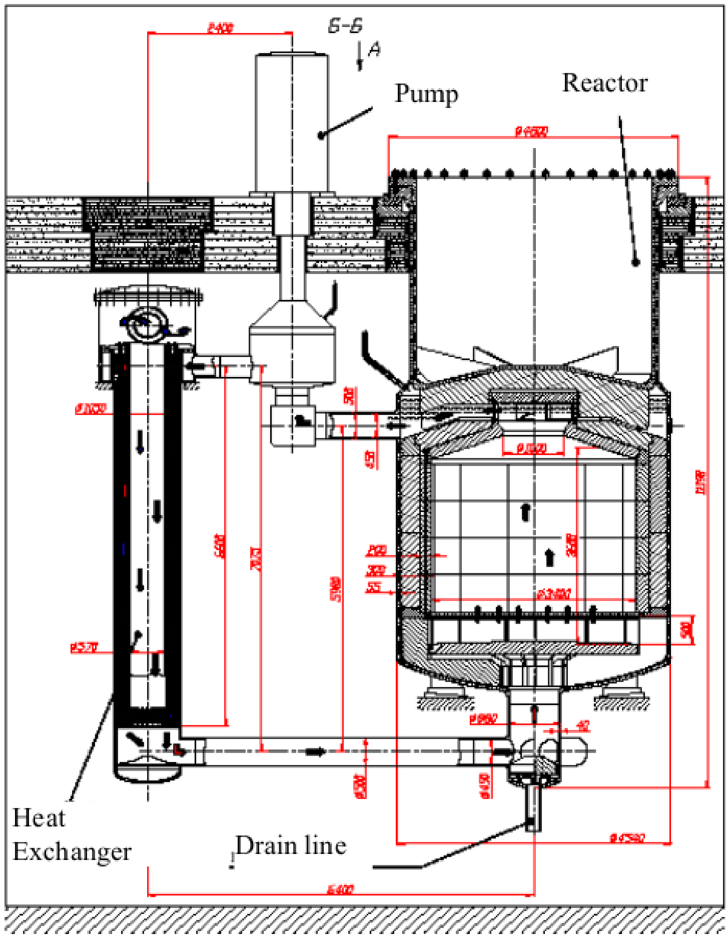


Figure C-2. MOSART core and primary loop [32]

REBUS

The REBUS design is a 3700 MW_t designed with a focus on sustainability, safety, economics, and non-proliferation [33]. The single-fluid reactor has a $(U/TRU)Cl_3$-NaCl fuel salt that contains both fissile and fertile material. Several lessons learned from the Molten Salt Breeder Reactor (MSBR) are incorporated to reduce technical design risk, including ratio of core to system volume. Similar to MOSART, the REBUS core consists of a metal-reflected cylindrical vessel. A simplified flow sheet for the REBUS design is shown in Figure C-3. There are only two key continuous operations for REBUS: (1) continuous natural uranium feed and (2) salt processing to remove gaseous fission products and noble metals.

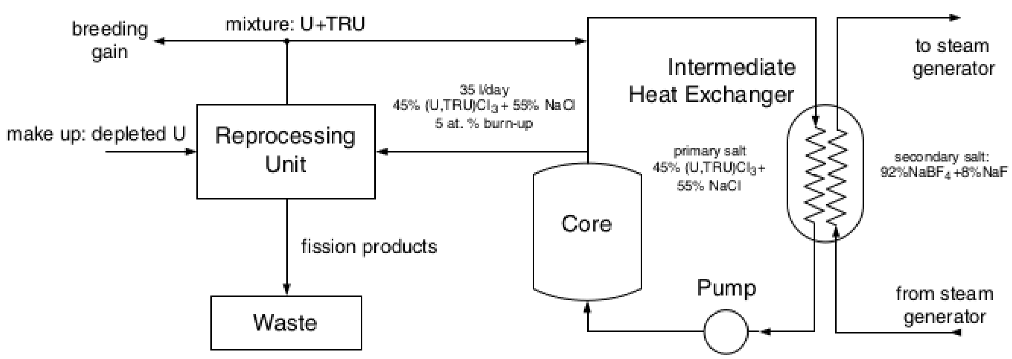


Figure C-3. REBUS core and primary loop [33]

MSFR

The Molten Salt Fast Reactor (MSFR) is a fast spectrum reactor with a thorium fuel cycle designed to take advantage of R&D developed during the MSRE [34, 38]. The 3000 MW_t design consists of a flowing UF_4-ThF_4-LiF fuel salt and a stationary ThF_4-LiF blanket salt. The reactor can be initially operated with either a ^{233}U-based salt or using a LWR TRU-based salt. The static blanket salt is used to shield secondary components from neutrons while breeding ^{233}U from ^{232}Th. A simplified schematic of the core design is shown in Figure C-4. There are three continuous operations for the MSFR including: (1) ^{232}Th feed to the blanket, (2) blanket separation options to feed fuel salt with ^{233}U, and (3) fuel salt processing to remove gaseous fission products and noble metals.

