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Safeguards for the Lithium Fluoride Thorium Reactor: A Preliminary Nuclear Material Control and Accounting Assessment

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August 2022

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OAK RIDGE National Laboratory

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Nuclear Nonproliferation Division

SAFEGUARDS FOR THE LITHIUM FLUORIDE THORIUM REACTOR: A PRELIMINARY NUCLEAR MATERIAL CONTROL AND ACCOUNTING ASSESSMENT

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ACRONYMS

CFR	Code of Federal Regulations
DOE	Department of Energy
EPRI	Electric Power Research Institute
FEI	Flibe Energy Inc.
GAIN	Gateway for Accelerated Innovation in Nuclear
HALEU	high-assay low-enriched uranium
IAEA	International Atomic Energy Agency
ICA	internal/item control area
LEU	low-enriched uranium
LFTR	lithium fluoride thorium reactor
MBA	material balance area
MC&A	material control and accounting
MSBR	molten salt breeder reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
MW_{th}	megawatt thermal
MW/MT_{IHM}	megawatt per metric tonne initial heavy metal
NDA	nondestructive assay
NE	Office of Nuclear Energy
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PWR	pressurized water reactor
SNM	special nuclear material
SQ	significant quantity

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EXECUTIVE SUMMARY

PURPOSE

This report was prepared by Oak Ridge National Laboratory (ORNL) for Flibe Energy Inc. (FEI), a US-based advanced reactor company founded in 2011 and headquartered in Huntsville, Alabama. FEI is developing the lithium-fluoride thorium reactor (LFTR), which is a modern two-fluid molten salt reactor (MSR) design operating on a thorium/^{233}U fuel cycle. FEI intends for LFTR to become a self-sustaining clean energy source that can create or breed its own fuel from thorium. Each LFTR is intended to breed enough fissile material to compensate for the amount it consumes. Consequently it would not require fissile replenishment during its operational lifetime. This self-sustaining nature would eliminate the need for uranium enrichment infrastructure to support LFTRs after the first generation [1].

However, this MSR design presents challenges for domestic safeguards as defined by the US Nuclear Regulatory Commission (NRC) as well as international safeguards as applied by the International Atomic Energy Agency (IAEA). The NRC defines domestic safeguards as material control and accounting (MC&A) and physical protection. Typically, MC&A licensee programs, in accordance with Title 10 of the *Code of Federal Regulations* (CFR) Part 74 [2], provide control and accounting measures to detect abrupt and protracted theft or diversion of special nuclear material (SNM) from authorized locations and processes within a facility. Physical protection licensee programs, in accordance with 10 CFR Part 73 [3], consist of a variety of measures to protect nuclear facilities and material against sabotage, malicious acts, and theft or diversion that result in the removal of licensed material from the facility. MC&A requirements work together with a licensee's physical protection programs to create an integrated and complementary domestic safeguards approach that results in a more robust protection against sabotage, theft, and diversion of licensed materials. The requirements within 10 CFR Part 74 [2] are a graded approach based upon the category of SNM.

The LFTR design is based on the 1967 two-fluid molten salt breeder reactor, which uses a ^{233}U-bearing salt as the fuel salt and a ^{232}Th-bearing salt as a blanket salt. The LFTR is a breeder reactor, which relies on the Th/^{233}U fuel cycle, which means that fertile thorium is converted to ^{233}Pa via neutron capture and subsequently decays to fissile ^{233}U. A portion of the blanket salt is continually processed in an electrolytic separator to remove ^{233}Pa and ^{233}U from the blanket salt, which are sent to a decay tank to allow time for the ^{233}Pa to decay. The LFTR design has two fluorinators. The decay fluorinator is used to remove uranium from the decay tank, while the fuel fluorinator is used to remove uranium from a portion of the fuel salt as the first step of removing fission products. The output of the two fluorinators are combined with cleaned (i.e., no fission products) FLiBe salt before reentering the reactor vessel. The fission products are removed from the residual salt from the fuel fluorinator by means of a second electrolytic separator. This process is described in more detail in Section 1.2.2. Note that the LFTR design currently proposed by FEI is an evolution of the design presented previously by FEI [4].

The Th/^{233}U fuel cycle is fundamental to LFTRs. Therefore, one key design feature of LFTRs is the production of nearly pure ^{233}U from its short-lived (half-life ~27 days) precursor ^{233}Pa, which is produced from the neutron capture of thorium and subsequently held in a decay tank before fluorination. This ^{233}U is central to the operation of LFTR. However, separated ^{233}U presents a potential target for theft and diversion. Furthermore, the production of ^{233}U within the decay tank results in the production or accumulation of fissile material outside the reactor vessel. Other challenging aspects of this design from a material accountancy perspective include the continuous online processing of the nuclear material within flowing molten-salt fuel within LFTR that is required as part of reactor operations in addition to the continuous chemical processing of the blanket and decay salts. This continuous processing, in addition to the changing nuclear material isotopic composition in the fluid salts, poses challenges for measuring and identifying what nuclear material is present and where it is present in the reactor and processing systems.

On the other hand, these challenges present opportunities to consider how to design the system to optimize the functionality of an MC&A system and how this system can support the future application of IAEA safeguards to the LFTR. The purpose of this report is to provide FEI with design recommendations for the implementation of nuclear material accountancy within a LFTR and enable FEI to meet its sustainability and operational goals for the LFTR.

This work was supported by the Gateway for Accelerated Innovation in Nuclear (GAIN) Program of the US Department of Energy's (DOE's) Office of Nuclear Energy (NE). This public-private partnership between ORNL and FEI presents an opportunity to consider MC&A aspects of LFTR at the design phase and examine critical elements of the reactor design from an MC&A perspective. The goal of this work is to provide recommendations to FEI to support its development of a LFTR design that helps progress it toward the practical implementation of MC&A including IAEA safeguards as part of the eventual commercialization of LFTR. The term *IAEA safeguards* refers to a set of measures and verification activities conducted by the IAEA at facilities and locations within a State under State-specific safeguards agreements. The technical objective of IAEA safeguards is "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection" [5]. This technical objective helps ensure the peaceful use of nuclear material in support of the global nuclear nonproliferation regime. IAEA safeguards depend on the application of MC&A and physical protection by a State to carry out its independent verification activities. The IAEA call this "state systems of accounting for and control of nuclear material."

The significant outcomes of this work were design recommendations for the process flow sheet within the 2018 LFTR design, which led to the generation of a revised 2021 LFTR design by FEI. Ultimately, understanding MC&A system design considerations for LFTR will provide input to the future MC&A planning process including MC&A system performance requirements.

MISSION NEED

Advanced nuclear power is proposed as an integral part of the expansion of the US nuclear energy industry. Advanced nuclear reactors and their associated fuel cycles will be required to address MC&A and IAEA safeguards for both domestic and international deployment, respectively. LFTR, and some MSRs in general, can be considered to contain more than one part of the fuel cycle in a single facility (e.g., fuel fabrication or synthesis, power generation or the nuclear reactor, and isotope separations), which creates unique challenges for MC&A and IAEA safeguards.

The mission of the DOE-NE GAIN program is to "Provide the nuclear energy industry with access to cutting-edge R&D, along with the technical, regulatory, and financial support necessary to move innovative nuclear energy technologies toward commercialization in an accelerated and cost-effective fashion" [6]. The mission need addressed by this report is to support the nuclear energy industry by moving a specific innovative nuclear energy technology, the LFTR, toward commercialization by considering MC&A needs at the design stage.

FEI has plans to develop a demonstration-scale LFTR (power ~60 MW_{th}) before developing a commercial-scale LFTR (power ~600 MW_{th}), and it is exploring potential US sites within the states of Alabama and Mississippi. The FEI LFTR design presents an opportunity to consider MC&A for a liquid-fueled MSR operating on a thorium/^{233}U fuel cycle and to do this at an early stage. Taking these nuclear safeguards aspects into consideration, in addition to safety and security, will prepare the designs for licensing and commercial deployment within the US and internationally.

KEY OUTCOME: 2021 LFTR DESIGN RECOMMENDATIONS

This report presents the results and findings from a preliminary MC&A assessment of the 2018 FEI LFTR chemical processing system design. The results from inventory calculations were performed using the 1967 ORNL two-fluid Molten Salt Breeder Reactor (MSBR) design [7] as a reference for the expected LFTR chemical processing system design. These calculations were used to inform recommendations for the next iteration of the LFTR chemical processing system design. Specifically, this report describes and evaluates the design features of 2018 LFTR against requirements for MC&A. The overarching goal is to provide recommendations to FEI that could be adapted to facilitate the implementation of more effective and efficient MC&A to future designs of the LFTR chemical processing system such as the 2021 version. The main features of the LFTR design are described, together with an overview of known and unknown operational aspects relevant to nuclear material accounting, based on currently available LFTR design information provided by FEI. The MC&A assessment was performed using a reactor physics model of an 1967 ORNL two-fluid MSBR design scaled to 600 MW_{th}. A reactor physics analysis was performed to estimate the nuclear material inventory and radiation source terms in the LFTR chemical processing system, and results are presented to show how LFTR design features and operations may influence MC&A as well as the application of IAEA safeguards.

The table below summarizes the design features of the 2018 LFTR chemical processing system representing domestic safeguards challenges that the ORNL and FEI team identified. This table also summarizes the design recommendations made by the team in this report and provides input to an updated design for the LFTR chemical processing system.

2018 LFTR Chemical Processing System Design Features and Potential Nuclear Material Control and Accountancy (MC&A) Challenges	Design Recommendations for improving MC&A within the LFTR Chemical Processing System
• A single large decay tank volume potentially leads to uncertainties in accounting measures (e.g., weighing precision and gamma self-shielding) and thus renders quantification of the decay tank nuclear material inventory challenging.	• Perform a systems design and explore the use of a series of smaller decay salt inventories while examining impacts to uncertainty in accountancy measurements for decay tank nuclear material inventory quantification (i.e., investigate tradeoffs between factors such as geometry, weighing precision, and self-shielding). • Design and build MC&A technologies into the decay process; for example, containment and surveillance technologies such as tamper indicating devices; or accountancy measurements using a custom nondestructive assay (NDA) system for the series of decay salt inventory(ies) potentially using both gamma-ray spectroscopy for isotopic composition analysis and neutron counting techniques for mass quantification. • Use the electrolytic cell in the 2018 LFTR chemical processing system design to validate other accountancy measures. Since thorium replacement at the electrolytic cell can be measured very precisely, it is possible to precisely quantify the mass of uranium and protactinium transferred to the decay tank.

2018 LFTR Chemical Processing System Design Features and Potential Nuclear Material Control and Accountancy (MC&A) Challenges	Design Recommendations for improving MC&A within the LFTR Chemical Processing System
• Continuous salt processing makes nuclear material inventory management and accounting challenging because it requires real-time accountancy of highly radioactive nuclear material in process, in a liquid form, at a high temperature, and in a highly corrosive environment.	• Convert to batch processing of the salt to enable additional static accounting measures (e.g., weight, volume). This may also help address the need for additional shielding and reduce the impact of high temperatures and corrosivity on instrumentation. • Design decay salt inventories in such a way that they become items (i.e., can be verified and then sealed to maintain continuity of knowledge). This would require the use of individually sealed containers of decay salt that can disconnect from the filling pipe. After several half-lives, the protactinium would be sufficiently decayed. Note that this option might be least favorable from a physical access standpoint.
• From a domestic safeguards standpoint, the decay tank inventory is not obviously self-protecting. Estimates of the decay tank inventory predicted that the ^{232}U content was 0.05 ppm with minimal fission products present, which are expected to be the dominant contributions to the radiation source term.	• Verify the expected self-protection of the decay inventory by expanding the model to explicitly account for decay salt composition and tank geometry. • Add fission products that have been separated from the fuel salt to increase the gamma activity within the decay tank and thus increase self-protection and deterrence to potential theft. Note that this may make measurement more difficult for both MC&A and IAEA independent verification, and trade-offs will need to be investigated. • Combine the decay inventory(ies) with a portion of the (clean or irradiated) fuel salt to reduce the purity of the uranium vector of the decay inventory. • Increase ^{232}U content by modified reactor operations, blanket composition, or both, such as by the addition of ^{230}Th [8] to improve self-protection.
• Approximately 3–8 kg of fissile ^{233}U and 27–28 kg Pa (~90% ^{233}Pa, ~10% ^{231}Pa) reside in the decay tank continuously with potential access, depending on the radiation source term.	• Rework the blanket salt processing to remove a dedicated decay salt inventory. For example, send blanket salt directly to the decay fluorinator (i.e., do not remove protactinium from the blanket salt). Note, this change is invasive and incompatible with the above suggestions. In addition, this will significantly reduce the thorium conversion efficiency by not protecting the protactinium from further neutron capture. Additionally, it may significantly alter the neutronics of the reactor.

2018 LFTR Chemical Processing System Design Features and Potential Nuclear Material Control and Accountancy (MC&A) Challenges	Design Recommendations for improving MC&A within the LFTR Chemical Processing System
• The uranium vector in the UF_6 output from the decay salt fluorinator was estimated to be 99.98% ^{233}U and 0.02% ^{234}U, with 0.05 ppm ^{232}U.	• Eliminate the decay fluorinator shown in Figure 2 in Section 1.2.2 entirely by allowing protactinium to decay in a small inventory of fuel salt outside the reactor vessel. • Reduce purity of the uranium vector from the decay fluorinator for example by routing the output of the fuel fluorinator to the input of the decay fluorinator. • Remove physical access to the UF_6 stream. For example, ensure the pipe between the fluorinators is too short for access.

*Note some recommendations are mutually exclusive.

KEY OUTCOME: DECAY TANK SYSTEM OPTIONS AND ROLE OF MEASUREMENT TECHNOLOGY

Five potential options were identified for the decay tank system to increase the effectiveness of nuclear material accountancy and facilitate potential future IAEA safeguards verification activities: (1) maintain the baseline of a single decay tank using a quasi-continuous process; (2) convert the operation of the decay tank to a series of smaller decay salt inventories that also use a continuous process; (3) convert the operation of the decay tank to a series of smaller decay salt inventories using a batch process; (4) convert the operation of the decay tank to a series of smaller decay salt inventories using a batch process that could be designed to become sealed items; and (5) eliminate the decay tank entirely. These options are illustrated in the schematic shown in Figure 1.

