Limitations of Overall Measurement Error for Molten Salt Reactors

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Abstract

Existing light water reactors utilize item accounting for tracking discrete fuel assemblies, while bulk handling facilities calculate material unaccounted for (MUF) as part of safeguards verification. However, some next-generation reactors are moving away from discrete fuel rods and proposing new design aspects that could require a MUF calculation for materials accountancy. One of the next-generation reactor classes of interest is the molten salt reactor (MSR), which could have dissolved fuel as a coolant component. Some MSR designs include more extensive on-site salt processing and replenish the fuel salt, while others include only minimal online salt processing and replace the fuel salt periodically. MSRs are being proposed for use in various markets including cargo ships and scalable electric markets, so identifying accurate ways of tracking the actinides is imperative to the deployment of the MSR. This study considers the molten salt demonstration reactor (MSDR), a 300 MWe reactor designed by Oak Ridge National Lab, to develop a material balance and calculate theoretical MUF.

The MSDR's 8-year fuel lifetime is modeled using beta tools in SCALE. SCALE is used to understand the evolution of the actinides over the fuel lifetime in the reactor. Similar to thermal LWRs, the MSDR has an inventory of hundreds of kilograms of plutonium that builds up over its lifetime. Consequently, the material balance uncertainty grows which has implications for detection of material loss. This work considers material losses where the duration and initial start time is varied. Theft scenarios are investigated at three times: early in operation, in the middle of operation, and near the end of operation. This is done to capture changes in safeguards performance that will arise from the change in plutonium inventory over the reactor lifetime. The material loss time was varied to evaluate both abrupt and protracted loss scenarios. Initial MUF calculations show that the probability of detection for material loss will likely decrease with operational time as a result of the increasing plutonium inventory. These results suggest that differing safeguards approaches may be needed for MSRs.

Introduction

Reactor vendors are proposing aggressive deployment timelines of generation-IV reactor designs, with many moving to deploy within the next 10 years. Cost effective safeguards strategies developed during the design phase will be key to reaching the deployment goal [1]. The liquid fueled molten salt reactor (MSR) represents one of the larger departures from current Light Water Reactors (LWRs) in terms of design. Whereas an LWR has fuel rods stored in fuel bundles; the liquid fueled MSR dissolves the fuel in the primary loop coolant. Many proposed

liquid fueled MSRs are based on the Molten Salt Reactor Experiment (MSRE) that was run in the 1960s at Oak Ridge National Laboratory (ORNL)[2]. Some liquid fueled MSRs designs include online reprocessing, a unique feature to liquid fueled MSRs.

Traditional safeguards for conventional LWRs have utilized an item accounting approach. The item accounting approach is very effective with LWRs where the fuel bundles are easy to count and movement is easily tracked throughout the reactor facility. However, this approach cannot be used for many of the proposed advanced reactor designs. As the liquid fueled MSR utilizes a mixed fuel and the coolant in the primary loop, safeguards strategies used for existing bulk handling facilities (i.e. enrichment and reprocessing) will probably be more effective. Item counting will likely be reserved for accounting of bulk canisters of material entering and leaving the facility (similar to cylinders of feed and product at enrichment facilities).

For liquid fueled MSRs there are two methods currently being proposed for safeguards[3]. The first is considered a "black-box" approach for liquid fueled MSRs. Item accounting is used to measure the nuclear material of the fresh fuel and the fuel leaving the reactor while avoiding quantification of the liquid fueled MSR inventory. The reactor itself is safeguarded using stringent containment and surveillance (C/S). The benefit of the "black-box" approach is that it is more like the standard approach used in LWR facilities, allowing current approaches used by the international atomic energy agency (IAEA) to be applied. However, implementation of the "black box" approach will be complicated by the unique characteristics of the liquid fueled MSR. The unique characteristics of the liquid fueled MSR, like chemical reprocessing, may introduce large material unaccounted for (MUF) measurements, if MUF is only calculated using fuel entering and exiting the reactor.