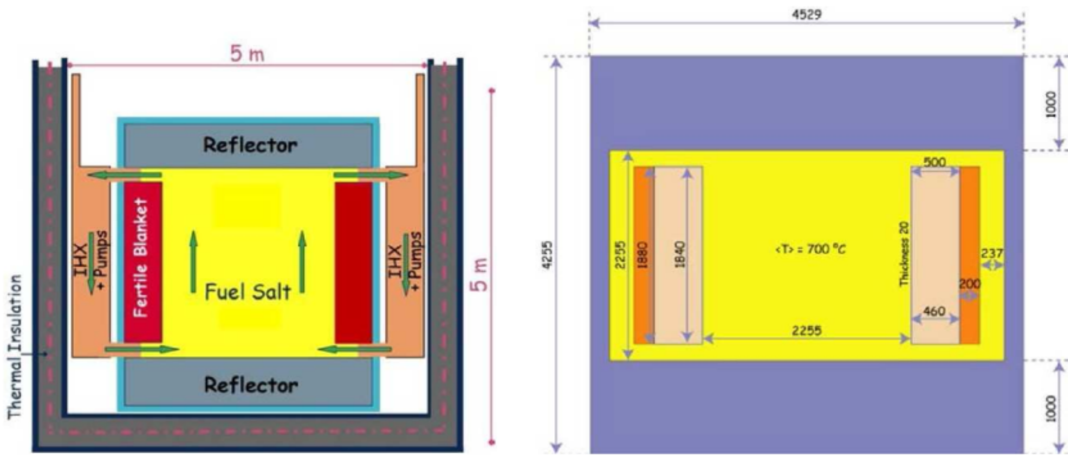


Figure C-4. Diagrams depicting the MSFR core design. Colors on right denote fuel salt (yellow), fertile salt (pink) and B_4C moderator (orange) and Ni-based structural material (blue) [34]

MCSFR

The Molten Chloride Salt Fast Reactor (MCSFR), which was designed by the United Kingdom Atomic Energy Authority in the mid 1970s, is a large 6000 MW_t design that consists of two NaCl-based salts [35]. Specifically, the fuel salt is a flowing (U/Pu)Cl_3 whereas the flowing blanket is UCl_3-NaCl. The core itself is designed to be an ideal configuration wherein the blanket salt forms several spherical shells around the active fuel salt. A slice of the core geometry is shown in Figure C-5. The MCSFR has three continuous processes: (1) natural uranium is added to both the fuel and blanket salt, (2) plutonium is separated from the blanket and transferred to the fuel salt, and (3) salt treatments remove gaseous fission products and noble metals.

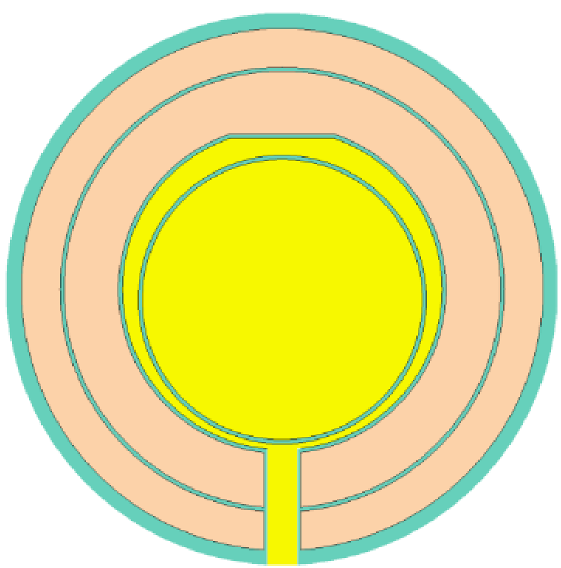


Figure C-5. MCSFR core cross section showing the fuel salt (yellow) and blanket salt (peach) [35]

APPENDIX D. DETAILED METHODOLOGY

D.1. Analytical approach

The MB at any given time t, where t is usually a multiple of the MBP, is a straightforward calculation that is defined in Equation D.1. Here, n refers to the number of locations.

$$\mathrm{MB}_t = \left(\sum_{i=1}^{n_\mathrm{I}} I_{i,t-1} + \sum_{i=1}^{n_\mathrm{in}} Tin_{i,t} - \sum_{i=1}^{n_\mathrm{out}} Tout_{i,t} \right) - \sum_{i=1}^{n_\mathrm{I}} I_{i,t} \tag{D.1}$$

Specific definitions for the terms in Equation D.1 are as follows:

- $\sum_{i=1}^{n_\mathrm{I}} I_{i,t}$ is the total inventory at time t across all locations
- $\sum_{i=1}^{n_\mathrm{in}} Tin_{i,t}$ is the total input at time t across all locations
  - If inputs are flows they should be summed over the time period of interest (i.e. $t-1$ to t)
- $\sum_{i=1}^{n_\mathrm{out}} Tout_{i,t}$ is the total output at time t across all locations
  - Similar to the inputs, if outputs are flows, they should be summed over the time period of interest

The MB is often calculated for each species of interest. For example, a thermal spectrum MSR with a U/Pu fuel cycle would primarily be concerned with Pu, so there would be a single material balance calculated on total Pu[4]. This work considers several different thermal and fast spectrum MSR designs, so the material of interest will vary based on the fuel cycle and design type.

MSRs have a number of unique features, which were discussed in Section 3, that require unique consideration with regards to the MB. As MSRs are undergoing constant nuclear transmutation, the material balance will require modification to include an estimate of actinide changes due to burnup that occurs during normal operation. This calculation will effectively lead to two different inventory terms; one observed inventory derived from direct measurement and a second inventory that is estimated using computational burnup codes. These quantities will never be exactly the same due to a variety of uncertainties (e.g., measurement conditions, incomplete knowledge of core conditions needed for modeling, nuclear data uncertainty)These two terms are defined as follows:

- $I_{m,t}$ - The measured (NDA and/or DA) inventory at time t
- $I_{c,t}$ - The calculated (from burnup code) inventory at time t

[4]Specifics of accountancy measures vary from stakeholder to stakeholder

The inventory terms cannot be directly observed and must be calculated using an estimate of the bulk salt mass and a concentration measurement. It is assumed that the calculated inventory and measured inventory will rely on a shared bulk salt measurement, which should remain approximately constant for fixed volume systems such that the inventory terms are expressed as follows:

- $I_{m,t} = B_{m,t}C_{m,t}$
- $I_{c,t} = B_{m,t}C_{c,t}$

Note that $B_{m,t}$ is the bulk salt measurement at time t whereas $C_{m,t}$ is the measured concentration derived from NDA or DA measurements and $C_{c,t}$ is the calculated concentration based on a burnup code. For MSRs, the most effective formulation is shown below in Equation D.2 (see Appendix B for additional discussion):

$$\text{MUF} = (I_{m,t} - I_{m,t-1}) - (I_{c,t} - I_{c,t-1}) \tag{D.2}$$

Equation D.2 can be extended to develop an expression for SITMUF, which is not included here for brevity[5]. Material balance evaluations performed in this work are conducted by applying Page's trend test to the SITMUF sequence. Specific parameters for Page's trend test are described in following sections, but the reader is referred to referenced material for additional background on SITMUF [15] and Page's trend test [20, 21].