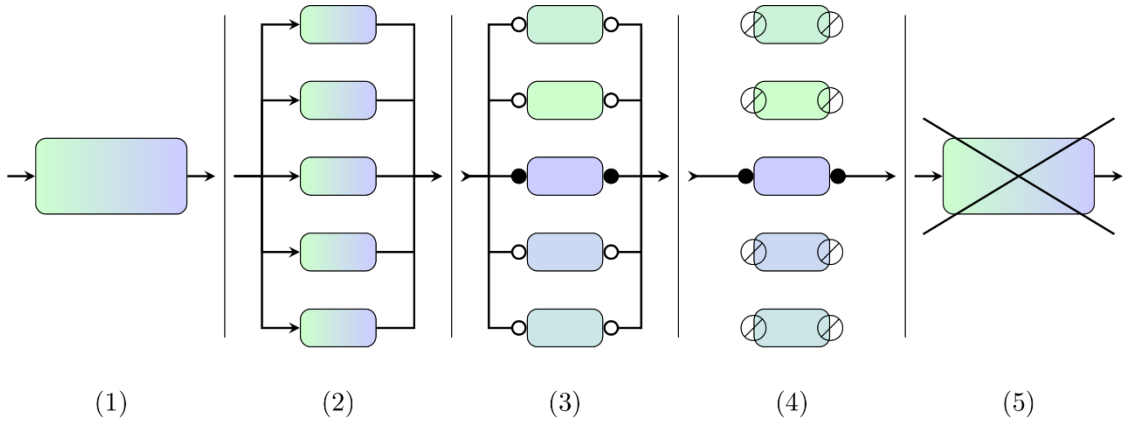

Figure 1. Diagram representing the decay tank recommendations. (1) Maintain the baseline. (2) Use series of smaller decay salt inventories with continuous processing. (3) Use series of decay salt inventories with batch processing. (4) Use series of decay salt inventories with batch processing using sealed containers while disconnected. (5) Remove the decay tank.

A range of options are presented that have different relative advantages and disadvantages to both the future LFTR operator from an MC&A perspective and a future inspector from an IAEA safeguards perspective. For example, options 2–4 are advantageous from a verification standpoint because they would reduce the salt volume within the individual decay tanks, reducing the radiation source term and

corresponding intensity of any interfering gamma-rays (e.g., ^{232}U decay progeny such as ^{208}Tl, fission products), which is advantageous for performing verification measurements via gamma-ray spectroscopy. This trade-off, and other similar trade-offs, need to be evaluated in detail as part of future work.

MC&A measurement technologies may be designed and built into the decay tank process. For example, a custom nondestructive assay system could be designed and developed for the series of decay tanks using mass and volume measurements in combination with gamma-ray spectroscopy for isotopic composition analysis and neutron counting techniques (i.e., neutron singles counting and correlated neutron counting including neutron coincidence counting) for mass quantification.

1. INTRODUCTION TO LIQUID FLUORIDE THORIUM REACTORS

The Flibe Energy Inc. (FEI) liquid fluoride thorium reactor (LFTR) is a proposed advanced[1] nuclear reactor under active development operating on a thorium/^{233}U fuel cycle and designed to achieve a conversion ratio of unity or above. Fundamentally, the LFTR concept aims to maximize thorium consumption efficiency and minimize actinides in the waste stream. Furthermore, FEI aims for LFTR to have a simplified chemical processing system, where possible, compared to molten salt reactor designs of the past.

In addition to these aims for LFTR, FEI would like to consider design features that would enable effective and efficient nuclear material control and accountancy (MC&A), within the context of the US Nuclear Regulatory Commission (NRC) domestic safeguards, as well as the application of International Atomic Energy Agency (IAEA) safeguards in the future. The remainder of this section outlines the main design features of the reactor followed by the history of the LFTR design. Then, Section 2 presents the goals for the project as well as the research questions to be answered throughout the report. Section 3 presents the model used to evaluate the LFTR design, and Section 4 contains the results of the neutronics model, which includes both isotopic inventories in the design as well as dose calculations. The report continues with the preliminary MC&A assessment in Section 5, which includes assessment of the material accountancy and material balance areas. Finally, Section 6 concludes with the potential MC&A and IAEA safeguards concerns of the LFTR design and recommendations for improving the design.

1.1 MAIN DESIGN FEATURES OF LFTR

According to FEI, the LFTR design contains several design features that distinguish it from the current fleet of reactors outside the non-traditional fuel form and non-water coolant. First, LFTR operates on a thorium/^{233}U fuel cycle, whereby fertile thorium is converted to fissile ^{233}U during operation. Compared to the single-fluid MSBR, the chemical processing system is simplified by separating the lanthanide and thorium isotopes into separate salt streams. Finally, FEI currently has plans to build LFTRs with two separate vessels with only one vessel filled with salt and operating at a given time, which is intended to reduce operational downtime by allowing maintenance on the out-of-use vessel while remaining at full power. Note that only one vessel at a time will contain nuclear material, and the second vessel will remain empty until ready for use.

1.2 HISTORY OF THE LITHIUM FLUORIDE THORIUM REACTOR (LFTR)

The original conception of the LFTR design began with the Oak Ridge National Laboratory (ORNL) Molten Salt Breeder Reactor (MSBR) program. The program explored several aspects of the salt processing system and included criticality experiments to prove the viability of operating reactors using uranium–fluoride molten salts.

As part of the MSBR program both a one-fluid (combined fissile and fertile salts) and a two-fluid (separated fissile and fertile salts) molten salt reactor design were created as well as a number of other pre-conceptual designs. Although the reactor core for the one-fluid design was simpler than the two-fluid design, the salt processing was more complicated because of the need to separate fission products and small amounts of higher actinides from the thorium and protactinium in the salt.

[1] According to the Nuclear Energy Innovation Capabilities act of 2017 (PL 115-248), an advanced nuclear reactor is “a nuclear fission reactor with significant improvements over the most recent generation of nuclear fission reactors”

1.2.1 Original LFTR design

The original LFTR design drew heavily from the design of the 1967 two-fluid MSBR. Since the MSBR project was canceled before a demonstration plant was constructed, FEI sought to restart and continue the exploration into MSBRs. The refreshed concept was originally published in the 2015 Electric Power Research Institute (EPRI) report [4]. The 2015 LFTR design used a mixture of lithium fluoride and beryllium fluoride salts as carriers for the nuclear material. Then, thorium fluoride salt was added to the blanket (fertile) material, and uranium fluoride salt was added to the fuel (fissile) salt.

The two-fluid reactor contains fuel salt and blanket salt fluids. Graphite is used as the neutron moderator. Irradiated blanket salt was sent to salt processing to remove protactinium and uranium, which was then held in a decay tank to allow ^{233}Pa to decay to ^{233}U. The uranium was extracted from the decay tank using a fluorinator, which was combined with uranium fluoride from the fuel salt processing. Uranium was continuously removed from the fuel salt using another fluorinator, which left fission products in the carrier salt. Thus, uranium could be selectively extracted (and combined with the uranium from the decay tank) before being reconstituted into refreshed fuel salt. For a more detailed explanation of the proposed salt processing, see the EPRI report [4].

1.2.2 2018 LFTR design

After the original LFTR design was presented in the 2015 EPRI report, FEI continued to iterate on the LFTR design, which resulted in the conceptual design shown in Figure 2. For the remainder of this report, we shall call this new design the *2018 LFTR* design, although further iterations on the design are ongoing. Like the original design, the reactor core contains graphite channels in the center of the core, which the fuel salt flows through. Most of the uranium in the core is contained in the fuel salt, and thus, nuclear fissions primarily occur in the fuel salt. The blanket salt, which contains thorium, flows outside of and between the fuel salt channels.

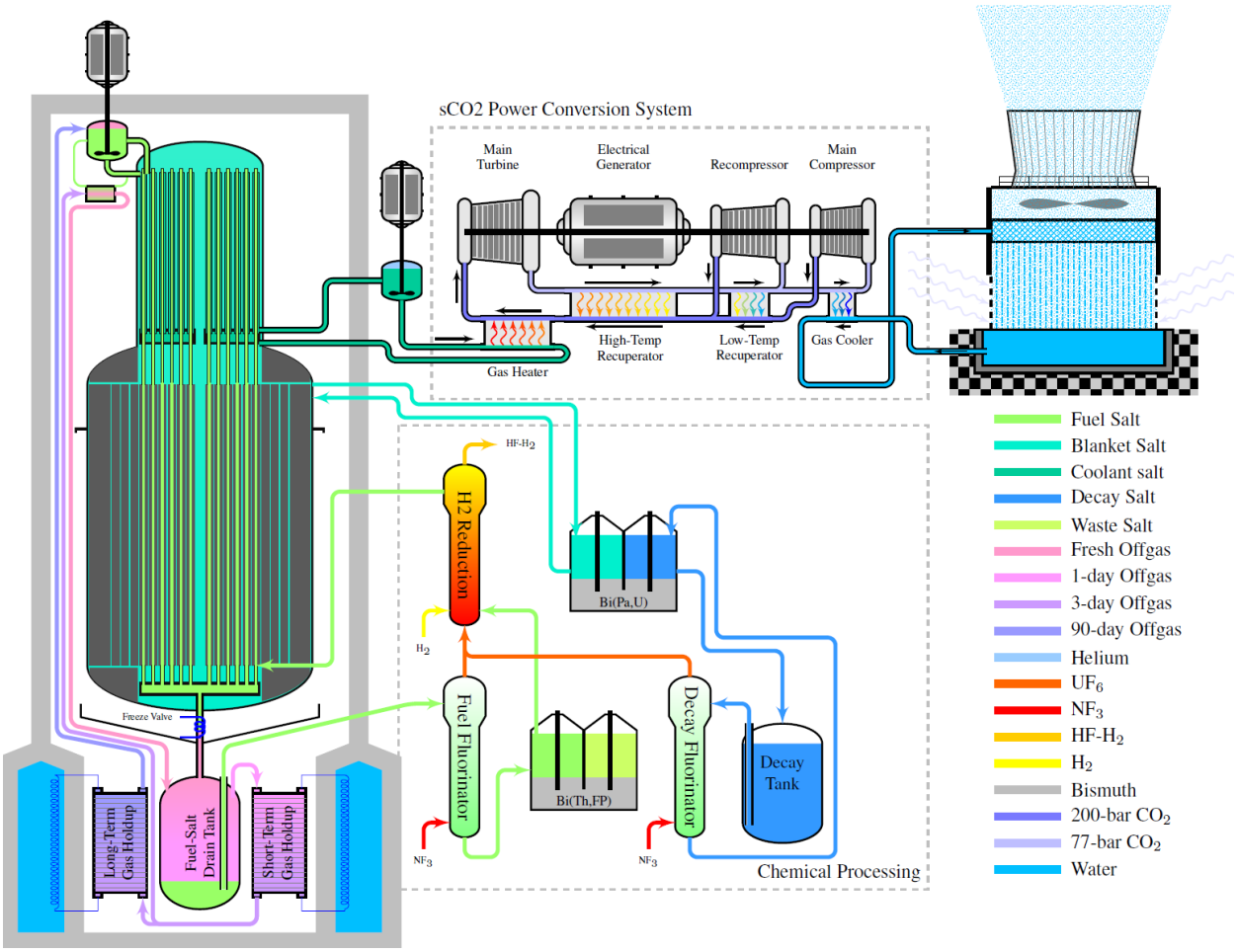

Figure 2. Flow chart of the 2018 LFTR design provided by FEI.

In FEI's conceptual design, a portion of the fuel salt (green) is continuously directed to salt processing, which removes fission products by reductive extraction. The fuel fluorinator is used to extract the uranium from the fuel salt before the salt proceeds to an electrolytic cell that uses bismuth reductive extraction, which removes the fission products from the residual salt. The uranium (orange) is then recombined with the cleaned salt using hydrogen reduction to produce refreshed fuel salt, which returns to the reactor vessel. The removed fission products are considered waste products and are further processed (not pictured in the figure) for long-term storage. Some uranium will likely exit the system in the waste stream(s), but this will be minimized to optimize the fuel use.

Additionally, a portion of the blanket salt (as shown in Figure 2 exiting near the top of the vessel in teal) is continuously sent to chemical processing, which removes protactinium and uranium from the blanket salt by reductive extraction in an electrolytic cell containing a consumable thorium metallic anode within a pool of metallic bismuth. FEI posits that an applied potential difference between the pool of metallic bismuth and the thorium metallic anode serves to drive any protactinium and uranium from the blanket salt into the bismuth pool while replacing those elements with thorium etched from the anode. The reduced protactinium and uranium species are then moved from the bismuth pool into the decay salt (blue) and held within a decay tank, where radioactive decay of protactinium to uranium occurs. Uranium is removed from the decay tank using the decay fluorinator where it is combined with the fluorination stream from the fuel salt (orange). The combined stream is then reduced and returned to the reactor vessel.

There are several notable differences between the 2018 LFTR and the original LFTR design. The most obvious is that the heat transfer system was moved to the upper portion of the reactor vessel, which allowed increased efficiency and a reduction in the total fissile mass in the reactor system.

In addition, the salt processing (protactinium and uranium extraction) was modified to use an electrolytic cell instead of reductive columns. The electrolytic cell used for blanket salt processing has the advantage of feeding the fertile thorium into the reactor as a solid metal rod from which thorium ions are etched. This process allows direct measurement of the thorium mass entering the system by measuring the current through the electrochemical cell, which has advantages for MC&A. According to FEI's ongoing research efforts, this electrolytic technique also appears to be a more selective technique than the prior reductive extraction via bismuth reductive columns. The disadvantage of the prior technique was that it extracted most of the thorium, in addition to the protactinium and uranium, from of the blanket salt. However, it is advantageous to avoid pulling thorium out of the blanket salt so more thorium returns to the reactor vessel for eventual neutron capture.

Note that the waste streams are not shown in the figure. The nonvolatile fission products are primarily removed by the electrolytic cell located between the fuel and decay fluorinators in the figure, while volatile fission products are removed by a helium off-gas system.

2. INTRODUCTION TO MC&A ASSESSMENT

2.1 GOAL

This MC&A assessment was performed to understand the LFTR design features that influence nuclear material accountancy. Consequently, one outcome of this assessment was to identify opportunities for the LFTR design and/or operations to optimize the implementation of MC&A and, therefore, minimize associated costs. Optimizing the implementation of MC&A will increase the effectiveness and efficiency of detecting abrupt and protracted theft or diversion of SNM [2] from authorized locations and processes within a future LFTR facility.

Another goal of this work was to provide recommendations to support FEI's revision of the existing LFTR design, which consider nuclear material accountancy and progresses LFTRs toward meeting NRC MC&A licensing requirements. In addition, this work enables future assessments to consider implications to the application of IAEA safeguards. The outcomes of this work were recommendations on the existing process flow sheet for the 2018 LFTR design, which led to FEI's generation of a revised LFTR design in 2021. Another outcome of this assessment was to identify measurement locations for the implementation of new MC&A technologies or approaches for potential development.

ORNL acknowledges that this study was completed assuming compliance with existing regulations of the NRC which were developed for the existing solid-fueled US reactor fleet based on low-enriched uranium. New regulations will likely be developed by the NRC to reflect MC&A challenges raised as new reactor designs (e.g., liquid fueled) and associated fuel cycles develop.

2.2 MC&A CONSIDERATIONS FOR ADVANCED REACTORS AND THEIR ASSOCIATED FUEL CYCLES BASED ON THORIUM/^{233}U

Thorium is under consideration for a fertile material for advanced nuclear reactors because it is relatively abundant compared to uranium and has advantageous nuclear properties [9]. Advanced reactor designs such as LFTR differ significantly from the current fleet of light and heavy water reactors by using molten salt fuel (i.e., liquid fuel). Nuclear material accountancy approaches, concepts, and measures are well developed for conventional nuclear reactors and their associated fuel cycle facilities operating on a uranium–plutonium fuel cycle. However, nuclear material accountancy approaches, concepts, and measures need to be developed for advanced reactor designs employing a thorium/^{233}U fuel cycle. The evaluation of a specific design such as the LFTR can proactively address these challenges.