The other proposed safeguard methodologies for liquid fueled MSRs include the development and use of process monitoring to quantify reactor inventory [3]. The benefits of a process monitoring system is that the system would utilize information measured for safety analysis and operation to increase the timely detection of nuclear material loss. Retrofits could be expensive, so the most cost-effective option would be incorporating these sensors during the design phase. Process monitoring data may also require joint use equipment if the data is needed by both the operator and inspector. The process monitoring approach could require the IAEA to develop new standards for the safeguards of liquid fueled MSRs.

As development of advanced reactors progresses, it is important to identify gaps in the current safeguards strategies for generation-IV reactors. This work models the molten salt demonstration reactor (MSDR) [4], a conceptual reactor developed by ORNL, and simulates three different abrupt material theft scenarios, at three different times of the fuels time in the reactor. The work covered in this paper, presents a candidate material balance for MSRs and discusses some general observations on system performance.

Data Generation and Simulation Setup

The MSDR radionuclide inventory is required to perform MUF calculations within the reactor. This is simulated using SCALE/TRITON[5] and SCALE/ORIGEN[6]. SCALE/TRITON and SCALE/ORIGEN are used to approximate the radionuclide generation, depletion, and decay of fuel salts over the fuel's lifetime in the reactor core. The MSDR is a 750 MW_{th}/ 350 MW_e reactor design that uses a LiF-U fuel salt. The MSDR has continuous fission product gas removal,

continuous removal of key noble metals, and continuous fresh fuel additions. The parameters used to model the MSDR are shown in Table 1. The inclusion of fission product removal and continuous feed for fresh fuel gives the model characteristics like the liquid fueled MSRs being proposed by vendors[1]. Figure 1 shows the notional uranium inventory as a function of time in the reactor.

Table 1. Parameters used in the SCALE simulations of the MSDR.

| MSDR Primary fuel salt composition		
^{6}Li	4.83E-06	wt frac
^{7}Li	4.83E-02	wt frac
^{19}F	3.29E-01	wt frac
^{235}U	3.08E-02	wt frac
^{238}U	5.91E-01	wt frac
Burnup parameter		
Specific Power	6.1968	MWD/MTHM
Irradiation Time	2880	days
Feed and removal isotopic groupings		
Group 1	Fission Product Gases	Kr, Xe, Ar, H, N, O
Group 2	Fission Product Solids	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te
Group 3	Enriched uranium feed	^{235}U, ^{238}U
Group flow rates		
Group 1	0.33	g/s (removal)
Group 2	0.33	g/s (removal)
Group 3	6.57E-05	g/s (addition)
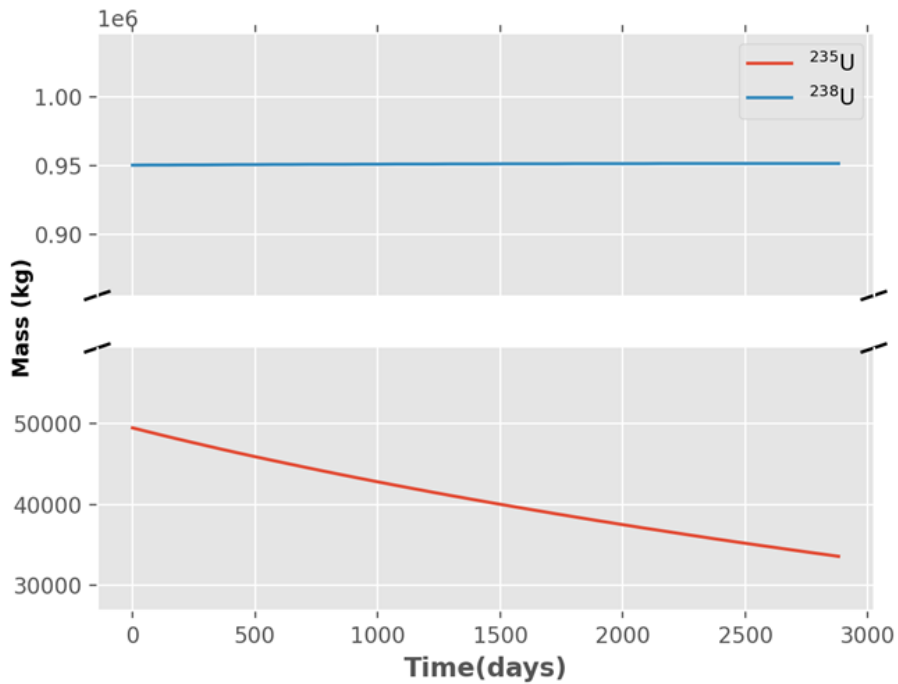
Figure 1. The notional Uranium inventory as a function of time in the reactor.