APPENDIX E. DETAILED SETUP INFORMATION

E.1. Baseline calculation

This work considers a wide range of potential material loss scenarios for multiple reactors, and as such, utilization of full-core 3D models would be prohibitively expensive to compute. Instead, representative 2D unit cell models are used.

The thermal reactor (MSDR) unit cell is explicitly based on the fuel channel assembly itself [7] whereas the unit cells for the fast reactors (MSFR, MCSFR, MOSART, REBUS) are generated by simplifying the full-core model. Fast reactor unit cells are created based on a number of constraints relating to the full core model [8, 9]:

1. Fuel-to-fertile salt ratio for each unit cell (u) consistent with full-core model (f)
    a) Applicable for two fluid systems only
    b) $\dfrac{V^f_{\text{core}}}{V^f_{\text{blanket}}} = \dfrac{A^u_{\text{core}}}{A^u_{\text{blanket}}}$
        i. V is volume and A is area

[5] Additional derivations available from authors upon request

2. Unit cell size adjusted for $k_\infty^u \approx k_{\text{eff}}^f$

   a) $k_\infty^u - k_{\text{eff}}^f < 300$ pcm

3. Structural material volume for unit cell adjusted to approximately match neutron energy spectrum shape to full-core spectrum

4. Pearson correlation coefficient r between neutron spectrum of models > 0.995

   a) $r = \dfrac{\sum_{i=1}^{N}(\phi_i^f - \overline{\phi^f})(\phi_i^u - \overline{\phi^u})}{\sqrt{\sum_{i=1}^{N}(\phi_i^f - \overline{\phi^f})^2 \sum_{i=1}^{N}(\phi_i^u - \overline{\phi^u})^2}}$

5. Approximate error in total neutron flux, $\delta < 3\%$

   a) $\delta = \left| \dfrac{\sum_{i=1}^{N}(\phi_i^f - \phi_i^u)}{\sum_{i=1}^{N}\phi_i^f} \right| < 0.03$

All baseline calculations are performed using parameters in Table E-1 below.

Parameter	Value
SCALE sequence	t-depl
Sequence parameters	2region, weight, addnux=4
Cross section library	252-group ENDF/B-VII.1
Total burn duration	30 years
nlib	5/year

Table E-1. SCALE parameters used to generate baseline results

NOTE: Full SCALE input decks are available upon request.

E.2. Material loss calculations

The SCALE code system was primarily designed to support reactor physics analysis for LWRs. As such, there is limited support for analysis of molten salt systems. Although SCALE does support continuous removals, there are some limitations that necessitate the development of supporting scripts to accurately simulate material loss. Most notably, SCALE/TRITON, the sequence responsible for simulating the evolution of actinide inventories over time, tends to discard input nuclides that are irrelevant for neutron transport calculations.

Material loss simulations have a number of parameters that must be incorporated into the reactor physics calculation:

- Elapsed time before initiation of material loss
  - Fissile content in MSRs can vary over time, so it is important to consider a range of material loss initiation times
- Target quantity of material to divert
  - Although there are regulatory targets for existing nuclear facilities, it is unclear if these will apply or be achievable for MSRs. A range of material loss targets are considered; each of which is a multiple of a significant quantity.
- Material loss rate
  - The difficulty of detecting a loss is directly proportional to the duration of a loss for a fixed removal quantity.
- Replacement material
  - Direct losses of material have been easy to detect historically owing to the high precision of process monitoring measurements (e.g., tank level measurements, bulk solution measurements, etc). Similarly, direct losses in MSRs will also likely be detectable through high precision process monitoring. Substitution losses to (i.e., replacing removed material with an equal mass surrogate) might be more difficult to detect using inventory estimation, but might have an impact on reactor neutronics, which might be detectable using process monitoring.

A Python script was developed to automate the creation and running of material loss scenarios within SCALE, which is available upon request. The script performs a number of steps:

1. Initial calculation of actinide inventories up to the initiation of the material loss
   a) The material loss should remove *all* species at a proportional rate until the desired quantity is obtained.
   b) It is impossible to know what to remove at the initiation time without calculating the inventory up to that point.
2. The script calculates removal rates for a specified number of species
   a) Not all species can be specified for removal due to limitations within SCALE.
   b) The script fills out a template with a timetable describing the loss parameters.
      i. Improvements in the SCALE beta have removed some limitations that caused certain isotopes to be inaccurately tracked due to multiple stacked SCALE input decks.
3. Script runs a SCALE input deck that includes the material loss rates at the correct times.
4. Script formats SCALE output data by writing relevant fissile isotopes to a structured format for subsequent analysis (i.e., application of statistical tests).

The structured data output from the script then can be imported into a Python notebook (also available upon request) that automates the MC&A analyses by applying measurement error, error propagation, and statistical testing.

E.3. Loss scenario assumptions

The loss scenario analysis focuses on raw statistical metrics with only a minimal set of assumptions. Specifically, Page's trend test will be applied to the time-dependent SITMUF sequence in order to determine a probability of alarm (i.e., detection) for different simulated material losses. Results reported in following sections are bounded such that $P(\text{Alarm}|\text{MUF}, \Sigma) \in [0, 1]$ where 0 indicates the worst performance (i.e., no detection of the scenario) and 1 is the best (i.e., perfect detection of the scenario) and are reported on a per year basis (e.g., 0.60 would be a 60% chance of detection per year).