The neutron irradiation of ^{232}Th leads to the production of ^{233}U. Figure 3 showcases some of the important reactions in this process [9]. The most direct pathway is the (n, γ) neutron capture reaction from ^{232}Th to produce ^{233}Th, which quickly decays to ^{233}Pa. The protactinium then β-decays with a half-life of about 27 days to ^{233}U.

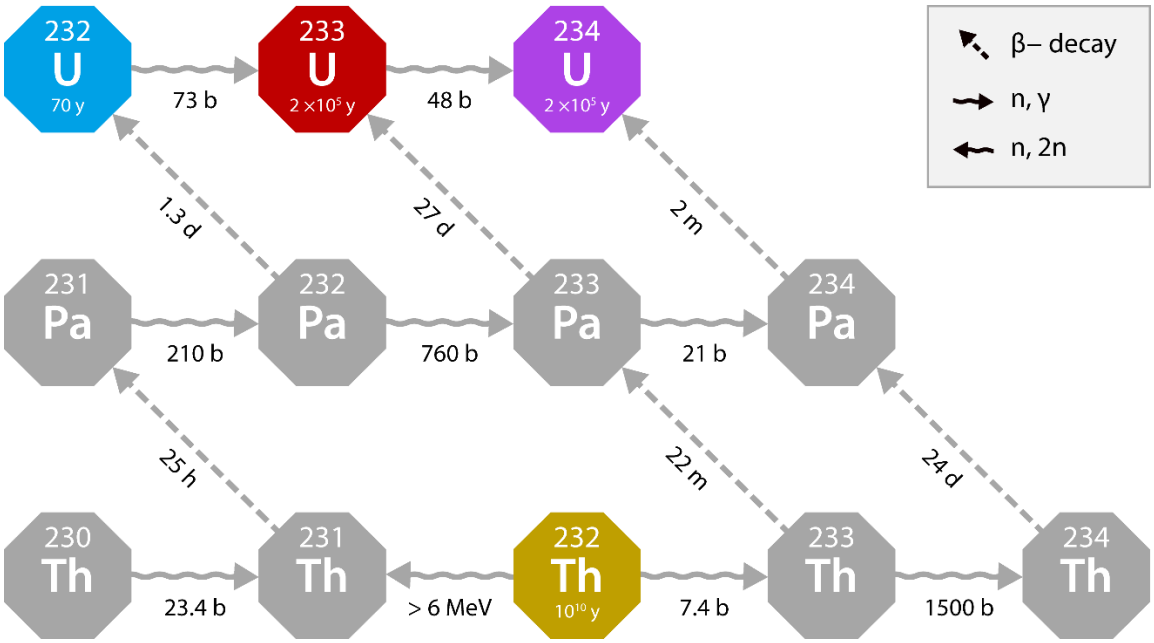

Figure 3. Uranium-233 production by the neutron irradiation of thorium-232.

Many advanced reactor concepts rely on this conversion of ^{232}Th to ^{233}U. The designs range from modifications to pressurized water reactors (PWRs) to MSRs. Some of the reactor designs breed 100% of the fuel needed for operation of the reactor, but others supplement the fuel with more traditional ^{235}U or plutonium. The broad range of potential designs creates several challenges for MC&A.

One of the challenges facing MC&A for all proposed thorium fuel cycles (i.e., not limited to MSRs) is that current detection technologies are tailored for the traditional uranium–plutonium fuel cycle, which includes detecting ^{235}U. Moreover, the induced fission cross section for ^{233}U and ^{235}U are similar, which makes it difficult to differentiate these isotopes using active neutron interrogation techniques, especially for reactor designs that use a mixture of fissionable isotopes. Another challenge for providing MC&A for advanced reactors in general is the range of fuel forms, which range from traditional fuel rods to spheres or pellets to molten salts.

2.3 OTHER NUCLEAR MATERIAL ACCOUNTANCY CHALLENGES OF THE LFTR

The LFTR design contains several potential new challenges for nuclear material accountancy different from the current US fleet of nuclear reactors. The first is that the fuel salt contains relatively high-^{233}U-content, which is potentially attractive SNM. Additionally, this nuclear fuel is also in the form of a liquid (i.e., molten salt), some of which exists outside of the reactor vessel and thus might be a potential target for theft or diversion. Another difference from the current fleet of US nuclear reactors is that LFTR can operate continuously with no need to shut down for refueling. Therefore, physical inventory and material balance measurements will need to be performed either while the reactor is generating power or during maintenance activities. These measurements are made more difficult by the continuous chemical and physical processing of fuel salts outside the reactor vessel, which is again different than currently operating reactors. These challenges are potentially offset by other differences, such as the salt inventories, are well-mixed in the LFTR design; consequently, salt samples are likely more representative of the whole inventory. Combining salt processing and reactor operation into a single facility adds additional challenges because different types of accountancy measures are used for fuel fabrication (fuel synthesis), reprocessing (separations), and nuclear reactors.

Furthermore, the LFTR is designed to operate using a thorium-based fuel cycle instead of the uranium–plutonium cycle used by current reactors. Thorium is considered source material and is not considered SNM as defined under Title 10 of the *Code of Federal Regulations* (CFR) Part 74 [2] "Material control and accounting of special nuclear material." Although thorium is not historically regulated under 10 CFR Part 74 for this reason, thorium tracking requirements do exist under 10 CFR Part 40 [10]. Design-specific MC&A requirements and the level of control of thorium (including the stage in the nuclear energy system at which the thorium is tracked) will need to be evaluated for the LFTR design. In other words, because SNM will be generated from thorium during reactor operation, the level of tracking and control may need to be modified. As the thorium fuel cycle evolves commercially, it is possible that the NRC could adopt an approach like the US Department of Energy (DOE), which does control thorium under MC&A regulations [11].

The ^{233}U inventory in the final LFTR design and its components will determine the material categorization for both MC&A and physical protection, as defined by the US NRC [3]. The calculated inventory level of ^{233}U in the reference design is 285 kg at equilibrium. While it can be concluded that there is a large quantity of ^{233}U in the fuel salt of the reference design, the diluteness and high radiation levels could be considered in future when making a final determination of whether the diversion of significant quantities of nuclear material is or is not credible in the final LFTR design. Therefore, these factors could potentially be taken into consideration when determining the physical protection and MC&A requirements for a final LFTR design, as discussed in reference [12]. The decay tank would likely be considered separately from the fuel salt, given the lower radiation levels, and thus points to a need to consider additional design solutions for both physical protection and MC&A. It is recommended that any final design solutions for the final LFTR design, including the fuel salt and decay tank, should consider nuclear material categorization and work towards reducing that categorization (e.g., from Cat I to Cat II for NRC categorization for MC&A) as opposed to increasing the categorization. The categorization will impact the physical protection and MC&A requirements applied to the final LFTR design and methods implemented within that final design. For completeness, a discussion of a graded safeguards approach for ^{233}U is given in reference [13]. However, note that DOE MC&A regulations are different from US NRC MC&A regulations.

Despite these challenges, LFTR offers potential MC&A benefits. High radiation fields can reduce the vulnerability for theft and diversion and potentially simplify the MC&A and security requirements. However, high radiation fields would potentially complicate access for IAEA safeguards inspections and placement of monitoring equipment. Another potential benefit is that if successfully deployed, LFTR would not require shipment of enriched nuclear material to the reactor site or on-site enrichment once the reactor is operating. However, the startup fuel will likely require enrichment for the fissile driver depending on design choices.

2.4 NUCLEAR MATERIAL ACCOUNTING CONSIDERATIONS FOR LFTRS—RESEARCH QUESTIONS

The following research questions are posed and addressed in this report. Note that the answers to these questions are dependent on the availability of specific design information on LFTR. Where no design information could be provided, the ORNL team adjusted the analysis accordingly and made assumptions (documented in Section 11) to complete the work.

1. What are the main design features of LFTR? (Section 3.6)
2. What are the nuclear material transfers, movements, and operations within LFTR? (Section 3.6)
3. What are the starting conditions (fuel and blanket salt compositions) and operating conditions (full power) for LFTR? (Section 3.6)

4. What is the total nuclear material inventory within a LFTR when operating under equilibrium conditions and how do operational choices and variations influence that inventory? (Section 4.3)
    i. Recognizing that ^{233}U is produced in the blanket and consumed in the reactor fuel, how much ^{233}U does LFTR produce and what is the isotopic composition of the uranium?
    ii. What is the equilibrium isotopic composition (uranium and plutonium vectors) of the fuel and blanket salts within the LFTR core?
    iii. What is the isotopic composition of the salt within the decay tank?
5. How does the equilibrium nuclear material inventory within LFTR compare in different system components? (Section 4.3)
    i. Reactor core
    ii. Blanket salt processing
    iii. Decay/drain tank
        a. What is the decay tank inventory?
        b. What is the rate of significant quantity production in the decay tank?
    iv. Fuel salt processing/feed
    v. Storage/heat exchanger
        a. The integrated primary heat exchanger is planned to reduce the overall salt inventory. The system is integrated into the vessel instead of a separate system (i.e., salt is heated in the core then heat is removed without needing to leave the vessel). The design is not yet finalized, so it is not yet modeled. In theory, it allows the neutronics and heat exchange to be separated to allow for better material selection, but not quite as well as if the units were completely separate.
    vi. Flush salt (non-nuclear flush salt picks up isotopes from the wetted surfaces of the components after the fuel salt is drained.)
6. How do LFTR operational choices impact nuclear material accounting? (Section 4.3)
    i. Residence times within the decay tank are considered.
7. How self-protecting is the nuclear material within LFTR? (Section 4.3)
    i. How much fuel salt (volume) equates to one significant quantity of ^{233}U at equilibrium and during normal operations?
    ii. How much blanket salt (volume) equates to one significant quantity of ^{233}U at equilibrium and during normal operations?
    iii. What is the dose from a cylinder containing a small salt sample at surface contact, at 1 foot, and at 1 meter?
    iv. What is the dose from a cylinder containing a salt volume equivalent to one significant quantity of ^{233}U?
8. How do variations in the main design features of the LFTR influence approaches to nuclear material accounting and thus, potentially IAEA safeguards (nuclear material security, acquisition pathways, and verification)? Furthermore, how can the main design features of a LFTR be varied to have a net benefit on nuclear material accounting? (Section 5.1) This analysis describes the features of the flow sheet in addition to modeling the below in detail:
    i. Blanket salt composition (four different salt matrices modeled)
    ii. Options to modify the decay tank salt composition were considered.
    iii. Options on eliminating (or diluting) pure streams of ^{233}U from the reactor design were analyzed.

iv. Changing the mixing point between the decay fluorinator and the fuel fluorinator was analyzed to reduce the potential for ^{233}U diversion.

9. Based on the above MC&A challenges that the current design introduces, how can FEI make early design changes (physical, operational, etc.) to modify the design to reduce security and MC&A costs such that it can be built at a price point to be competitive? (Section 6.1)
10. What technology gaps exist (if any) to ensure appropriate MC&A? (Section 6.2.2)
11. This work can provide the basis for future work that considers how IAEA safeguards might be applied to LFTR. For example, what concepts, approaches, measures, technologies, and inspection frequency requirements are needed? (Section 6.2.3)

3. MC&A ASSESSMENT METHOD: LFTR MODEL

To evaluate the LFTR design, ORNL developed a reactor physics model to represent the 2018 LFTR design. Primarily, the model enabled the calculation of the LFTR nuclear material inventories at equilibrium, which informed the preliminary MC&A assessment. This section presents the main design features of LFTR as well as the nuclear material movements and operating conditions within the reactor system. This information is used to answer some of the research questions outlined in Section 2.4.

During much of this work, the parameters of the 2018 LFTR core design concept and their dimensions were still being developed by FEI and were in flux. Therefore, as a reference model, the team selected the 1967 ORNL two-fluid MSBR, which was the precursor to the original LFTR design. The 1967 two-fluid MSBR design was well documented and allowed for sufficient neutronic modeling to support general analysis and conclusions applicable to LFTR. Note that future iterations of this analysis using a model more closely resembling the LFTR design will likely result in small changes to the output, which will influence the conclusions made from those results.

The remainder of this section presents the neutronic model used for the MC&A evaluation of the LFTR. Section 3.1 discusses the major design features of the LFTR. Section 3.2 summarizes the major nuclear material movements and process operations in the system. Section 3.3 summarizes how the nuclear movements were modeled. Sections 3.4 and 3.5 present the model geometry and parameters, respectively. Finally, Section 3.6 contains a summary of the information as it relates to the research questions from Section 2.4.

3.1 1967 ORNL TWO-FLUID MSBR AS THE PRECURSOR TO LFTR

As stated in Section 1.2, the 1967 ORNL two-fluid MSBR [7] was used as a reference for the anticipated LFTR design to protect proprietary design information while iterating on the LFTR design. In the 1967 two-fluid MSBR reactor vessel design, the fuel salt enters the core through the bottom. It then travels through the hexagonal graphite fuel cells (upward in the inner channel and downward in the outer channel) to exit through the bottom of the vessel. The graphite cells comprise an outer hexagonal tube with an inner cylindrical tube. The fuel salt flows into the core in the channel between these tubes and out of the core through the center of the cylindrical tube.

The blanket salt enters the reactor vessel near the top of the core, and the flow is directed downward along the inside of the vessel wall. The salt collects at the bottom of the core where it is directed upward through the interstitial space between the graphite fuel cells as well as through graphite tubes surrounding the fuel cells. The blanket salt exits the reactor vessel near the top of the core.

By separating the fuel and blanket salts, the two-fluid design is more complex than a single fluid design but has simplified salt processing. Thorium-232 in the form of fluoride salt is added to the blanket salt, which is irradiated to produce ^{233}Pa. The protactinium is then removed from the blanket salt for processing. Protactinium-233 is the short-lived (half-life ~27 days) precursor to ^{233}U. The ^{233}Pa is allowed to decay in a holding tank to ^{233}U, which is transferred to the fuel salt to undergo fission. Nearly all fissions occur in the fuel salt, and thus fission product removal is only necessary for the fuel salt.

As the LFTR design is continually evolving while it remains under development, the 1967 two-fluid MSBR was chosen as the starting point for evaluating the LFTR design. Compared to the 1967 two-fluid MSBR design, the LFTR is planned to have similar reactor power (600 MW_{th} compared to the 575 MW_{th} of the 1967 two-fluid MSBR) as well as similar fuel salt composition. FEI is considering different options for the blanket salt composition, including removing beryllium fluoride and possibly adding sodium fluoride.

Much of the salt processing system design is under consideration as FEI work to build upon the salt chemistry explored by the original 1967 ORNL two-fluid MSBR design. Due to the relatively unknown chemistry, the salt processing will largely be treated as a "black box" process assuming the efficiencies and removal times suggested by the 1967 two-fluid MSBR report [7].