The ORIGEN outputs include the radionuclide inventory as a function of time in the reactor. The plutonium inventory is tracked throughout the fuel's lifetime in the MSDR core. Figure 2 plots the notional Pu inventory as a function of the time in the reactor. The maximum uncertainty due to the nuclear data for ^{242}Pu is 3% and the minimum uncertainty of the ^{239}Pu is 1.12%; however, the actual value of the uncertainty is a function of the specific isotope and the burnup. A 4% uncertainty is derived from the propagating error of all the plutonium isotopes and taking the final end-of-life uncertainty. The Pu inventory increases over the time in the reactor, and an equilibrium mass is not reached over the fuel's eight-year lifetime. The growing Pu mass presents concerns for using static safeguards criteria. The Pu inventory grows to over 1200 kg during the fuel's time in the reactor.

With the high amount of Pu within the reactor, the likelihood of identifying a significant quantity (SQ) loss is decreased [7]. 9 material loss scenarios were simulated for this study, with the adversary attempting to siphon one SQ of plutonium over a designated time. Early material loss A is an abrupt loss where B and C are increasingly protracted loss scenarios.

- Early Lifetime Material Loss Scenario
  - Early Material Loss A
  - Early Material Loss B
  - Early Material Loss C
- Mid-Lifetime Material Loss Scenario
  - Mid-Lifetime Material Loss A
  - Mid-Lifetime Material Loss B
  - Mid-Lifetime Material Loss C

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- End of Lifetime Material Loss Scenario
  - End of Lifetime Material Loss A
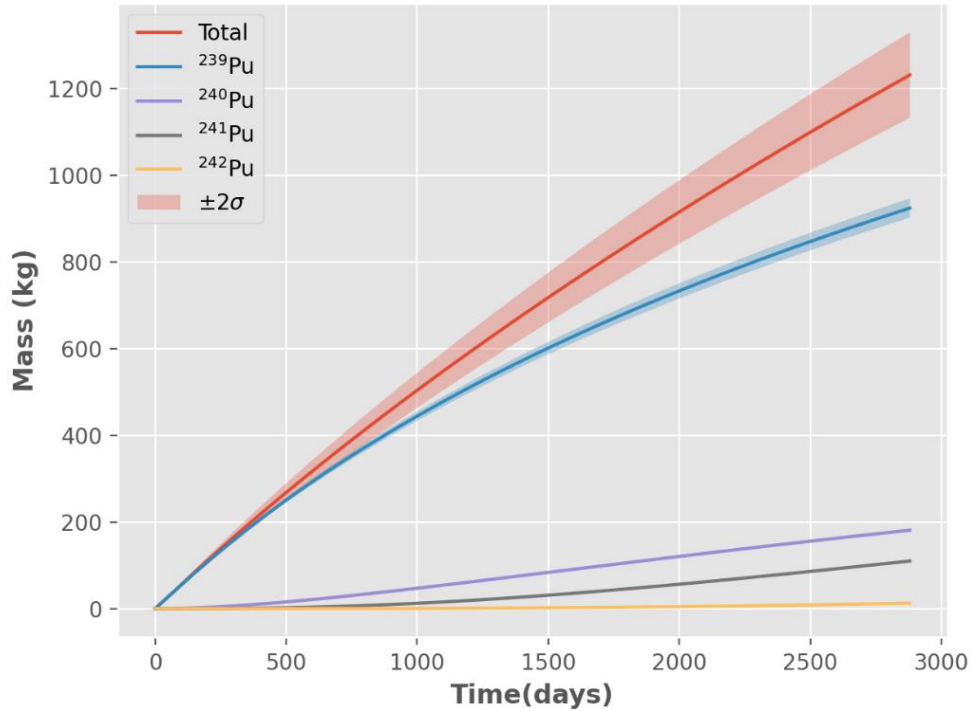 - End of Lifetime Material Loss B
  - End of Lifetime Material Loss C