The h value used in the trend test are tuned to provide a 5% false alarm probability per year during normal operations whereas the k is fixed to 0.5 for all scenarios. Setting $k = 0.5$ is a common choice in the material accountancy literature as $k = 0.5$ is optimal for detection of a 1 significant quantity loss [14, 21].

Specification of a false alarm takes careful analysis as a single operational run of a MSR simulation is roughly 30 years, which corresponds to a nominal salt lifetime. Therefore, each MSR simulation run is divided into a single yearly segment and combined with all other segments to determine the FAP.

Molten salts represent a challenging measurement environment which may lead to higher uncertainties than in other contemporary bulk facilities (e.g., enrichment, reprocessing, fabrication, etc.). As such, consideration for a range of measurement uncertainties can provide insight into expected MC&A performance (using traditional statistics approaches) for a range of measurement technologies. Consequently, several sets of measurement errors are considered. It is well understood that measurement error, which is a function of the measurement device, has a strong impact on the probability of detection for a specified material loss. One set of results was included for illustrative purposes; namely the results reflecting 0.01% measurement error. Currently infeasible in real-world conditions, these are included to draw contrast to current state-of-the-art levels of measurement uncertainty (i.e. > 0.01%).

Generally, losses from blanket salts (where applicable) are more challenging than the fuel salt to model as they frequently have low equilibrium actinide inventories. There may not be a significant quantity present in some cases. In the analyses below, there are some instances where certain material losses have been omitted. This is because the uniform structure applied to generate these loss scenarios (i.e., the ordered set of parameters for loss quantity, duration, and start time applied to most designs and materials) would fail in some instances. This can occur when the requested material loss is larger than the inventory, for example. This has resulted in fewer losses from the blanket salts. Future work will endeavor to develop additional loss scenarios for blanket salts to improve the coverage of these analyses.

For the analyses presented in this work, multi-fluid systems were assumed to consist of separate inventories, but nonetheless reside in the same material balance area. That is, both fuel and blanket salts (where applicable) were both included in the same MUF calculation rather than individual consideration. Although less favorable from a statistical standpoint, this was seen as the most logical choice given the loops will likely occupy the same physical spaces (i.e., building or room).

APPENDIX F. BASELINE OPERATION

The following sections establish baseline inventories for each reactor design to serve as a point of reference for the evaluated loss scenarios.

MSDR

The MSDR was designed by ORNL and intended to demonstrate technologies developed during the molten salt reactor experiment [38]. For the purpose of this work, a LEU (5%) fluoride-based fuel salt is considered [5]. The reactor was designed to have continuous removal of noble metals and gaseous fission products with continuous feed of fuel material. As the MSDR is a thermal flux reactor, some of the observed behavior in the actinide inventory evolution over time will resemble light water reactors (LWRs). For example, the plutonium content present in the salt tends to build in over time, as shown in Figure F-1.

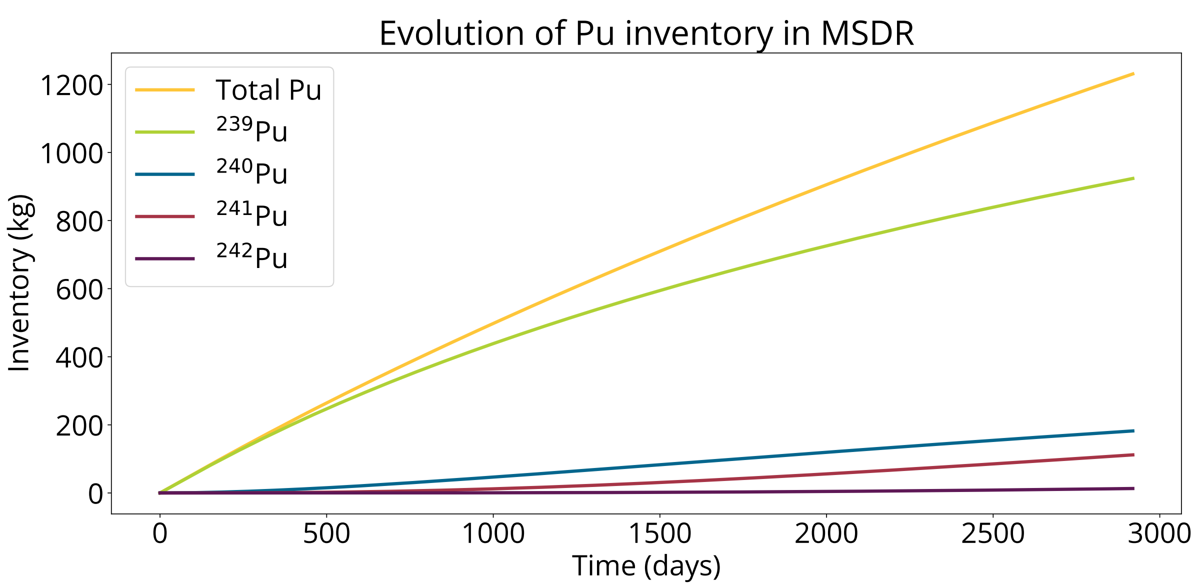


Figure F-1. Total Pu as a function of time for MSDR inventory

MOSART

MOSART is distinct from the other fast spectrum designs considered as LWR spent fuel transmutation was a key design objective. Criticality is maintained primarily using a Th/U fuel cycle while transmuting spent LWR. Consequently, there are multiple security relevant species present. Both Pu and ^{233}U are present in the primary salt. Figures F-2 and F-3 show the evolution of Pu and ^{233}U respectively under normal operating conditions.