For this work, the following removal constants were assumed: (1) from the blanket salt, protactinium and uranium were removed with 96% efficiency over a 3 day period; (2) from the fuel salt, noble gases and metals were removed with 90% efficiency over a 14.2 second cycle, and other fission products were removed with 99% efficiency over a period of 60 days. In addition, the reactor was assumed to be a "second generation" LFTR initialized using equilibrium fuel from another LFTR, and transient conditions (e.g., startup and shutdown) were not considered in this work. The specific details of initial fuel enrichment (fissile driver), if not obtained from another LFTR, and subsequent fuel fabrication (i.e., salt synthesis) processes were also unknown. Note that in the basic fuel cycle for the LFTR design, it is assumed that the initial reactor must obtain fissile material from an external source for startup and to reach equilibrium. However, the ^{233}U bred from this initial reactor can then be used as the fissile driver to start up subsequent LFTRs ad infinitum.

3.2 LFTR SYSTEM DESIGN AND FLOW DIAGRAM

A simplified process diagram of the LFTR is shown in Figure 4. The process is designed to convert fertile ^{232}Th into fissile ^{233}U, which is used as fuel. The salt processing is greatly simplified in the figure, but the movement of important isotopes is captured. The thorium feedstock enters the blanket salt processing by reductive replacement of protactinium and uranium, (i.e., each atom of protactinium and uranium removed from the blanket salt during processing is replaced by thorium). Note this point has MC&A implications and is discussed in Section 5.1.3. The removed isotopes are moved into a decay tank to allow the ^{233}Pa to decay into fissile ^{233}U, which is a crucial step because if the ^{233}Pa is left in the blanket salt it can absorb an additional neutron and will not result in a ^{233}U daughter. Meanwhile, processed blanket salt travels in a loop to the reactor vessel where ^{232}Th is converted to ^{233}Pa by neutron irradiation within the reactor core before returning to the blanket salt processing.

The uranium from the decay tank enters the fuel salt loop where it becomes fuel for the reactor where it undergoes fission and produces fission products. Most of the fuel salt is cycled continuously to/from the heat exchanger (not pictured) to extract heat, and a small portion is routed for processing to remove fission products and other activation products created via neutron absorption. The 1967 two-fluid MSBR conceptual design routes less than 0.0003% of the fuel salt and 0.05% of the blanket salt to processing. However, the LFTR will likely differ from these values.

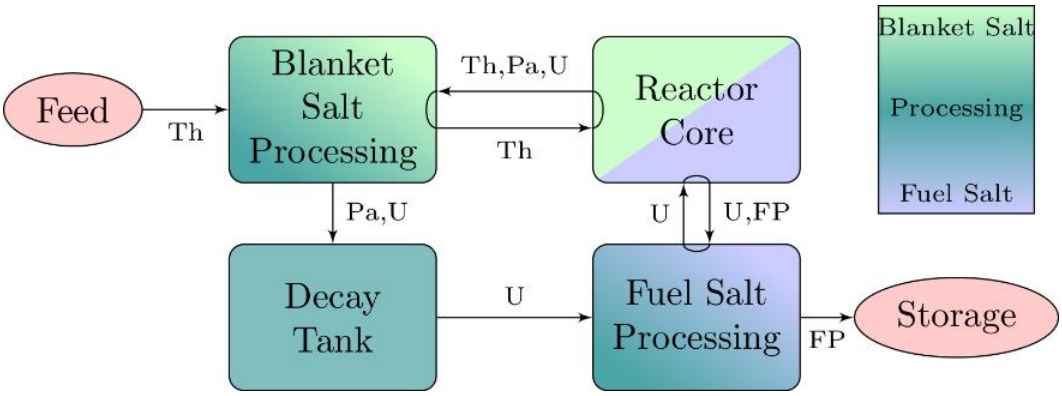

Figure 4. Simplified fuel cycle flow chart of the LFTR design. Thorium enters the blanket salt, which is irradiated to form ^{233}Pa that decays into ^{233}U. These products are moved to a decay tank. Uranium is moved from the decay tank into the fuel salt. Fission products are continually removed from the fuel salt.

3.3 REACTOR PHYSICS MODEL DESCRIPTION

To evaluate the nuclear material inventory and material flows for a LFTR, a neutronics model of the reactor vessel was designed, which was based on the 1967 two-fluid MSBR vessel geometry. The modeling tool TRITON, which uses the Monte Carlo solver KENO-VI, from the SCALE package [14] was chosen to evaluate the neutronics of the reactor vessel.

New features in SCALE 6.3 allow for movement of material, which was used to simulate the effects of processing on the fuel and blanket salts. Notably, thorium was fed into the blanket salt, and the model moved the protactinium and uranium vectors from the blanket salt to decay outside the geometry, thus simulating the decay tank. Protactinium in the decay tank is converted into uranium.

Ideally, uranium from the decay tank would be moved successively into the fuel salt in the model, but such a chain of material movement was not recommended by the SCALE team to ensure conservation of material. Instead, a uranium feed into the fuel salt equal to the production rate of uranium in the decay tank was created to mimic the ideal behavior of material being transferred (i.e., uranium accumulated in the decay material while the fuel salt was supplied by a constant feed rate). Finally, fission products were moved from the fuel salt into waste streams.

The model was initialized at assumed steady-state conditions then allowed to deplete to 1,000 effective full-power days. Any differences between the initial and final states of the model were used to correct the initial conditions until the model converged (i.e., steady-state conditions were found). Primarily, the amount of uranium in the fuel salt, the uranium vector of the fuel salt, and the salt feed rates (thorium into the blanket salt and uranium into the fuel salt) were adjusted until convergence.

3.4 MODEL GEOMETRY

The dimensions for the reactor vessel and components were primarily taken from the 1967 two-fluid MSBR report [7]. An X-Y cross section of the reactor vessel is shown in Figure 5, where the 420 hexagonal fuel cells take up most the area. The vessel has a diameter of 14 ft, and the core diameter is 10 ft. The blanket section is approximately 15 in. thick, and the graphite reflector at the edge of the vessel is approximately 6 in. thick. At the center of the core is a graphite control rod that is used to make minor changes to reactivity; power is increased as the rod is inserted. Major changes to reactivity require adjusting the fuel salt composition.

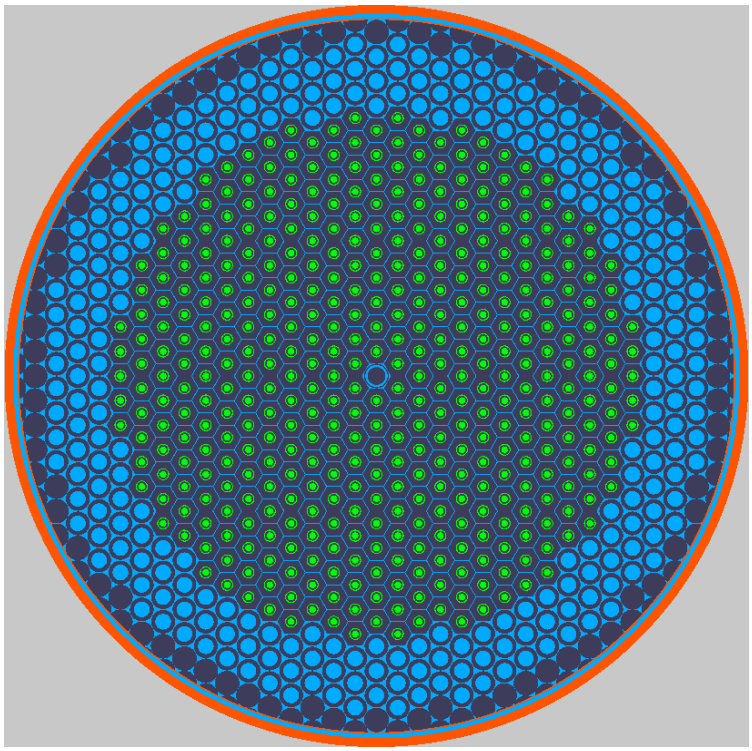

Figure 5. X-Y cross section of the reactor vessel model. Green is the fuel salt, blue is the blanket salt, dark blue is the graphite, and orange is Hastelloy.

Each 15.3 ft long graphite fuel cell is 5.375 in. from flat-to-flat edges and arranged on a 5.056 in. pitch. The graphite takes up 80.2% of the volume, and the fuel and blanket salts take up 13.4% and 6.4% of the volume, respectively. The 15 ft long blanket elements are arranged with the same pitch spacing but are cylindrical tubes 5.375 in. in diameter for volume fractions of 0.58 and 0.42 for blanket salt and graphite, respectively.

Figure 6 presents the Y-Z view of the model. The vessel was 19.5 ft long, with an active core length of 13.25 ft. Note that the top of the vessel is filled with helium and is not consistent with the stated design in the 1967 two-fluid MSBR report, which seemed to suggest the area was filled with blanket salt. However, to match the stated quantity of blanket salt within the entire reactor vessel volume more closely, the top region was instead filled with inert gas. The depletion model contained approximately 700 ft^3 of blanket salt and approximately 182 ft^3 of fuel salt.

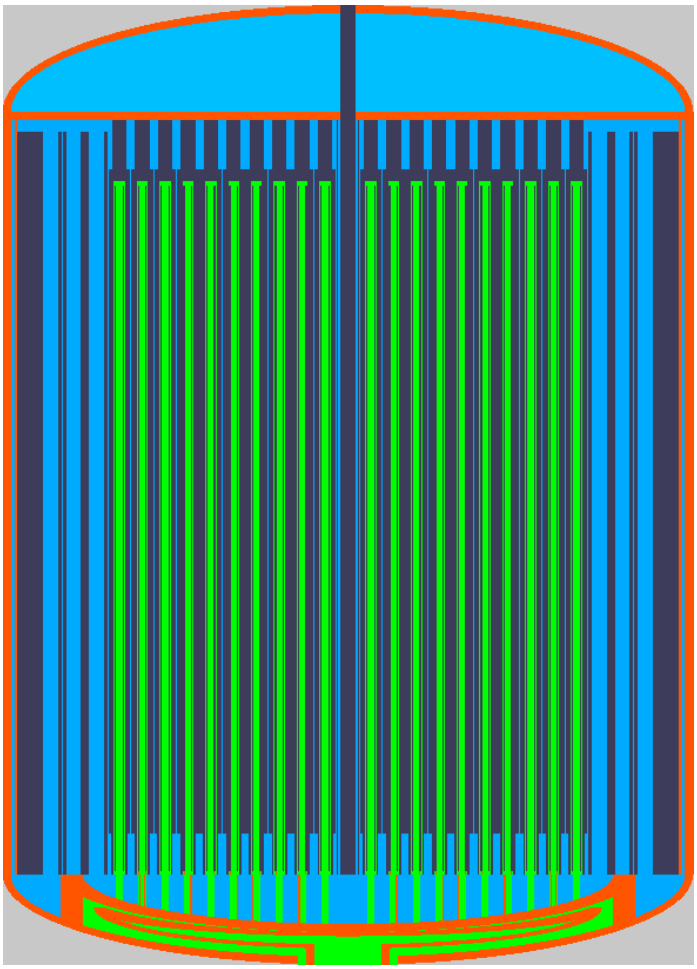

Figure 6. Y-Z cross section of the reactor vessel model. Green is the fuel salt, blue is the blanket salt, dark blue is the graphite, and orange is Hastelloy. The light blue at the top of the vessel is helium.

Nearly all dimensions are taken from Figures 5.1 and 5.2 of the 1967 ORNL two-fluid MSBR report [7]. Dimensions for the areas above and below the core were estimated from the figures, whereas the core region was well documented.

3.5 MODEL INPUT PARAMETERS

The fuel salt was composed of ^{7}LiF and BeF_2 salt mixed with UF_4 (66.44, 33.22, and 0.338 mol%, respectively). The uranium vector in the 1967 two-fluid MSBR report was stated to be 69% ^{233}U, 17% ^{234}U, 6% ^{235}U, and 8% ^{236}U. The reactor was operated at a power density of 19.890 MW/MT_{IHM} adjusted to account for only 49.6% of the fuel salt present in the reactor vessel.

In the 1967 two-fluid MSBR report [7], the blanket salt was a ternary mixture of ^{7}LiF, BeF_2, and ThF_4 (71, 2, and 27 mole%, respectively). However, FEI was also considering other blanket options including removing BeF_2. Additionally, NaF was proposed as an addition to the blanket salt. Therefore, four different compositions were modeled, which are summarized in Table 1.

Table 1. Blanket salt compositions in mole percent. Each option was simulated separately.

Design	Composition name	^{7}LiF (%)	ThF_4 (%)	BeF_2 (%)	NaF (%)
MSBR	FLiBeTh	71.54	26.46	2	0
LFTR	FLiTh	73	27	0	0
LFTR	FLiNaTh_low	58.907	30.557	0	10.534
LFTR	FLiNaTh_high	51.1	24.6	0	24.3

As mentioned, material flow parameters were defined to feed thorium into the blanket salt. Protactinium and uranium were moved from the blanket salt to the decay tank. Uranium was fed into the fuel salt. Fission products were moved from the fuel salt to waste streams. The flow parameters are used to modify the decay constants of the materials used by ORIGEN in the TRITON depletion sequence. The general formula for these adjustments is

$$\lambda = \frac{ln(1 - \text{fraction})}{\text{time}},$$

where the λ is the decay constant adjustment, *fraction* is the removal efficiency fraction, and *time* is the processing time in seconds. For the simulation, the decay constant adjustments were calculated using the removal times and efficiencies presented in Section 3.1.

After converging the model feed rates, approximately 13.5 mg/s of thorium was fed to the blanket salt, and 13.5 mg/s of uranium was fed to the fuel salt. Note that for the model inputs, these numbers were reduced by the fraction of blanket and fuel salts that were in the reactor vessel (i.e., 0.56 and 0.496). The uranium feed consisted of 99.98220% ^{233}U, 0.01779% ^{234}U, and the balance ^{232}U, which was the converged uranium vector in the decay tank.

3.6 ANSWERS TO LFTR DESIGN RESEARCH QUESTIONS

In this section, some of the research questions from Section 2.4 have been answered including discussing the major design features as well as the nuclear material movements and operating conditions. The 600 MW_{th} 2018 LFTR design and the 1967 ORNL two-fluid 575 MW_{th} MSBR design are similar. The fuel and blanket salts are kept separate in the two-fluid design, which allows for simplified salt processing at the cost of a more complicated core design. The fuel and blanket salts are mixtures of lithium and beryllium fluorides mixed with either uranium or thorium fluoride. The equilibrium enrichment of the fuel salt will be explored in the next section.

The reactor breeds ^{232}Th into ^{233}U, which is used as fuel. Thorium enters as a feed to the blanket salt, which is irradiated in the core to produce ^{233}Pa, which in turn decays to ^{233}U. The blanket salt is processed to move ^{233}Pa and ^{233}U from the blanket salt to the decay tank, where most of the protactinium decay occurs. The decay salt is fluorinated to remove uranium, which is transferred to the fuel salt, where it is consumed as nuclear fuel and produces fission products. These waste isotopes are continually removed from the fuel salt. In the next section, the nuclear material inventory results are presented.

4. MC&A ASSESSMENT RESULTS: LFTR NUCLEAR MATERIAL INVENTORY, RADIATION SOURCE TERM, AND DOSE CALCULATIONS

In this section, we use the reactor core model discussed in the previous section to explore the nuclear inventories in various parts of the reactor design. In this section, some of the research questions posed in Section 2.4 are answered. Firstly, the total nuclear material inventory within LFTR when operating under equilibrium conditions is calculated and compared across different system components in Section 4.1. The section goes on to discuss the impact of some operational choices on nuclear material accounting. Section 4.2 assesses the associated radiation fields and doses for different types and quantities of fuel salt that would be at potential risk for abrupt and protracted nuclear material diversion by simulating the neutron and gamma source terms resulting from a samples of material using MAVRIC [14], which is another tool in the SCALE suite of codes.