Figure 2. The notional plutonium mass and propagated uncertainty as a function of time in the reactor

MUF Setup and Equations.

MUF [7] is a key statistical measurement for discriminating between material loss scenarios and nominal conditions at bulk nuclear facilities. MUF is calculated at the end of each material balance period (MBP). One possible way to calculate MUF is shown in Equation 1. This MUF calculation assumes that there are three components used to calculate the material balance. A concentration monitoring system for the reactor monitors or calculates the plutonium concentration within the reactor. The bulk measurement system measures the total inventory mass to convert the concentration of plutonium to mass. The final component is the result of a neutronic calculation that models the reactor and estimates the current radionuclide concentration within the reactor. Related research is currently investigating the viability of these measurements and is not considered in detail here.

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

- I = inventory

- m = measurement
- c = neutronic calculation
- t = time

Since there are currently no MSR datasets with which to perform this analysis, simulation forms the basis of the analysis. A baseline dataset is generated from SCALE. Then, errors are applied to generate representative datasets for measured and calculated salt concentrations. Errors are also applied to the constant salt volume to simulate measured bulk salt mass. Equation 1 determines the difference between the neutronic calculation and the measurement. The following uncertainties are used to estimate the standard deviation of the MUF: uncertainties in the bulk measurement, concentration measurement and the neutronic calculation. Table 2 lists the uncertainty for each of the measurement systems.

Table 2. Measurement Uncertainties used in these simulations.

	Random Error	Systematic Error
Bulk Salt Measurement	1%	1%
Calculated Concentration	1%	4%
Measured Concentration	1%	1%

Figure 3 plots the average MUF and associated σ_{MUF} for the nominal fuel conditions of the MSDR as a function of time in the reactor. The equations used to calculate MUF and σ_{MUF} are shown in more detail in equation 3 and equation 4. The measurement uncertainties listed in table 2 are used to calculate σ_{MUF} and MUF.
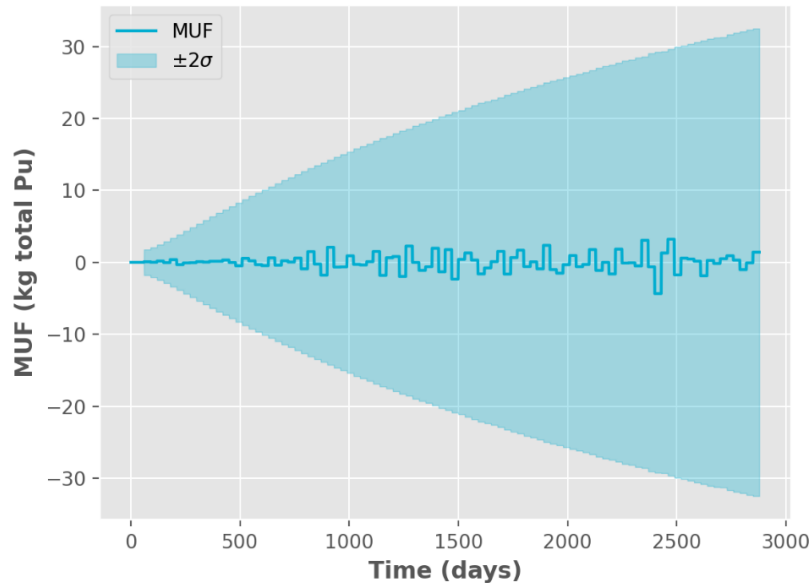

Figure 3. The Average MUF during normal operation using *Equation 2*.