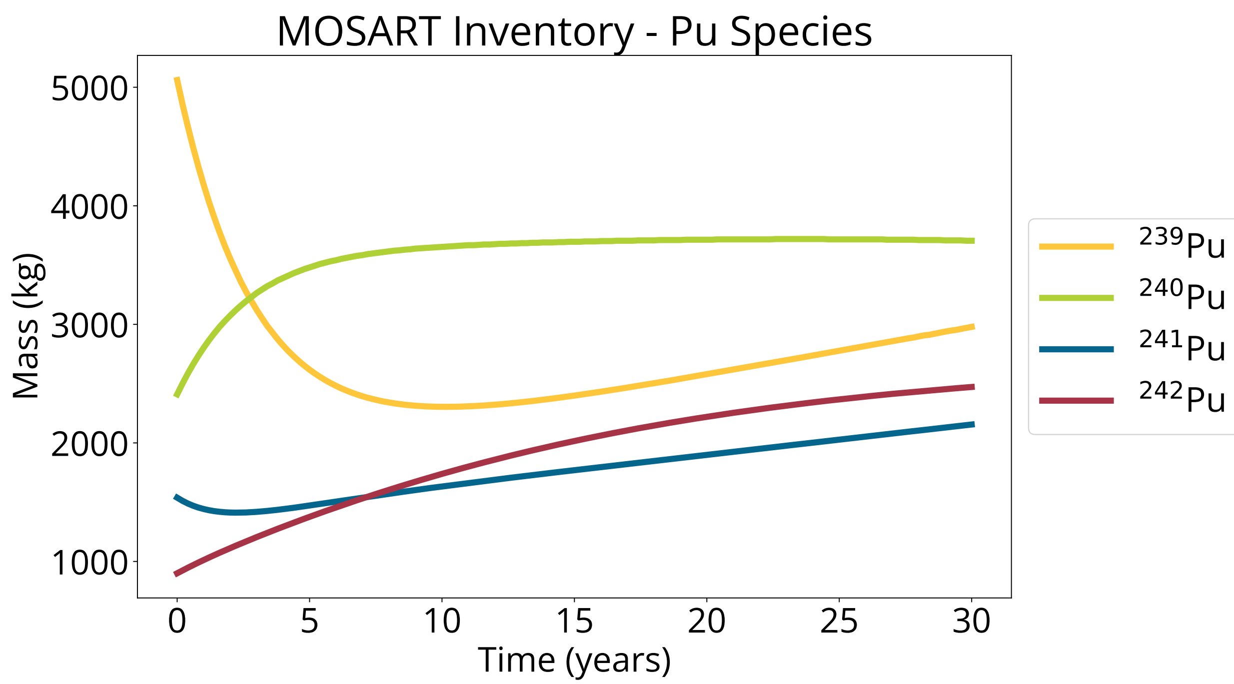


Figure F-2. Pu present in MOSART primary salt

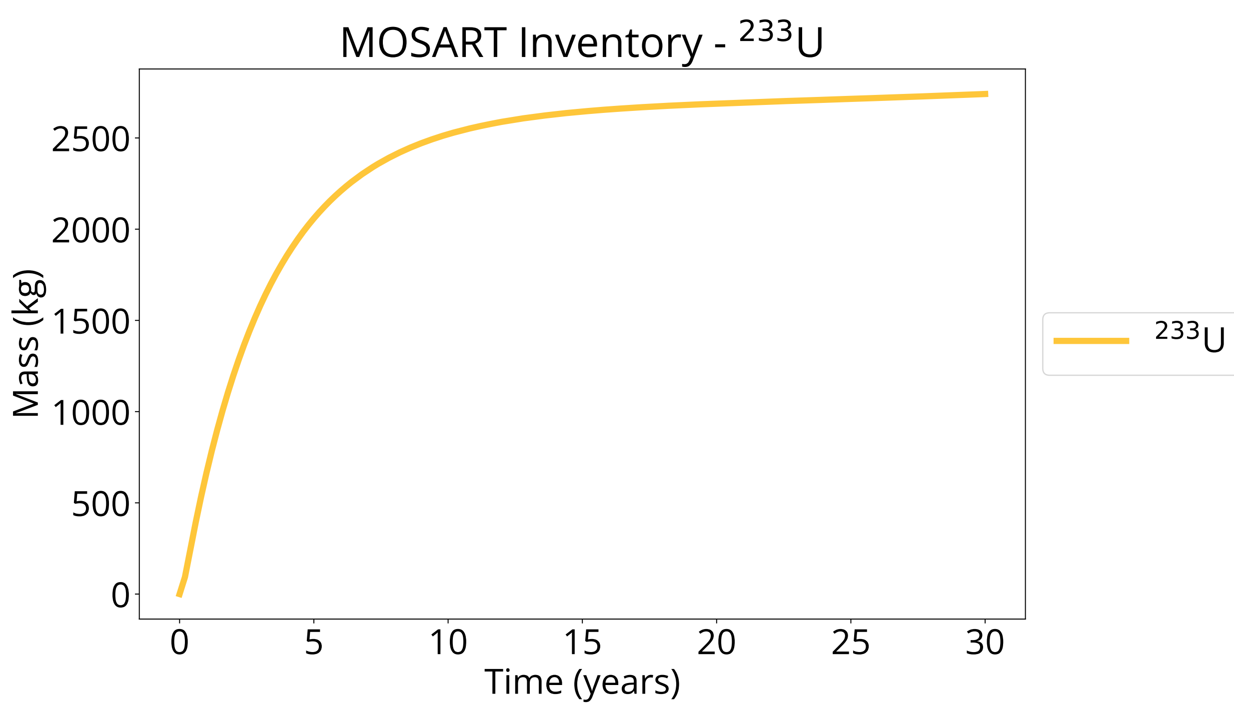


Figure F-3. U present in MOSART primary salt

This reactor maintains relatively high inventories of both ^{233}U and plutonium species throughout the operational lifecycle. As such, loss scenarios analyses will focus on removal of both species.