4.1 NUCLEAR MATERIAL INVENTORY CALCULATIONS

4.1.1 Steady-state uranium vector

The first step to evaluating the nuclear material inventories relied on converging the model input parameters, including the material feed rates as well as the isotopic composition of the fuel salt. Of note, ORNL-4528 [7] estimated that the fuel salt uranium isotopic composition was 69% ^{233}U, 17% ^{234}U, 6% ^{235}U, and 8% ^{236}U, whereas in conversation (private communication from K. Sorensen, FEI, to R.L. Reed and L.G. Evans, Oak Ridge National Laboratory, May 2021), FEI had originally estimated the uranium isotopic composition to be 90% ^{233}U, 9% ^{234}U, 1% ^{235}U, and 200 ppm ^{232}U. These two estimates along with the converged equilibrium estimate are compared in Figure 7 and Figure 8. From the figures, all three estimates converge with enough time, which suggests that the final composition is representative of the steady-state condition given the model assumptions.

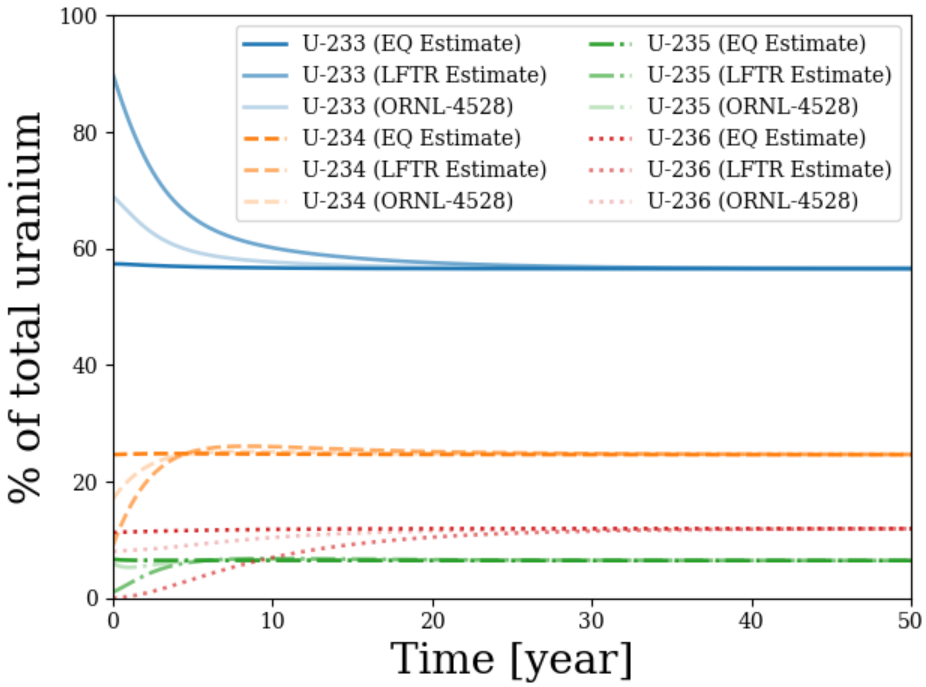

Figure 7. Uranium vector in the fuel salt as a function of LFTR operating time compared across different starting estimates.

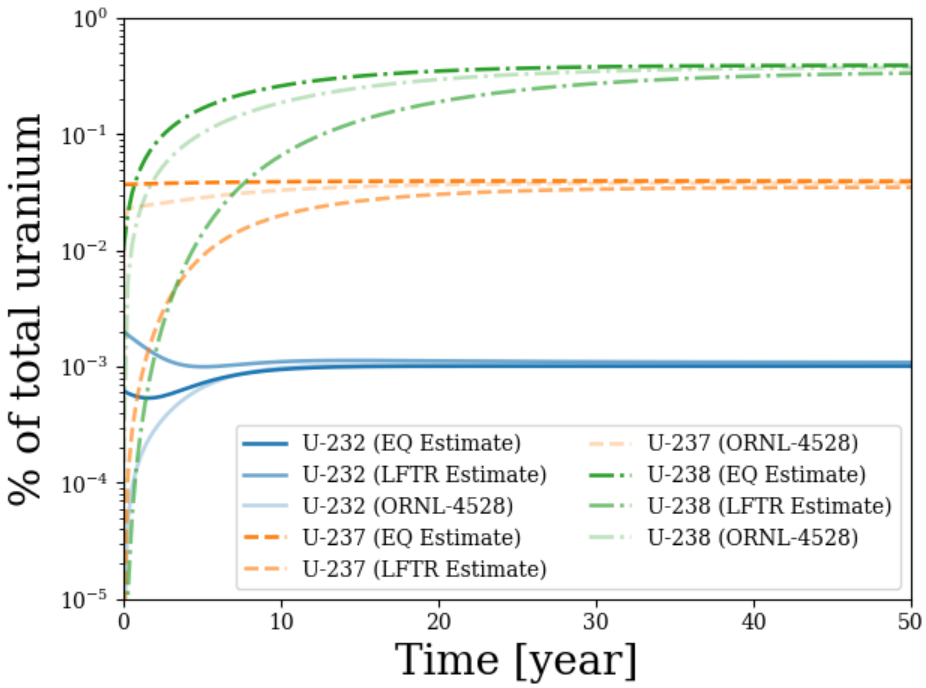

Figure 8. Uranium vector in the fuel salt as a function of LFTR operating time compared across different starting estimates.

These plots were created by using the f33 library created by KENO-VI to deplete the fuel salt using ORIGEN from the SCALE suite. As observed, both the ORNL-4528 report and FEI vectors overestimated the ^{233}U fraction. The model converges to the uranium vector shown in Table 2, which is lower than estimated at about 56.6% ^{233}U. The model suggests that neutron capture, which creates heavier uranium species, is more prevalent than previously estimated.

Table 2. Uranium vector in the fuel salt at equilibrium for the base case (FLiBeTh blanket salt).

^{232}U	^{233}U	^{234}U	^{235}U	^{236}U	^{237}U	^{238}U
10.1 ppm	56.59%	24.63%	6.44%	11.91%	0.04%	0.39%

To assess the potential perturbations to this equilibrium uranium vector, the additional blanket salt compositions proposed by FEI (shown in Table 3) were also modeled, which resulted in the fuel salt uranium vectors shown in Table 3. As shown, the different blanket compositions had very little effect on the equilibrium uranium isotopic composition in the fuel salt model.

Table 3. Uranium vector in the fuel salt at equilibrium for the various blanket salt compositions. The final column is the base case (FLiBeTh blanket salt).

	FLiTh	FLiNaTh_high	FLiNaTh_low	FLiBeTh
^{232}U	10.03 ppm	10.39 ppm	9.97 ppm	10.1 ppm
^{233}U	56.61%	56.64%	56.69%	56.59%
^{234}U	24.62%	24.61%	24.60%	24.63%
^{235}U	6.45%	6.45%	6.47%	6.44%
^{236}U	11.89%	11.90%	11.80%	11.91%
^{237}U	0.04%	0.04%	0.04%	0.04%
^{238}U	0.40%	0.37%	0.40%	0.39%

Another brief study explored how the equilibrium uranium vector changed as a function of temperature. Thus, for the base case (FLiBeTh blanket salt), the equilibrium uranium vector composition was computed for both the inlet and outlet fuel salt temperatures of 810.9 and 977.6 K, respectively. The corresponding uranium vectors are compared to the base case (temperature of 894.2 K) in Table 4. As shown in the table, temperature had a relatively small effect on the equilibrium uranium vector.

Table 4. Uranium vector in the fuel salt at equilibrium as a function of temperature.

	810.9 [K] (inlet)	894.2 [K] (average)	977.6 [K] (outlet)
^{232}U	10.05 ppm	10.10 ppm	10.10 ppm
^{233}U	56.56%	56.59%	56.62%
^{234}U	24.66%	24.63%	24.60%
^{235}U	6.42%	6.44%	6.45%
^{236}U	11.93%	11.91%	11.89%
^{237}U	0.04%	0.04%	0.04%
^{238}U	0.39%	0.39%	0.39%

4.1.2 Steady-state isotopic vectors

For completeness, the isotopic vectors for other important elements were computed for all cases. However, as in the uranium cases discussed in the previous section, blanket salt composition and temperature did not create significant differences in the isotopic vectors. Thus, only the base case (FLiBeTh blanket salt) is presented here. Tables 5–7 show the equilibrium vectors across material regions for uranium, plutonium, and protactinium.

Table 5. Uranium vector for base case at equilibrium for material regions.

	Fuel salt	Blanket salt	Decay tank
^{232}U	10.10 ppm	1.91 ppm	0.05 ppm
^{233}U	56.59%	99.37%	99.98%
^{234}U	24.63%	0.63%	0.02%
^{235}U	6.44%	0.00%	0.00%
^{236}U	11.91%	0.00%	0.00%
^{237}U	0.04%	0.00%	0.00%
^{238}U	0.39%	0.00%	0.00%

Table 6. Plutonium vector for base case at equilibrium for material regions.

	Fuel salt (%)	Blanket salt (%)	Decay tank (%)
^{238}Pu	52.42	28.05	0.00
^{239}Pu	13.86	2.61	46.03
^{240}Pu	8.12	3.3	50.08
^{241}Pu	6.66	2.97	3.17
^{242}Pu	18.86	15.39	0.00
^{243}Pu	0.01	0.00	0.00
^{244}Pu	0.08	47.61	0.00

Table 7. Protactinium vector for base case at equilibrium for material regions.

	Fuel salt (%)	Blanket salt (%)	Decay tank (%)
^{231}Pa	99.03	0.02	9.64
^{232}Pa	0.75	0.00	0.00
^{233}Pa	0.22	99.98	90.36

Although the tables in this section are useful for comparing isotopic ratios across the material regions, the various regions do not contain the same amount of each element. In the next section, elemental masses are presented for various locations in the reactor design.

4.1.3 Nuclear material inventory results

As stated in Section 3.5, equilibrium conditions for the 600 MW_{th} model required a uranium burn rate (and thus thorium replacement rate) of approximately 13.5 mg/s for convergence. The core model was divided into several material streams including fuel salt, blanket salt, decay tank, and waste. This enabled calculating the nuclear inventory for each of these streams. Further, the 1967 two-fluid MSBR report [7] states that 56% of the blanket salt and 49.6% of the fuel salt is in the core region. These values allow estimation of the nuclear inventory contained in the heat exchanger, pipes, and other components that were not included in the core model. Finally, the average residence time in the decay tank (and thus volume) was not specified in ORNL-TM-4528 [7]. Values of 30, 60, and 90 day residence times were assumed and corresponding nuclear material inventories are presented in this section.

As shown in Tables 8–10, under equilibrium conditions, most of the thorium resides in the blanket salt, and most of the uranium resides in the fuel salt. Most of the protactinium is in the decay tank. Note, as the average residence time in the decay tank increases, the volume of the tank and the nuclear material inventory also increase. Further, the inventory of protactinium is constant with respect to uranium production. This is because the rate of ^{233}U consumption is constant in the model, so the rate of ^{233}U generation (i.e., ^{233}Pa mass) is also constant. Uranium is the only element of interest that is affected by changing the average residence time in the decay tank. Finally, the fissile uranium mass is the sum of ^{233}U and ^{235}U.

Table 8. Nuclear inventory for decay tank with 30 day residence time.

		Fuel salt		Blanket salt		
	Total mass (kg)	**In core (%)**	**Out of core (%)**	**In core (%)**	**Out of core (%)**	**Decay tank (%)**
Pa	27.56	0.0	0.0	2.0	1.6	96.3
Pu	0.3	49.6	50.4	0.0	0.0	0.0
Th	95,707.2	0.0	0.0	56.0	44.0	0.0
Fissile U	317.67	49.0	49.7	0.0	0.0	1.3
Total U	501.59	49.2	50.0	0.0	0.0	0.8

Table 9. Nuclear inventory for decay tank with 60 day residence time.

		Fuel salt		Blanket salt		
	Total mass (kg)	**In core (%)**	**Out of core (%)**	**In core (%)**	**Out of core (%)**	**Decay tank (%)**
Pa	27.56	0.0	0.0	2.0	1.6	96.3
Pu	0.3	49.6	50.4	0.0	0.0	0.0
Th	95,707.2	0.0	0.0	56.0	44.0	0.0
Fissile U	321.78	48.3	49.6	0.0	0.0	2.6
Total U	505.71	48.8	49.6	0.0	0.0	1.6

Table 10. Nuclear inventory for decay tank with 90 day residence time.

		Fuel salt		Blanket salt		
	Total mass (kg)	**In core (%)**	**Out of core (%)**	**In core (%)**	**Out of core (%)**	**Decay tank (%)**
Pa	27.56	0.0	0.0	2.0	1.6	96.3
Pu	0.3	49.6	50.4	0.0	0.0	0.0
Th	95,707.2	0.0	0.0	56.0	44.0	0.0
Fissile U	325.89	47.7	48.5	0.0	0.0	3.8
Total U	509.82	48.4	49.2	0.0	0.0	2.4

4.2 DOSE CALCULATION RESULTS

To evaluate any self-protecting capabilities for the nuclear materials used in the LFTR design, commensurate with nuclear material security and MC&A considerations, a simple model was used to evaluate the neutron and gamma-ray photon dose from a small sample. To begin, 100 mL of the target material was modeled as a solid sphere in MAVRIC [14], which represented a small sample taken for monitoring salt chemistry. Note this sample size is likely much larger than any required for modern measurement techniques. Then detection regions were placed at the sphere surface, 30.48 cm (1 ft) from the surface, and 1 m from the surface to represent sample contact and standoff distances. Track-length tallies in conjunction with the 1991 ANSI standard flux-to-dose-rate factors [14] resulted in the dose rates shown in Table 11. The radiation source for these calculations was generated using ORIGEN.

Table 11. Dose rates for 100 mL of LFTR fuel/blanket salt compared to 100 mL of irradiated PWR fuel. All values are with no cooling time (i.e., immediately after removal from the core).

	Distance	Neutron dose rate (rem/h)	Gamma dose rate (rem/h)
Fuel salt	0 ft	2660 ± 1.5%	5,146,000 ± 0.02%
	1 ft	13.0 ± 1.3%	25,340 ± 0.02%
	1 m	0.157 ± 1.3%	307.0 ± 0.02%
Blanket salt	0 ft	9.27 ± 3.4%	81,090 ± 0.02%
	1 ft	0.0442 ± 2.5%	390.1 ± 0.02%
	1 m	0.00056 ± 2.5%	4.73 ± 0.02%
Reference PWR	0 ft	9901. ± 1.5%	10,480,000 ± 0.03%
	1 ft	47.98 ± 1.3%	47,880 ± 0.03%
	1 m	0.581 ± 1.3%	579.9 ± 0.03%

From the table, the fuel salt dose rates are approximately 20% of the corresponding doses resulting from irradiated PWR fuel. In this case, the PWR reference is assuming 4% enriched UO_2 irradiated to 3 GWd of burnup with cladding removed. Note that the values in the table are the result of zero cooling time, which means that short-lived fission products increase the dose rate. Table 12 shows the same materials' dose rate after 3.65 days of cooling.