The error model for measured values in the system is shown in Equation 2. The error model describes the relationship between the true, unobservable value and the error contaminated measured value. In the model setup two key types of error are identified: systematic error, S_i, and random error, $R_{i,t}$. The systematic error arises from measurement conditions or settings such as calibration curves which are not changed for some period of time, vary in an unpredictable way and is difficult to reduce. The random error varies in an unpredictable way under repeatable conditions and can be reduced through repeated measurements. Both systematic and random error are assumed to have a normal distribution. For each concentration and bulk variable in the MUF equation, equation 2 is used to model the errors related to each of the measurement systems.

$$M_{i,t} = G_{i,t}(1 + S_i + R_{i,t}) \tag{2}$$

- $M_{i,t}$=Measured value at location i and time t
- $G_{i,t}$=Ground truth value at location i and time t
- S_i=Systematic error random variate at location i
- $R_{i,t}$=Random error variate at location i and time t

For both the neutronic calculation and the concentration measurement in the reactor it is assumed that the fuel and coolant in the reactor are homogenously mixed. The standard deviation for Equation 1 is complicated by the covariance that arises due to the shared bulk measurement. The variables used to derive the MUF and the standard deviation of MUF derivation are shown:

- $I_{m,t} = B_{m,t}C_{m,t}$
- $I_{m,t} = B_{m,t}C_{m,t}$
- t=time
- B=bulk
- C=concentration
- c=calculated
- m= measured

Equation 1 is redefined in terms of the concentrations, masses, and associated errors as the inventory cannot be observed directly and shown in equation 3. The shared bulk measurement creates a covariance term related to the bulk measurement for both the neutronic calculation and concentration measurement, the standard deviation of the MUF is shown in equation 4.

$$
\begin{aligned}
\text{MUF} = {} & C_{m,t}B_t\left(1 + R_{Cm,t} + S_{Cm,t}R_{B,t} + S_{B,t}\right) - \\
& C_{m,t-1}B_{t-1}\left(1 + R_{Cm,t-1} + S_{Cm,t-1} + R_{B,t-1} + S_{B,t-1}\right) - \\
& C_{c,t}B_t\left(1 + R_{Cc,t} + S_{Cc,t} + R_{B,t} + S_{B,t}\right) + \\
& C_{c,t-1}B_{t-1}\left(1 + R_{Cc,t-1} + S_{Cc,t-1} + R_{B,t} + S_{B,t}\right)
\end{aligned} \tag{3}
$$

$$\sigma_{MUF} = [(C_{m,t}B_t)^2(\delta_{R_{m,t}}^2 + \delta_{S_{m,t}}^2 + \delta_{R_{b,t}}^2 + \delta_{S_{b,t}}^2) +$$
$$(C_{m,t-1}B_{t-1})^2\left(\delta_{R_{m,t-1}}^2 + \delta_{S_{m,t-1}}^2 + \delta_{R_{b,t-1}}^2 + \delta_{S_{b,t-1}}^2\right) +$$
$$(C_{c,t}B_t)^2\left(\delta_{R_{c,t}}^2 + \delta_{S_{c,t}}^2 + \delta_{R_{b,t}}^2 + \delta_{S_{b,t}}^2\right) +$$
$$(C_{c,t-1}B_{t-1})^2\left(\delta_{R_{c,t-1}}^2 + \delta_{S_{c,t-1}}^2 + \delta_{R_{b,t-1}}^2 + \delta_{S_{b,t-1}}^2\right) -$$
$$2C_{m,t}B_tC_{m,t-1}B_{t-1}(\delta_{S_{m,t}}^2 + \delta_{S_{b,t}}^2) - 2C_{c,t}C_{m,t}B_t^2(\delta_{R_{b,t}}^2 + \delta_{S_{b,t}}^2)$$
$$+$$
$$2C_{m,t}B_tC_{c,t-1}B_{t-1}\delta_{S_{b,t}}^2 + 2C_{m,t-1}B_{t-1}C_{c,t}B_t\delta_{S_{b,t}}^2 -$$
$$2C_{m,t-1}C_{c,t-1}B_{t-1}^2\delta_{S_{b,t}}^2 - 2C_{c,t}B_tC_{c,t-1}B_{t-1}(\delta_{S_{c,t}}^2 + \delta_{S_{b,t}}^2)]^{\frac{1}{2}}$$