REBUS

REBUS is a large design (3700 MWth) designed to utilize a single fluid with a U/Pu cycle. The core has a relatively high initial inventory of 114 MTIHM due to the size of the reactor and homogeneous mix of both fertile and fissile species. Given the U/Pu fuel cycle, Pu species will be the primary element of interest for MC&A purposes. Inventories of primary Pu species are shown below in Figure F-4.

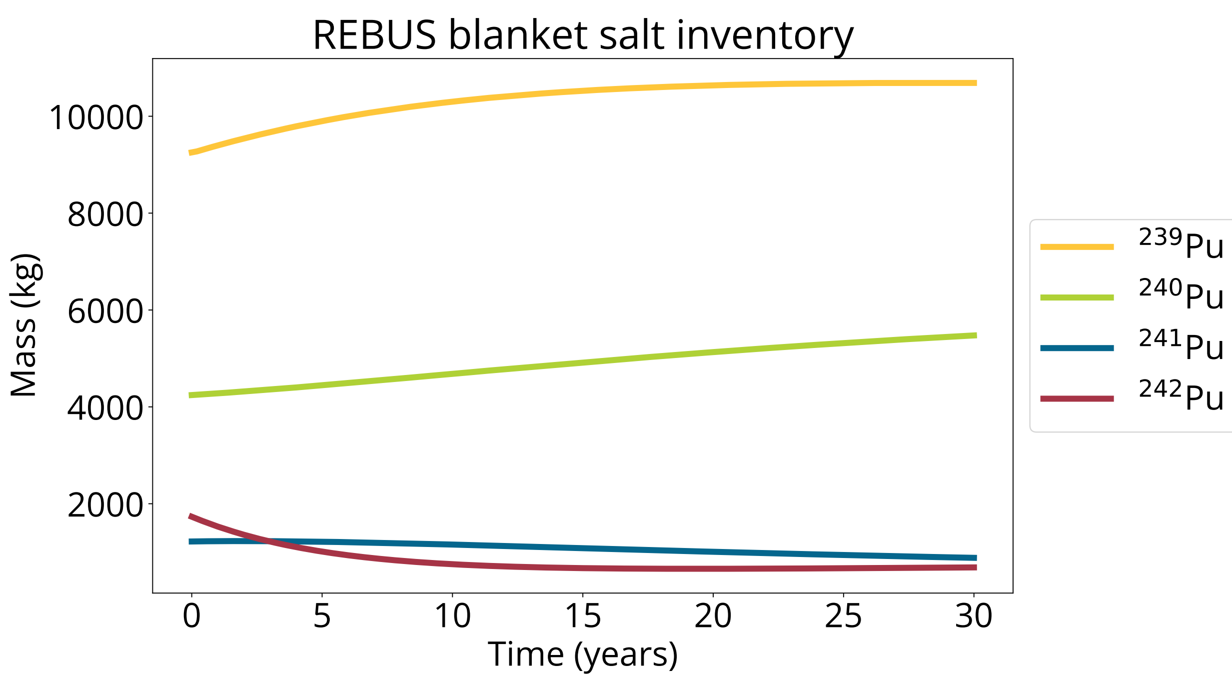


Figure F-4. Evolution of Pu species in REBUS primary salt.

Generally, the REBUS design has stable plutonium inventories over the salt lifetime. The upward inventory trend is likely due to a feed flowrate, which in this case is ^{238}U, that is slightly too high. The large inventory present during operation could present challenges to traditionally NMA.

MSFR

The MSFR is a two-fluid fast reactor designed around a Th/U fuel cycle. ^{232}Th is fed to both the blanket and fuel salts, although most of the breeding of ^{233}U occurs in the blanket. Separation operations on the blanket salt send ^{233}U to the fuel salt. ^{233}U is the dominant MC&A species of concern. The evolution of the MSFR inventory for both the blanket and fuel salts are shown in Figure F-5.

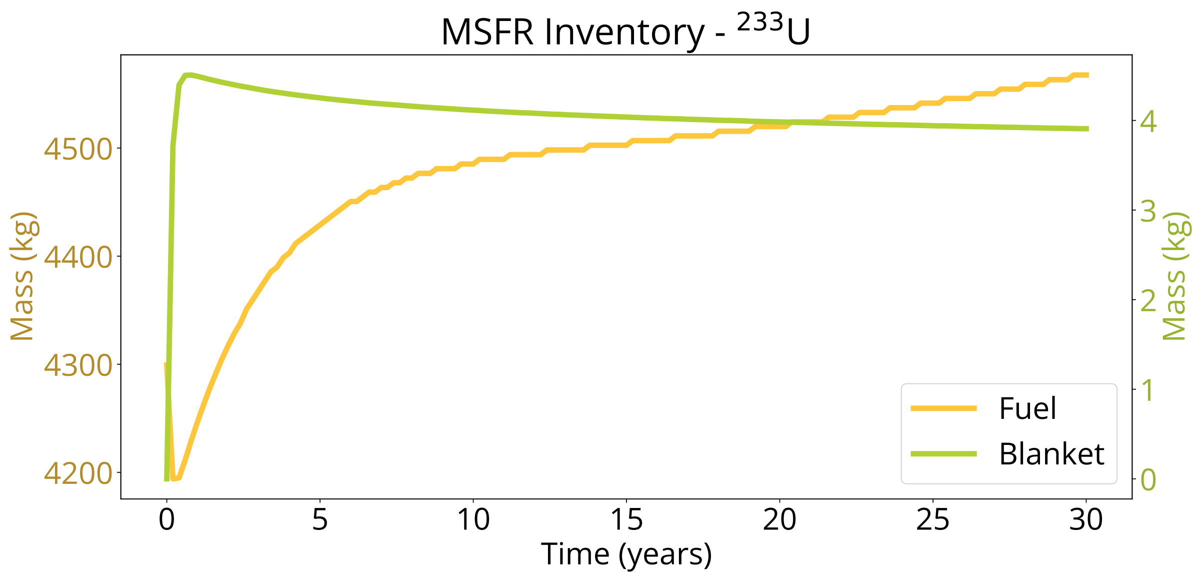


Figure F-5. ^{233}**U time-dependent behavior in fuel and blanket salts of the MSFR.**

The blanket salt contains a very small inventory of ^{233}U as it is constantly separated and added to the fuel salt. In contrast, the fuel salt has an appreciable ^{233}U inventory that slightly increases over time. Again, the increase in fuel salt inventory and decrease in blanket salt inventory are due to slight imprecisions in the flowrate. The MC&A focus for the MSFR will be on the ^{233}U content in the fuel salt.