Table 12. Dose rates for 100 mL of LFTR fuel/blanket salt compared to 100 mL of irradiated PWR fuel. All values are with 3.65 days of cooling time

	Distance	Neutron dose rate (rem/h)	Gamma dose rate (rem/h)
Fuel salt	0 ft	0.061 ± 20.4%	107,000 ± 0.02%
	1 ft	0.00029 ± 17.8%	530. ± 0.01%
	1 m	3.53e-06 ± 17.8%	6.42 ± 0.01%
Blanket salt	0 ft	0.00	9650 ± 0.02%
	1 ft	0.00	46.5 ± 0.01%
	1 m	0.00	0.564 ± 0.01%
Reference PWR	0 ft	0.00	894,000 ± 0.03%
	1 ft	0.00	4090 ± 0.02%
	1 m	0.00	49.5 ± 0.02%

Note that the dose rates for the decay tank salt (see the blue salt in Figure 2) were not computed as part of this work. Because the salt processing chemistry is not well specified or understood, the decay salt from the model did not include any processing elements (i.e., those used for the extraction of protactinium and uranium from the blanket salt). Since the mass and composition of the decay tank could vary widely based on salt chemistry, accurate dose calculations were not feasible because the radiation source strength and spectra of a 100 mL sample depend on the tank composition. However, it is likely that the dose from a decay tank sample would be greater than the blanket salt sample because of the absence of thorium in the decay tank, which would lead to a sample with a larger fraction of ^{232}U, which is a major contributor to the dose rate in the blanket and decay salt because of its decay progeny, which are high-intensity, high-energy gamma emitters (e.g., ^{208}Tl).

Figure 9 presents how the gamma-ray doses vary over time. The figure also shows a cutoff for self-protection of 100 rem/h, which is recommended by the US NRC at a distance of 1 m [3]. Using these criteria, a 100 mL sample of salt (e.g., fuel, blanket, decay) is only self-protecting on the order of days.

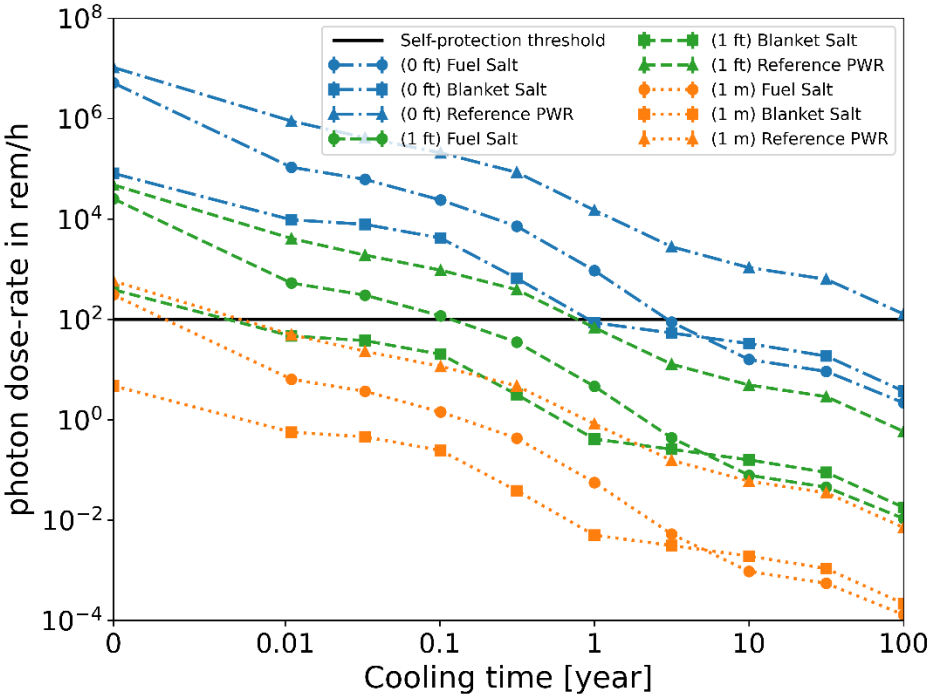

Figure 9. Photon dose rate from 100 mL samples of select material as a function of cooling time.

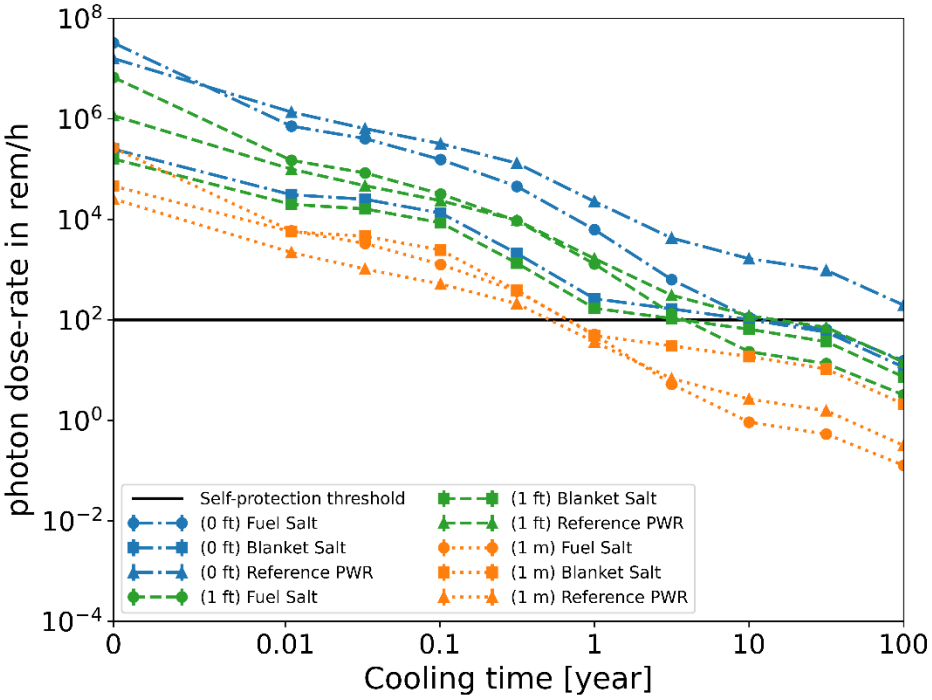

Figure 10. Photon dose rate from sample of select material containing 8 kg of ^{233}U + ^{235}U + ^{233}Pa as a function of cooling time. Sample volumes were 266.4 L for fuel salt, 275,800 L for blanket salt, and 21.9 L for reference PWR.

Dose calculations were also performed for volumes of material containing a significant quantity (SQ) of nuclear material. In this case, 8 kg of $^{233}U + ^{235}U + ^{233}Pa$ was selected to account for fissionable material. For conservatism and for simplification, the mass of one SQ for ^{235}U was assumed to be 8 kg (not 25 kg), and ^{233}Pa was considered the same as ^{233}U because of the short half-life for its decay to pure ^{233}U. Figure 10 presents the time-dependent dose calculations for the two salt and the reference materials at the distances of interest. For these calculations, 1 SQ of material required 266.4 L (541 kg) of fuel salt, 275,800 L (1.22×10^6 kg) of blanket salt, or 21.9 L (219 kg) of reference PWR fuel (irradiated to 3 GWd burnup). At a distance of 1 m, the samples remained above the self-protection threshold until about 1 year. As previously mentioned, doses for the decay salt were not computed because of large uncertainties in the decay salt composition, but the dose is likely between the fuel- and blanket-salt rates. Finally, the NRC uses a formula quantity to assess amounts of SNM [15], which is computed as the mass of ^{235}U plus 2.5 times the mass of ^{233}U + Pu. Counting ^{233}Pa as equivalent to ^{233}U, the 1 SQ samples contained 18.80, 19.97, and 21.68 kg of formula-quantity SNM for fuel salt, blanket salt, and reference PWR, respectively.

4.3 ANSWERS TO NUCLEAR MATERIAL INVENTORY AND DOSE RESEARCH QUESTIONS

In this section, material inventory and dose calculations were presented for the 1967 two-fluid MSBR model, which was used as the reference model for the 2018 LFTR design. This model and the resulting data were used to answer the research questions posed in Section 2.4 related to nuclear inventory.

The starting (transient) conditions were not explored in this report, but the uranium vector for steady-state operation was found to be about 56.6% ^{233}U, which is significantly lower than the estimated vector from ORNL-4528 (69% ^{233}U). ORIGEN was used to show how two nonequilibrium uranium vectors converge over time.

The total nuclear material inventory within LFTR when operating under equilibrium conditions was approximately 500 kg of uranium (285 kg ^{233}U and 32 kg ^{235}U) and 95 MT of thorium. Approximately half of these amounts are within the core, and the remainder is in piping or the salt processing. The reactor consumes (thus must produce) about 13.5 mg of ^{233}U per second.

At equilibrium, most of the uranium is in the fuel salt, most of the thorium is in the blanket salt, and most of the protactinium is in the decay tank. The uranium vector in the blanket and decay salts was nearly pure ^{233}U, but in the fuel salt, only about 56.6% of the uranium was ^{233}U. Depending on the decay tank residence time, around 1 SQ of uranium was present in the decay tank.

Variations in the blanket salt composition and the fuel salt temperature were explored for potential consequences on to nuclear material accounting. However, the equilibrium isotopic concentrations and material inventories were not strongly dependent on these variables.

One operational choice that impacts nuclear material accounting is the residence time in the decay system. The decay tank residence time influences the amount of uranium held in the tank. A 30 day residence time leads to approximately 0.5 SQ of ^{233}U in the tank, whereas a 90 day residence time leads to approximately 1.5 SQ of ^{233}U in the tank.

Potential abrupt and protracted nuclear material theft and diversion scenarios for LFTR will be affected by the radiation field and doses associated with the material. Therefore, the doses resulting from irradiated samples were modeled. Since ^{233}U makes up approximately 1.32% of the fuel salt mass, approximately 602 kg (297 L at a density of 2.03 g/cm^3) are required to contain 1 SQ (8 kg) of ^{233}U. For the blanket salt, ^{233}U makes up approximately 0.00153 ppm of the mass. Thus approximately 52,300 kg

(11.8 m^3 at a salt density of 4.43 g /cm^3) are required to contain 1 SQ of ^{233}U. If the blanket salt sample contains 1 SQ of ^{233}U + ^{233}Pa, then only 1,220 kg (0.27 m^3 at a salt density of 4.43 g/cm^3) is required.

Small 100 mL samples of fuel and blanket salts were found to have a surface contact photon dose rate of 107,000 ± 0.02% rem/h for the fuel salt and 9,650 ± 0.02% rem/h for the blanket salt after 3.65 days of cooling. At a distance of 1 m, these values reduced to 6.42 ± 0.01% and 0.564 ± 0.01% for fuel and blanket salts, respectively. This is below the self-protection threshold of 100 rem/h at a distance of 1 m. However, 2,970 such sample containers of fuel salt would have to be diverted to amount to 1 SQ. After cooling, the neutron dose rate was below microrem/h levels. As presented in Section 4.2, samples containing 8 kg of ^{233}U + ^{235}U + ^{233}Pa, remained above the self-protection threshold for almost 1 year.

The isotopic concentrations and doses presented in this section were obtained from the MSBR model, which was chosen as a surrogate for directly modeling the LFTR design. In the next sections, these results will be used for a preliminary analysis considering nuclear material control and accountancy for the LFTR design to identify the areas of potential challenges and to suggest possible mitigation methods.

5. LFTR NUCLEAR MATERIAL ACCOUNTING SYSTEM DESIGN CONSIDERATIONS

The 2018 LFTR design and associated process flow diagram were revisited and discussed with FEI during an in-person meeting on June 29, 2021, to provide an analysis of the potential to design nuclear material accounting into the process flow sheet for both the reactor core and chemical processing system. This section answers the remaining research questions related to the influence of nuclear material movements and operations on nuclear material accountancy.

5.1 DESCRIPTION OF KNOWN NUCLEAR MATERIAL OPERATIONS AND MOVEMENTS WITHIN 2018 LFTR

Here the nuclear material operations and movements within the 2018 LFTR design are described. This description is derived based on discussion with FEI about nuclear material locations at a LFTR and nuclear material operations (e.g., transfers, receipts, storage, waste) distinct from the description of the chemical processing of these nuclear materials. For each of these descriptions, the analysis considers where nuclear material is expected to accumulate, either by design or accidentally.

Each of the sections below walk through the nuclear material operations, movements, and transfers throughout the LFTR. The 2018 LFTR process flow diagram is provided in Figure 2 for reference. Recommendations are provided for ongoing assessments and future work. MC&A measurements are considered for nuclear material accounting and potential monitoring approaches for nuclear material control. Specific technological solutions for MC&A measurements will be the subject of future work. Any potential pathway for nuclear material to leave the reactor containment (e.g., via a waste stream) was also considered and is discussed here.

5.1.1 Fuel fabrication

Carrier salt synthesis from its constituents is not performed at the reactor site. Instead, the carrier salt is manufactured off-site and transported to the reactor as the salt compound FLiBe. The carrier salt is combined with fissile uranium (in the form UF_4) on-site.

5.1.2 Fresh fuel component storage area

The fresh fuel component storage area is not pictured on the original LFTR process flow diagram, but it will be located outside the reactor vessel. FEI should consider where to locate any nuclear material storage areas.

5.1.3 Initial fuel loading

At initial loading, neither the fuel nor blanket salts in the reactor vessel contain fissile material; fissile material will be added through the chemical processing system during the approach to criticality. The FLiBe fuel salt (without nuclear material) is the only material present in the reactor vessel. However, the fresh blanket salt requires an initial loading of nuclear material (thorium). Fresh blanket salt is brought on-site, heated, melted, and then loaded into the decay tank.

During equilibrium operation, the thorium for the blanket salt arrives at the reactor site as thorium metal rods. This thorium is leached out of the rods within the electrolytic system and replaces uranium and protactinium atoms in the blanket with thorium atoms. Since the current through the electrolytic cell could be measured, the resultant signal could be correlated to the number of thorium atoms and thus the mass of thorium in the blanket. This would be a new application of MC&A measurements for the LFTR.

The decay tank is where the fuel is initially loaded into the reactor. Therefore, fissile nuclear material is present in the decay tank at startup (likely as UF_4). Then, the decay tank fluorinator can be used to convert the uranium to UF_6 and transfer that fissile material to the vessel after reconstitution (again as UF_4). For the purposes of this assessment, it is assumed that the LFTR is started up on ^{233}U from another LFTR (e.g., first generation LFTR). However, this is an area of active research and development and is being investigated by FEI.

For future planning for LFTR, it will be necessary to determine the specific startup fissile material, which is currently being explored. One option being explored by FEI is to start up the first LFTR on high-assay low-enriched uranium (HALEU) in the fuel salt up to 19.95% ^{235}U and then the rest (future generations) of LFTR will operate on ^{233}U. However, starting the LFTR on ^{238}U might be undesirable in terms of sustaining reactivity. This is not only an active area of research and development for FEI from a reactor operations standpoint, but it is also subject to US supply of HALEU and ^{233}U and dependent on whether that material could be made available for LFTR use in the future.