(4)

Results
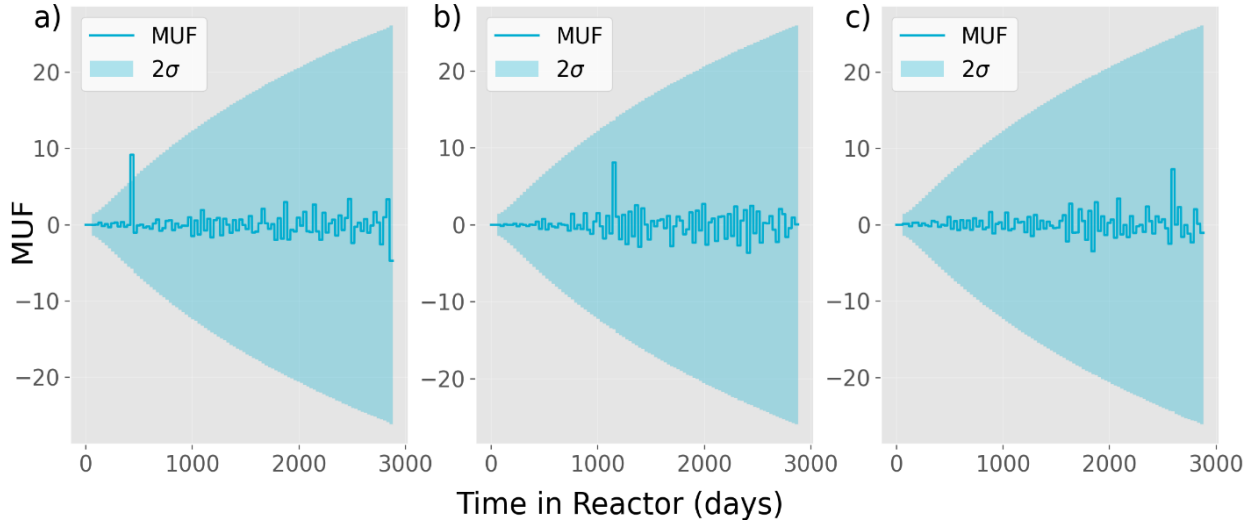
Figure 4. The average MUF measurements as a function of time fuel spends in the reactor for the three loss scenarios. a) Early Lifetime Loss, b) Mid-Lifetime Loss, c) End of Lifetime Loss.