MCSFR

The final design considered in this work, the MCSFR, is a large (6000 MWth) two-fluid design based on NaCl carrier salts. The reactor operates on a U/Pu cycle wherein Pu is largely bred in the blanket region before being transferred to the fuel salt. Pu species are the primary species of MC&A interest and their nominal behavior is shown in Figure F-6 and F-7 below.

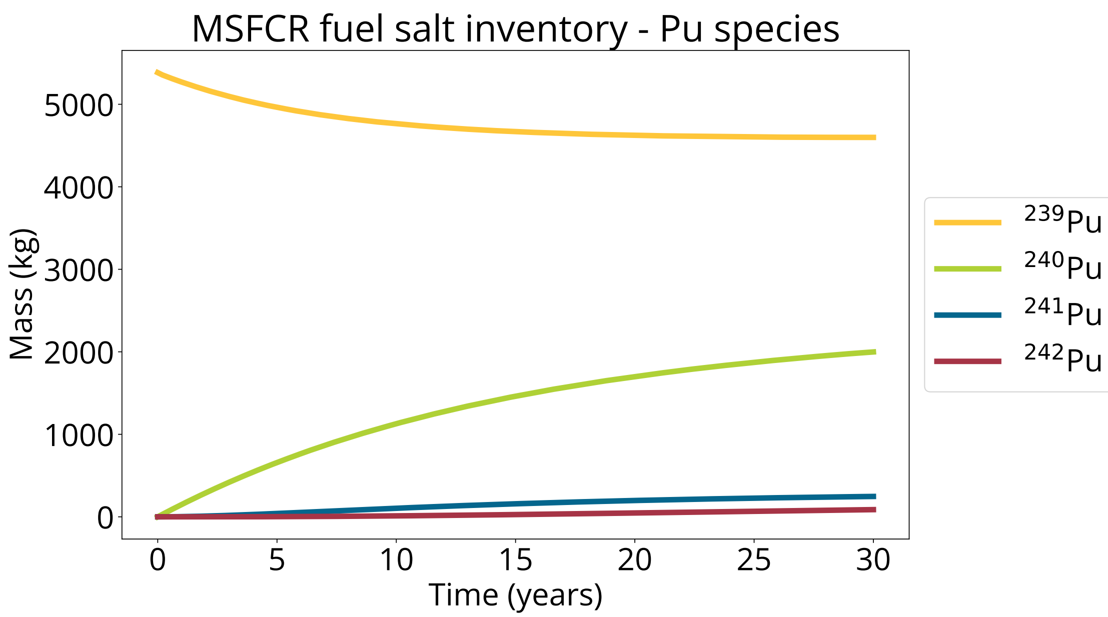


Figure F-6. MCSFR salt inventory

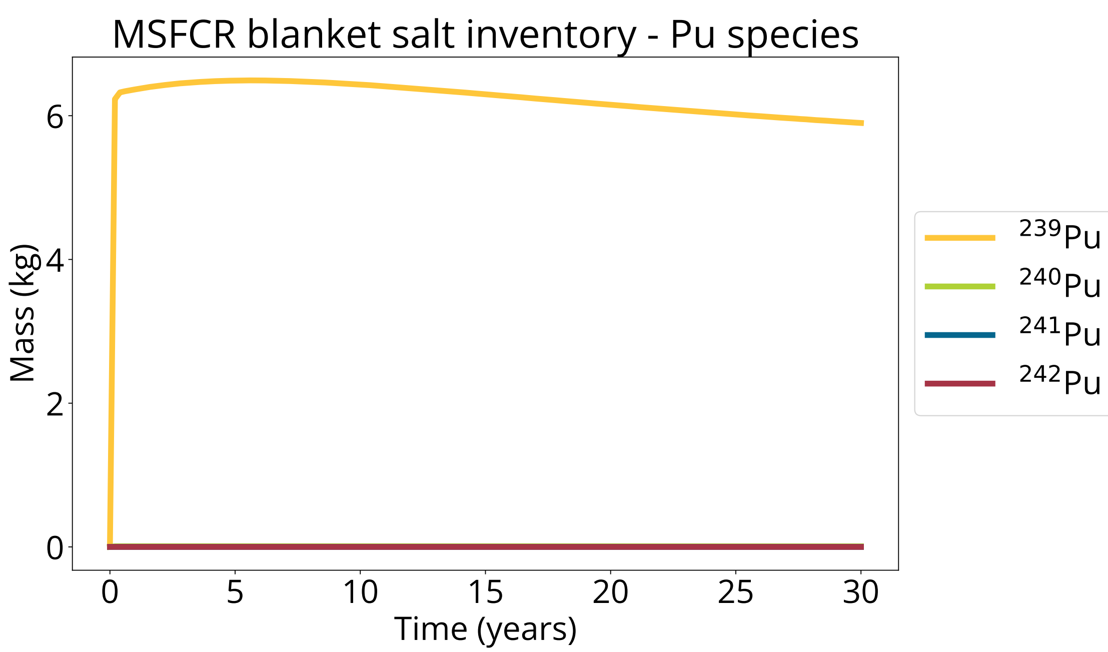


Figure F-7. MCSFR blanket inventory

Similar to the MSFR, the blanket inventory remains comparatively low across the entire salt lifetime. The actinide inventory gradually decreases as a result of a unoptimized feed rate to the blanket. Nonetheless, the fuel salt inventory remains largely stable with a comparatively large plutonium inventory.