5.1.4 Nuclear material entry points

Entry points for the initial startup blanket and fuel fissile components are planned to be at the electrolytic cell. The electrolytic cell has a holdup tank (not shown in the process diagram) for salt before the inlet to the electrolytic cell, which provides a constant feed into the electrolytic cell. During normal operation, some quantity of both thorium and uranium will be present in multiple chemical forms within the electrolytic cell(s). The nuclear material mass present in the cell(s) will be dependent on design specifications for the salt processing.

If sampling is implemented for process monitoring during plant operations, access points may be provided at other parts in the plant (e.g., the decay tank). The next revision of the LFTR process flow diagram would include material entry and exit points for both operations and potentially sample lines for measurement, as well as nuclear material storage areas and waste streams.

Additionally, the correlation between reactor power and the SNM inventories should be explored. Since the reactor power should be directly correlated with the fissile material input, which in turn is dependent on the breeding of ^{233}U, a systematic investigation on how correlations of reactor power histories can inform the production and utilization of SNM, source material (thorium), and protactinium and the MC&A approach.

5.1.5 Nuclear material holdup

Since LFTR operates using liquid fuel salt, there is the potential for nuclear material accumulation (holdup) within any pipe or component, particularly at filters throughout the plant during normal operations. A nuclear material holdup monitoring approach should be developed accordingly. Additionally, if components are removed and replaced, the nuclear material hold up in that component must be documented.

5.1.6 Off-gas system

The purpose of the off-gas system is the treatment of volatile fission products. Once the design of the off-gas system has been finalized, the potential accumulation of nuclear material being captured in the filters and components and resulting in holdup will need to be evaluated. Further, a holdup measurement approach will need to be determined. It is possible that some UF_6 remains dissolved in the fuel salt that could lead to uranium in the off-gas system. It is unlikely that thorium or uranium will end up in the off-gas system, but it should be considered.

5.1.7 Waste stream

There are essentially two different waste streams, which are not shown on the LFTR process flow diagram: (1) fission products from the fuel salt cleanup will undergo further processing to convert them to the form that would be desirable for long-term storage and (2) the anodes from the electrolytic cell at the end of life (to be determined by FEI from chemistry research and development). Note that because waste stream 1 is the output of the process to remove fission products from the fuel salt, there is the potential for some fissile material to end up with the fission products in the waste stream. However, this would be minimized by FEI to prevent wasting potential fuel.

As part of planning for future MC&A measurements, further analysis is required to determine both (1) what can be measured from the waste stream and how to perform the measurement and (2) and what fissile material has the potential to end up in the waste stream (i.e., could potentially [inadvertently or otherwise] leave the reactor containment). For example, because the fuel fluorinator has process inefficiencies, the quantity of uranium in the waste stream should be evaluated. FEI has planned further research to gather experimental data to support this decision.

Future modeling efforts should also consider fission product accumulation in the blanket over time. Although only a small fraction of fissions occur in the blanket salt, accountancy measures should be in place to track any produced actinides. If the blanket salt ultimately goes to waste as a disposal strategy, then this could be a potential path to actinides leaving the reactor containment. In this regard, it is recommended to consider additional monitoring at points of potential exit to the reactor containment. Such monitoring points could function like a radiation portal monitor, which is designed to simply alarm if a radiation signature is detected, to indicate nuclear material exiting reactor containment.

5.1.8 Decay tank

There is ~5 kg of uranium and ~25 kg of protactinium present in the decay tank at any time, depending on the operating parameters and residence times. This is detailed in Section 4.1. It is possible that samples of the decay tank salt could be measured outside the decay tank for characterization and quantification. The composition of the entire decay tank is currently unknown, in addition to the specific decay tank geometry, which would both affect decay tank accountancy measurements.

5.1.9 Fuel salt drain tank and storage

The drain tank and storage are not shown on the LFTR process flow diagram. The drain tank is used whenever the fuel salt needs to be removed from the reactor core (i.e., during emergency shutdown or maintenance shutdowns/vessel transfers). During normal operations, most of the fuel salt continues through a cycle loop and does not go through the drain tank. Currently, FEI has no plan to drain the blanket salt. However, if the blanket salt could be removed to the drain tank, then this should be accounted for in future measurement planning. If the reactor was shut down quickly and the salts were removed from the vessel to the drain tank, then there would be a small amount of thorium conversion ultimately leading to uranium production.

5.2 DESCRIPTION OF PLANNED OPERATIONAL DOWNTIME

5.2.1 End of cycle (two reactor vessels)

At the end of a complete cycle, the reactor will be shut down to transfer the fuel from one reactor vessel to another (two reactor vessels). This is planned every 4 years. Therefore, one MC&A consideration is whether a physical inventory can be performed without shutting down the reactor. Such a physical

inventory could be performed from sampling followed by destructive analysis. Given that NDA might be challenging in some areas of LFTR and physical access for measurements is likely difficult, a measurement system should be included in the next version of the LFTR design.

5.2.2 Component maintenance

Because nuclear material may be held up within plant components, a monitoring plan according to FEI's maintenance plans and schedule needs to be considered.

5.3 POTENTIAL LFTR MATERIAL BALANCE AREA (MBA) STRUCTURE AND MEASUREMENT POINTS

The previous section detailed potential locations of nuclear material in the LFTR facility. This section discusses how to ensure that nuclear material is in the expected locations (i.e., accounting for the material). Material balance areas (MBAs) are required for MC&A. A LFTR would likely have one MBA for both MC&A and IAEA safeguards. For MC&A, additional internal control areas (ICAs, sometimes referred to as *item control areas*) would then be defined with measurement points at the input and output points to those ICAs. Five main ICAs should be considered for the 2018 LFTR design: (ICA 1) reactor vessel, (ICA 2) blanket salt processing/electrolytic cell (where thorium weight is the input), (ICA 3) decay tank (the tank could potentially be mounted on a scale (e.g., load cell) and monitoring system around the decay tank), (ICA 4) drain tank, and (ICA 5) the fuel salt processing system. The proposed ICAs are shown in Figure 11. Note that ICA 1 (the reactor vessel) may be better divided into a fuel salt side and blanket salt side because these streams do not mix within the reactor vessel. Furthermore, the storage areas including fresh fuel storage and waste storage areas would be a ICAs. Note that additional MC&A would need finer resolution on the ICAs to account for and localize any inventory differences.

Consideration needs to be given to the measurements that can be performed within LFTR—both from an operational and MC&A perspective—and where those measurements can occur. Operational measurements and MC&A measurements can be the same measurement system. In fact, the operator's MC&A system and associated measurements are required by the US NRC. For IAEA safeguards, independent verification is required, and generally inspector measurement systems are independent of the operational measurements. A paradigm shift toward joint use of the MSR operator's equipment for IAEA measurements will likely be necessary, which is outside the scope of this discussion.

For IAEA safeguards, nuclear material movements into and out of the LFTR facility would need to be declared to the IAEA. As part of 10 CFR Part 75, the licensee must complete a design information questionnaire that the IAEA uses to perform a design information verification to verify the design features of LFTR. This is of vital importance to the IAEA because many process operations within LFTR are likely not accessible after the start of operations and will be challenging to independently authenticate and directly verify.

One important design feature from an MC&A perspective is the decay tank. FEI will need to be able to determine the contents of the decay tank and will likely need to determine a method for measuring what enters and exits the decay tank, as well as the nuclear material flow rates. The decay tank could be positioned on a scale for the measurement of the total mass of material in the tank. A monitoring requirement would be to ensure that the decay tank weight stays consistent over time, and any deviation would indicate material removal due to a process loss elsewhere in the system or nuclear material removal from the tank itself. However, the decay tank is currently a large single tank. Where relying on a weight measurement for MC&A or verification purposes, a smaller volume in the decay tank could allow for more precise weight measurements. FEI should perform a system evaluation to evaluate how measurement uncertainty would be effected by a smaller decay tank, or series of smaller decay tanks,

which could make it easier to detect changes in the tank. Alternatively, the decay tank volume and decay salt residence time would be necessary to distinguish changes in the decay tank. Note, this is change detection rather than an absolute quantification. If absolute quantification is required, then a measurement method would need to be developed that could apply to the decay tank.

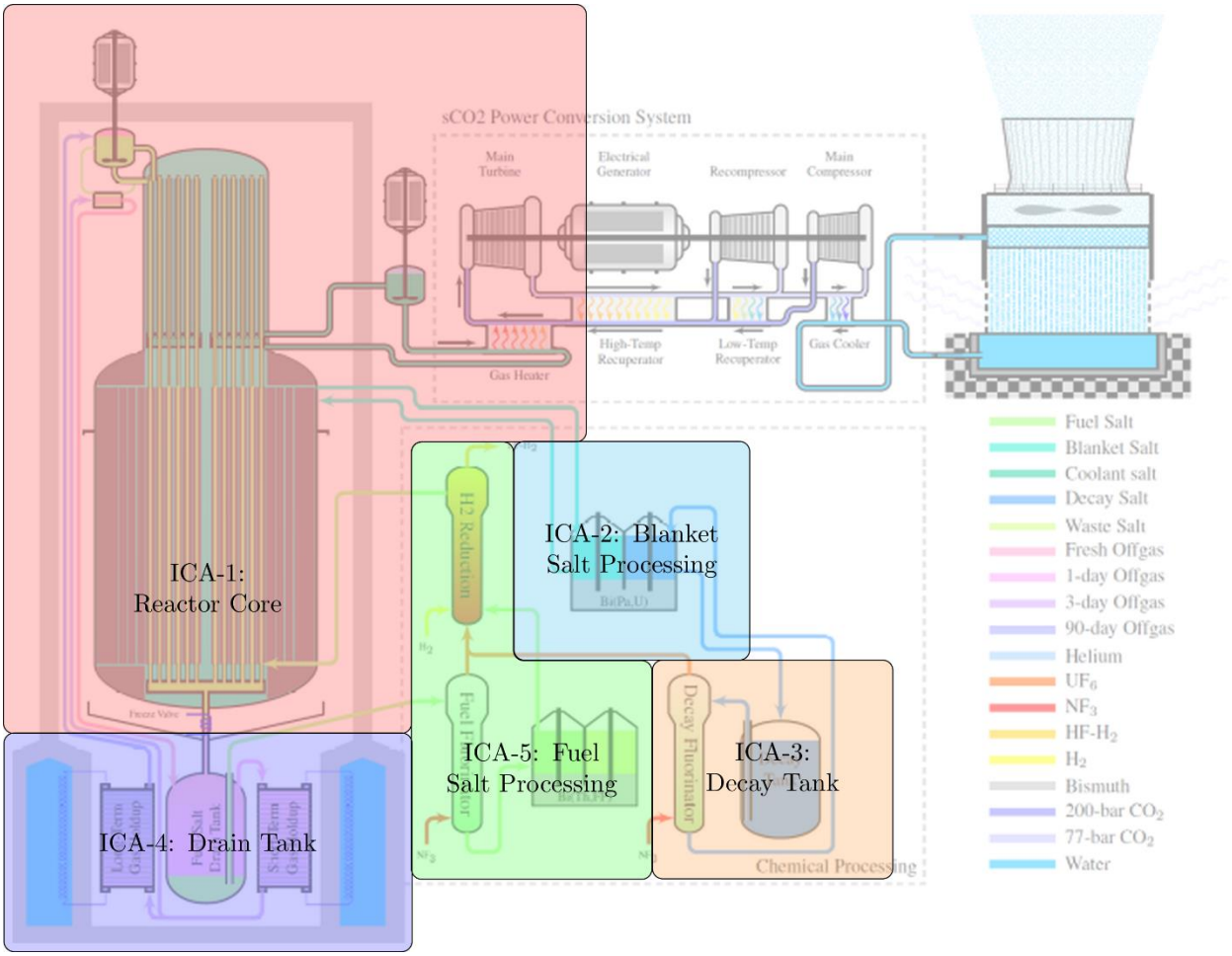

Figure 11. Proposed ICAs for the LFTR.

The drain tank could also be placed on a scale for the determination of the total mass of material. In both the case of the decay tank and drain tank, additional MC&A measurement technologies would need to be developed for the characterization (i.e., isotopic composition) and quantification (i.e., isotopic mass) of the nuclear material within those tanks. For example, these measurements might include gamma or neutron detection and monitoring around the perimeter of the tanks or other state-of-the-art monitoring techniques such as advanced Raman [16] or laser induced breakdown spectroscopy (LIBS) [17]. Additionally, the use of flow meters on the input and output pipe of the decay tank would provide another measurement to achieve material balance on the decay tank.

The LFTR chemical processing system is based on continuous processing. Establishing material balances for nuclear material accountancy is more challenging for continuous processing; it is recommended that process unit operations be revisited to consider batch processing instead. For example, places in the design where the nuclear material is purposely held up, such as the decay tank, could be treated as a batch process instead of a continual process. Furthermore, the electrochemical process is currently a continuous process that could be changed to a batch process. This move toward batch processing would be beneficial

for nuclear material accounting because batches of nuclear material are easier to measure. Continuous versus batch processing is discussed further in the next section as part of the recommendations about MC&A for the decay tank process.

6. CONCLUSIONS AND MC&A DESIGN RECOMMENDATIONS FOR LFTRS

FEI developed the LFTR to support environmental sustainability and to optimally use nuclear material (thorium). The LFTR design builds upon MSR technology that was conceived in the 1960s as a unique and innovative approach to obtaining energy from nuclear fission while maximizing operational flexibility, eliminating the need for the precise fabrication of solid fuel, and eventually eliminating the need for the enrichment of uranium and separate reprocessing facilities. However, these unique design benefits also pose unique challenges for the accounting of nuclear material. As such, the considerations of how to best achieve this material accounting in this early portion of the design stage of LFTR would be beneficial to FEI.

This section describes the major recommendations from this assessment, which are summarized in Section 6.1. Several specific recommendations are made regarding LFTR MC&A for FEI's consideration. Before performing the inventory calculations, the fuel salt was anticipated to contain the highest ^{233}U inventory and thus present the highest priority for MC&A. The uranium vector in the fuel salt, although still relatively high ^{233}U content, was found to contain lower ^{233}U content than suggested by historic studies, which is likely due to the improved accuracy of modern modeling tools. Once the ^{233}U is in the fuel salt system either inside or outside the vessel, its composition is relatively constant and SNM could potentially be tracked with reasonable accuracy. It is when blanket salt is being processed that many of the complexities in tracking SNM occur. Thus, many of the recommendations center on the blanket salt processing and decay tank. Note that the recommendations in this section do not represent an exhaustive list, and additional studies will be required to address MC&A challenges in more detail including where MC&A design recommendations require further investigation (e.g., operational versus safeguards trade-offs). Section 6.2 identifies clear areas for future study that build directly on this work.