Figure 4 shows the impact an adversary's choice on start time can have on identifying the material loss scenario. The first loss scenario occurs a little after a year in the reactor. For this case, the material loss scenario causes the MUF to increase to over 2σ, increasing the likelihood that an operator would be able to detect that a loss has occurred. However, in the case of the mid-lifetime and end of lifetime losses in the reactor, the loss is well within 2σ making it harder to detect whether a loss has occurred. MUF measurements by themselves are not a good indicator for whether a loss has occurred.
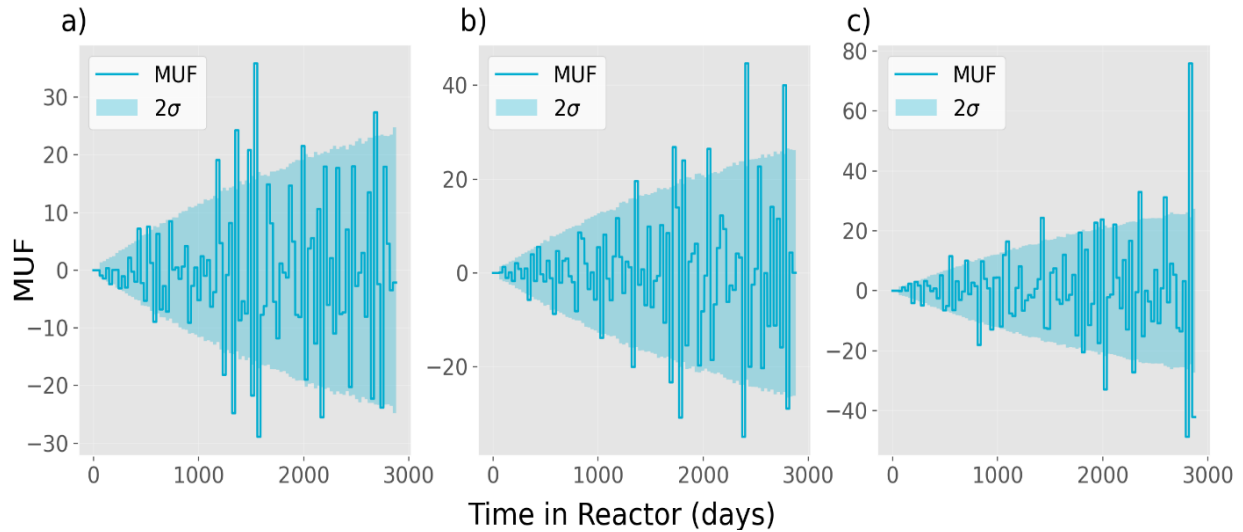
Figure 5. Individual MUF measurements as a function of time fuel spends in the reactor for the three loss scenarios. a) Early Lifetime Loss, b) Mid-lifetime Loss, c) End of Lifetime Loss.

Figure 5 plots individual MUF cases for the three loss scenarios, and this plot further proves that MUF measurements will not be a good indicator of loss scenarios. The fluctuations in MUF are due to the measurement error. For MUF to be a successful measure of losses, either the uncertainty surrounding the systematic, bulk, and cross-sections would need to decrease or MUF by itself will not be an efficient method for identifying loss scenarios.

Conclusions

The liquid fueled MSR reactor presents a unique safeguards challenge, with the mass of plutonium growing to high levels over the fuel's time in the reactor core. The large mass of plutonium causes the standard deviation of the MUF to increase with fuel salt burnup. Future work will consider the individual error components to prioritize future R&D efforts for the measurement systems of a liquid fueled MSR.

This work shows that MUF for a thermal spectrum LEU MSR can grow to large values over time, but the actinide growth is dependent on the design. Future work may be required to reduce the material balance uncertainty. Effective safeguards strategies will be required to meet aggressive deployment timelines proposed by vendors. There are two aspects of the liquid fueled MSR that could improve the safeguards of this design. The first is the dilution of the fuel concentration within the coolant salt. Although over a thousand kilograms of plutonium are generated over the fuel's time in the reactor, the plutonium mass is still a small ratio of the material in the reactor, which makes it difficult for an adversary to siphon a single SQ of plutonium without doing chemical processing on a much larger mass within the reactor core. The reactor core is radioactively hot, which also limits an adversary's ability to remove material. The radioactive heat limits loss pathways, and a credit system can be utilized to take into account the self-protecting nature of the reactor core. More standard safeguards approaches could be used to safeguard the reactor core. For example, process monitoring measurement systems used for operating the reactor can be used to decrease the uncertainty in the MUF calculation.

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Containment and surveillance could provide additional assurances that reactors have not been tampered with during operation.

Acknowledgements

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