APPENDIX G. UNCERTAINTY ANALYSIS

The uncertainty in the burnup calculations is not as well understood as measurement uncertainty. One study conducted for the Nuclear Regulatory Commission (NRC), NUREG/CR-7249 [39] has considered the uncertainty in SCALE for LWR applications. The report compared the predictions of a detailed SCALE model against measurements of a single fuel rod, MKP109, from a Combustion Engineering 14x14 assembly that was irradiated in Calvert Cliffs Unit 1. The calculation uncertainty ranged anywhere from a low of 1% to greater than 10%. The study identified numerous sources of error in the predicted isotopics, however, a key component for accurate predictions was detailed modeling conditions. For example, accurate modeling of the soluble boron concentration had a large impact on the overall prediction accuracy.

The results from NUREG/CR-7249 suggest that high uncertainties could exist unless the MSR material flows are modeled with sufficient detail[6]. This could prove to be challenging given the lack of literature on many complex phenomena that might dominate the behavior of the fuel salt. Impacts from model fidelity cannot be accurately captured as of this time. While SCALE does provide tools to probe arbitrary parameters to discover how they might impact the isotopic predictions, SCALE does not yet include modeling for many potentially dominant phenomena, such as the complex chemical interactions between the salt and pipe walls.

A lower bound on the uncertainty from the burnup calculation can be determined by considering the uncertainty arising from the underlying nuclear data used for depletion calculations. This would provide insight into expected uncertainties when perfect knowledge of the operating MSR was used to develop a sufficiently detailed model for use within SCALE. Specifically, the SAMPLER sequence within SCALE was used to perturb the nuclear data (cross-sections, decay data, and fission yields) to determine the impact on predicted isotopics. A total of 500 different perturbations were run to provide a good estimate ($\approx \pm 5\%$[7]) of the uncertainty.

The nuclear data uncertainty varies greatly between thermal and fast reactors. This is largely due to resolution of cross-section resonances in the thermal spectrum. First, consider the nuclear data uncertainty in the MSDR (thermal design) for plutonium species in the fuel salt, shown in Figure G-1.

Error for individual isotopes generally increases with number of neutrons (i.e. ^{239}Pu has the lowest uncertainty and ^{242}Pu is the highest). This is unsurprising given there are an increasing number of events that must occur to form the larger plutonium isotopes. The uncertainty also increases with the

[6] Here, sufficient detail refers to models that capture relevant phenomena as simply having a high resolution model (i.e. small dx and dt would be insufficient)

[7] This is just an estimate as it does not account for covariance between perturbed terms.

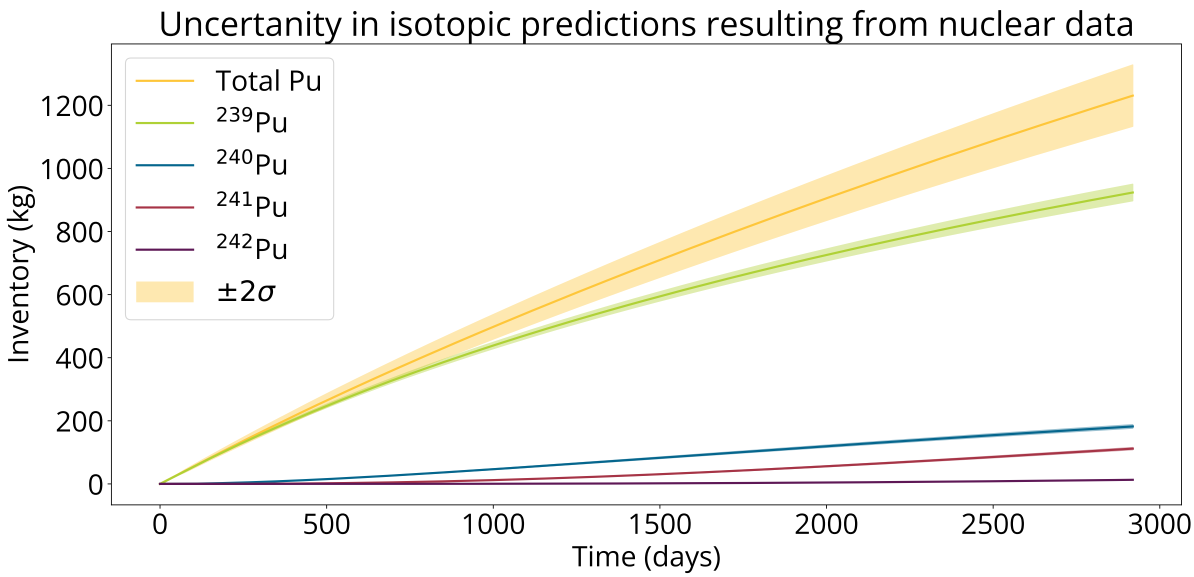


Figure G-1. Uncertainty arising from nuclear data. Shaded regions are $\pm 2\sigma$.

salt lifetime, however, this is largely due to the larger inventory present at a given time. The combined plutonium uncertainty can be a sizable 4% at end of salt lifetime.

Uncertainties in the MSDR (i.e., thermal spectrum) calculation are much larger than the fast spectrum designs, which do not require resonance resolution. Generally, uncertainties in the predicted isotopic concentrations calculated by SCALE/TRITON were on the order of 0.25% or less for the fast designs. Note that these uncertainties are likely lower than their thermal spectrum counterparts as a consequence of resonance cross section (i.e., resolution of resonance regions are required for thermal spectrum cross sections, but not fast spectrum cross sections).

Nuclear data uncertainties present in fast reactors will be ignored in analysis presented in subsequent sections given the relatively low magnitude (i.e., measurement uncertainty >> 0.25%). However, this uncertainty **will** be included for the MSDR where the contribution is much larger.