6.1 MC&A DESIGN RECOMMENDATIONS FOR LFTRS

2018 LFTR Chemical Processing System Design Features and Potential Nuclear Material Control and Accountancy (MC&A) Challenges	Design Recommendations for improving MC&A within the LFTR Chemical Processing System
• A single large decay tank volume potentially leads to uncertainties in accounting measures (e.g., weighing precision and gamma self-shielding) and thus renders quantification of the decay tank nuclear material inventory challenging.	• Perform a systems design and explore the use of a series of smaller decay salt inventories while examining impacts to uncertainty in accountancy measurements for decay tank nuclear material inventory quantification (i.e., investigate tradeoffs between factors such as geometry, weighing precision, and self-shielding). • Design and build MC&A technologies into the decay process; for example, containment and surveillance technologies such as tamper indicating devices; or accountancy measurements using a custom nondestructive assay (NDA) system for the series of decay salt inventory(ies) potentially using both gamma-ray spectroscopy for isotopic composition analysis and neutron counting techniques for mass quantification. • Use the electrolytic cell in the 2018 LFTR chemical processing system design to validate other accountancy measures. Since thorium replacement at the electrolytic cell can be measured very precisely, it is possible to precisely quantify the mass of uranium and protactinium transferred to the decay tank.
• Continuous salt processing makes nuclear material inventory management and accounting challenging because it requires real-time accountancy of highly radioactive nuclear material in process, in a liquid form, at a high temperature, and in a highly corrosive environment.	• Convert to batch processing of the salt to enable additional static accounting measures (e.g., weight, volume). This may also help address the need for additional shielding and reduce the impact of high temperatures and corrosivity on instrumentation. • Design decay salt inventories in such a way that they become items (i.e., can be verified and then sealed to maintain continuity of knowledge). This would require the use of individually sealed containers of decay salt that can disconnect from the filling pipe. After several half-lives, the protactinium would be sufficiently decayed. Note that this option might be least favorable from a physical access standpoint.

2018 LFTR Chemical Processing System Design Features and Potential Nuclear Material Control and Accountancy (MC&A) Challenges	Design Recommendations for improving MC&A within the LFTR Chemical Processing System
• From a domestic safeguards standpoint, the decay tank inventory is not obviously self-protecting. Estimates of the decay tank inventory predicted that the ^{232}U content was 0.05 ppm with minimal fission products present, which are expected to be the dominant contributions to the radiation source term.	• Verify the expected self-protection of the decay inventory by expanding the model to explicitly account for decay salt composition and tank geometry. • Add fission products that have been separated from the fuel salt to increase the gamma activity within the decay tank and thus increase self-protection and deterrence to potential theft. Note that this may make measurement more difficult for both MC&A and IAEA independent verification, and trade-offs will need to be investigated. • Combine the decay inventory(ies) with a portion of the (clean or irradiated) fuel salt to reduce the purity of the uranium vector of the decay inventory. • Increase ^{232}U content by modified reactor operations, blanket composition, or both, such as by the addition of ^{230}Th [8] to improve self-protection.
• Approximately 3–8 kg of fissile ^{233}U and 27–28 kg Pa (~90% ^{233}Pa, ~10% ^{231}Pa) reside in the decay tank continuously with potential access, depending on the radiation source term.	• Rework the blanket salt processing to remove a dedicated decay salt inventory. For example, send blanket salt directly to the decay fluorinator (i.e., do not remove protactinium from the blanket salt). Note, this change is invasive and incompatible with the above suggestions. In addition, this will significantly reduce the thorium conversion efficiency by not protecting the protactinium from further neutron capture. Additionally, it may significantly alter the neutronics of the reactor.
• The uranium vector in the UF_6 output from the decay salt fluorinator was estimated to be 99.98% ^{233}U and 0.02% ^{234}U, with 0.05 ppm ^{232}U.	• Eliminate the decay fluorinator shown in Figure 2 in Section 1.2.2 entirely by allowing protactinium to decay in a small inventory of fuel salt outside the reactor vessel. • Reduce purity of the uranium vector from the decay fluorinator for example by routing the output of the fuel fluorinator to the input of the decay fluorinator. • Remove physical access to the UF_6 stream. For example, ensure the pipe between the fluorinators is too short for access.

*Note some recommendations are mutually exclusive.

As presented briefly in the preceding table, five potential options were identified for the decay tank system to increase the effectiveness of nuclear material accountancy and facilitate potential future IAEA safeguards verification activities: (1) maintain the baseline of a large decay tank using a continuous process; (2) convert the decay tank to a series of smaller decay salt inventories that also use a continuous process; (3) convert the decay tank to a series of smaller decay salt inventories using a batch process; (4)

convert the decay tank to a series of smaller decay salt inventories using a batch process that could be designed to become sealed items; and (5) eliminate the decay tank. These options are illustrated in the schematic shown in Figure 1. The different options have relative advantages and disadvantages, which are discussed in more detail in the following subsections.

Depending on the selected option, differing MC&A measurement technologies may be designed and built into the decay tank process. For example, a custom NDA system could be designed and developed for the series of decay tanks using both gamma-ray spectroscopy for isotopic composition analysis and neutron counting techniques (i.e., neutron singles counting and correlated neutron counting including neutron coincidence counting) for mass quantification.

6.1.1 Single large decay tank to series of smaller decay tanks

The LFTR design considered in this work made use of a single decay tank as part of the continuous blanket salt processing. As stated in Section 4.1, the decay tank holds approximately 5 kg of uranium and 28 kg of protactinium depending on the residence time. Although not determined for this work, the decay tank would likely also contain some amount of other material such as the carrier salt FLiBe. A relatively large inventory makes it more difficult to quantify small fluctuations in the nuclear material inventory by increasing the uncertainty in accounting measures such as mass measurements or gamma-ray spectroscopy.

To enable easier nuclear material quantification, the large decay tank should be replaced with a series of smaller decay tanks (option 2, Figure 1). Each of the tanks would have a reduced inventory, which should reduce the measurement uncertainty. Furthermore, the process could be improved by designing a custom NDA system to build verification measurements directly into the decay tanks or decay tank configuration.

Replacing one larger decay tank with several smaller ones would increase the length of piping and number of flow loops, increasing the potential access points for diversion. However, because this equipment will be in a high radiation area during reactor operations, access will be limited, and as such, this will be a minor concern.

6.1.2 Continuous to batch blanket salt processing

As designed, the blanket salt processing is designed to operate as a continuous chemical process. As material is flowing in a bulk state, a continuous process would require real-time accountancy of the highly radioactive liquid containing nuclear material. Similar real-time accounting is performed in other fuel cycle facilities such as reprocessing or enrichment, and analogies may be drawn to derive MC&A best practices. Enrichment processes cannot be shut down, but the input and output can be closely monitored and controlled, and the material is not highly radioactive. Reprocessing facilities process highly radioactive material in batches and thus can perform accounting when the material is “static” (i.e., not flowing or “dynamic”).

If blanket salt processing were redesigned as a batch process (option 3, Figure 1), the nuclear material could be contained in containers or tanks where the volume and mass could be controlled. In theory, this would make accountancy simpler. A batch of irradiated blanket salt could be processed to extract the protactinium and uranium, which would then be placed into a decay tank. This tank could be verified and then sealed for a specified amount of time to enable the protactinium to decay before being used to add to the fuel salt (option 4, Figure 1). Tools such as NDA, density, volume, full height, and weight measurements could be used to verify the contents of the decay tank.

6.1.3 Increased self-protection of the decay tank

The standard for self-protection is a dose rate of greater than 100 rem/h at a distance of 1 m, and it is unlikely that the decay tank as designed would exceed this dose rate. The uranium vector in the decay tank contains a minimal amount of ^{232}U, which can significantly increase the dose rate due to highly active decay progeny such as ^{208}Tl. Furthermore, because most of the nuclear fissions occur in the fuel salt, minimal radioactive fission products reside in the decay tank.

Several methods are available to increase the activity of the decay tank, which would raise the tank above the self-protection threshold, relevant to physical protection. The first is to add fission products that have been separated from the fuel salt, which would greatly increase the activity of the decay tank. Ideally these fission products would remain in the decay salt during processing by the decay fluorinator, which removes uranium to be sent to the fuel salt. A downside of adding highly active fission products is the increased uncertainty in NDA methods and increased measurement difficulty in general. These trade-offs would need to be evaluated further to determine if adding any or all of the fission products are helpful for MC&A quantification or IAEA inspections. Although the IAEA does not consider self-protection in its safeguards approaches, any increase in the decay tank ^{232}U or fission product content could hinder access by IAEA inspectors to perform any kind of measurement. Therefore, it would be necessary to investigate these trade-offs for the LFTR decay tank radiation source term.

Another possible pathway toward increasing the activity of the decay tank is to increase the concentration of ^{232}U in the blanket salt and the corresponding concentration in the decay salt. One method for this is to add a small fraction of ^{230}Th to the blanket salt. This isotope can be found in thorium that has been codeposited with uranium, and ^{230}Th is readily converted into ^{232}U by neutron irradiation.

Since the uranium vector in the decay tank is about 99.98% ^{233}U as discussed in Table 5 in Section 4.1, another recommendation is to direct a fraction of the fuel salt into the decay tank. Because the fuel salt uranium vector contains around 56% ^{233}U, this should significantly reduce the ^{233}U purity in the decay tank.

6.1.4 Decay tank removal

As the previous sections describe, there are several challenges associated with the present decay tank design ranging from several significant quantities of continuous nuclear material inventory to measurement and verification uncertainties. Instead of addressing these challenges, another approach is to redesign the blanket salt processing to entirely remove the decay tank and the associated decay salt loop (option 5, Figure 1). In theory, this is equivalent to not removing the protactinium from the blanket salt and instead using the current decay fluorinator to remove uranium from the blanket salt. This has the downside of no longer protecting the protactinium from neutron irradiation, which reduces the thorium consumption efficiency and may increase the actinides present in the blanket salt by increasing the amount of ^{233}U in the blanket salt. In other words, this option is likely counter to the design goals stated by FEI but has been added here for completeness.

6.1.5 Limit access to decay salt fluorinator output

As suggested, the decay tank contains nearly isotopically pure ^{233}U, which means that the outlet stream from the decay salt fluorinator will contain the same purity albeit in the form of UF_6. Physical access to this output should be limited such as by shortening the pipe connecting the decay fluorinator and the fuel salt reconstitution process. Alternatively, it may be possible to reduce the purity of the decay fluorinator output by operating the two fluorinators in series (fuel salt then decay salt) instead of in parallel, which would make the outlet stream more closely match the fuel salt uranium vector.

6.1.6 Blanket salt composition

FEI is considering modifying the blanket salt composition from the historic MSBR design by ORNL. During this work, several potential compositions were modeled to provide reactor physics input to FEI as well as for this assessment. Options included removing beryllium and exchanging beryllium for sodium. These design variations were found to have minimal influence on the equilibrium nuclear material isotopic vectors throughout the system. Note, the blanket salt material (particularly sodium) was found to affect the system neutronics such as criticality, so potential compositions warrant further study to determine optimal makeup.

6.2 FUTURE WORK

This work was intended as a preliminary exploration into nuclear material control and accountancy to support future design considerations for LFTR. This section is devoted to outlining a few of the possibilities for building upon the work in this report.

6.2.1 Expanding LFTR Safeguards Modeling

As stated in Section 11, the design for LFTR was in flux for much of this study. Thus, the 1967 two-fluid MSBR design by ORNL was chosen as a reference in order to develop general MC&A conclusions regarding LFTR, which was based on the that MSBR. In order to improve the accuracy of the findings in this report, an accurate model of LFTR should be developed and used for analysis. Such a model should account for the heat exchanger modifications at the top of the reactor vessel as well as more accurate salt processing chemistry. Note that future iterations of this analysis using a model more closely resembling the LFTR design will likely result in small changes to the output and thus the conclusions made from those results.

In addition, this work assumed equilibrium conditions for LFTR, which is representative of operating a LFTR that was fueled by another LFTR. To better represent MC&A challenges, future study should be devoted to model transient conditions such as startup and shutdown as well as the operation using non-LFTR fuel to initialize the reactor. Such a study could also explore transient conditions such as emergency shutdown or end-of-life conditions. These studies are not trivial as they depend on operational parameters such as reactor lifetime and startup fuel/salt composition.

6.2.2 Design of MC&A Measurement Technologies for LFTR

As discussed in Section 5, MC&A measurement technologies will need to be developed for LFTR. These might include NDA systems based on radiation signatures, destructive assay techniques, or the use of other signatures.

One important step will be for FEI to determine what operational and process control measurements they plan to put in place including components such as sample lines and sampling ports in the process flow sheet.

6.2.3 Model MC&A Plans for Designs of this Type

The application of domestic safeguards at US domestic nuclear facilities includes MC&A for SNM. The current MC&A regulatory framework and published guidance, including for advanced non-light water reactor nuclear energy systems, are not necessarily directly applicable to all MSR nuclear energy system designs. Therefore, MC&A system design assessments, as well as identification of MC&A program features and required performance measures, are recommended to provide input to both the design

licensing process and planning for future MC&A regulations and regulatory guidance for these reactor types.

This work provides preliminary input to a future detailed MC&A design assessment of the LFTR. Such a future assessment should provide an independent, authoritative examination to provide a model MC&A plan for MSRs based on current US Nuclear Regulatory Commission (NRC) licensing requirements under Title 10 of the *Code of Federal Regulations* (CFR) [1] Part 74 "Material control and accounting of special nuclear material." In the future, ORNL could provide a technical report on a model MC&A plan for LFTR to FEI, which would provide input to a future NRC Regulatory Report that will be prepared by FEI.

The Draft US Nuclear Regulatory Commission (NRC) report NUREG-2159, "Acceptable Standard Format and Content for the Material Control and Accounting Plan Required for Special Nuclear Material of Moderate Strategic Significance," [2] may provide useful information regarding the future regulatory basis for an MC&A Plan for LFTR. The NRC currently does not have guidance for moving fuel reactors (i.e., reactors with continuous online refueling) nor does it have guidance for reactors that use fresh uranium fuel that is enriched between 5% and 20% in ^{235}U (i.e., high-assay low-enriched uranium or HALEU). Since the current fleet of LWRs under NRC regulations use low-enriched uranium (LEU) fuel <5% ^{235}U, extrapolating future MC&A regulations and guidance will be necessary for application to liquid fueled reactors using alternative fuels (e.g., HALEU, Th/^{233}U).

Future work will require the formulation of a new approach or framework for MC&A system design considerations that may be based on a combination of guidelines for "bulk-type" facilities, such as enrichment or reprocessing plants, as a model for a continuously operating process. An applicable reference facility might also be a pebble bed reactor since it has continuous online refueling.

6.2.4 IAEA Safeguards, Security, and Proliferation-Resistance Engagements

The outcomes of this preliminary assessment serve as the foundation for MC&A system design that could be useful in future IAEA safeguards-by-design assessment for FEI. 10 CFR Part 75 "Safeguards on nuclear material-implementation of safeguards agreements between the United States and the International Atomic Energy Agency" states that US nuclear facilities that are placed on the eligible facilities list may be selected for the application of IAEA safeguards. Thus, LFTRs may still have to comply with some aspects of IAEA safeguards even if built in the United States.

The US government has various offices that can assist FEI with the consideration and potential implementation of IAEA safeguards, security, and proliferation-resistance evaluations. FEI should take advantage of this assistance to further inform its design and to incorporate these topics.

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