

DEVELOPMENT OF NUCLEAR SAFEGUARDS APPROACHES FOR A MOLTEN
SALT REACTOR DESIGN

A Thesis

by

ANNA GENEVIEVE ARMSTRONG

Submitted to the Office of Graduate and Professional Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

Chair of Committee,	Sunil Chirayath
Committee Members,	Craig Marianno Matthew Fuhrmann
Head of Department,	Michael Nastasi

May 2021

Major Subject: Nuclear Engineering

Copyright 2021 Anna Genevieve Armstrong

ABSTRACT

The original molten salt reactor (MSR) concepts were developed at Oak Ridge National Laboratory (ORNL) in the 1960s. Increasing energy demands and concerns about global CO₂ emissions have emphasized the need for advanced nuclear energy systems, such as MSRs. The inclusion of MSRs in the Generation IV reactor designs bolstered global interest in the development of MSR technologies beyond the Nuclear Weapons States (NWSs). This has created proliferation concerns relating to MSRs, especially as they can be considered small modular reactors (SMRs), which makes them an economically attractive entry into nuclear reactor development for Non-Nuclear Weapons States (NNWS) and potentially for States that are looking to acquire nuclear material for illicit weapons programs. However, MSRs present unique challenges for the application of nuclear safeguards, and there are currently no existing technical solutions. One potential technology is hybrid K-edge densitometry (HKED), which could be used to determine isotopic concentrations at strategic points in the system to support implementation of material balance areas (MBAs) that are required to determine if any special nuclear material (SNM) is unaccounted for. The value of material-unaccounted-for (MUF) must be lower than one significant quantity (8 kg for plutonium, 75 kg of ²³⁵U for low enriched uranium, 25 kg of ²³⁵U for high enriched uranium) in a specified time frame, known as a material balance period (MBP). This regulation is to ensure that the MSR can meet international safeguards requirements as applied by the International Atomic Energy Agency (IAEA). Therefore, to simulate SNM accountancy measurements results from a

reference MSR design, a uranium-based reactor core design was developed as part of this research. The reactor was designed to operate in the thermal neutron spectrum with a power output of 300 MW_{th}. The reactor core consisted of a hexagonal design with 61 fuel-salt channels and 108 blanket-salt channels, each connected to salt plena on the top and bottom of the core. The core was surrounded by a graphite radial reflector and a boron carbide (B₄C) neutron absorber, which are enclosed in a Hastelloy-N reactor vessel. The fuel-salt was composed of uranium tetrafluoride (UF₄) with ²³⁵U enrichment of 3.5%, constituting 20% of the fuel-salt by core weight fraction, and mixed with molten lithium and beryllium fluoride (FLiBe) at a temperature of 900 K. The fuel-salt channels were surrounded by depleted UF₄ blanket-salt channels. The fuel-salt and blanket-salt both had the same density (2.892 g/cm³). This proposed reactor model was developed using Monte Carlo N-Particle Transport (MCNP version 6.2) code and the Standardized Computer Analyses for Licensing Evaluation (SCALE version 6.2) code system. These models were used to compare effective neutron multiplication factor values under steady-state operating conditions. Fuel burnup simulations were performed using the MCNP model only. Based on the results of SNM production and fuel-salt flow rates, MSR safeguards approaches were developed.

Results of the MCNP fuel burnup simulations were used to determine SNM quantities over time in each of the three designated MBAs for the MSR by utilizing HKED as an SNM assay methodology at the KMPs. A combined (systematic and random) measurement uncertainty of 0.28% provided for HKED by the IAEA ITV document was assumed. When considering the combined three loops of the MSR, the IAEA conditions

($MUF < SQ$, $MUF < 3\sigma_{MUF}$, and $3\sigma_{MUF} < SQ$) were met for both ^{235}U and plutonium for the MBP of 12 months. Further analysis was done considering a multi-module site with four MSRs. This analysis showed that the $3\sigma_{MUF} < SQ$ condition would not be satisfied for either ^{235}U or plutonium for the 12-month MBP.

This work demonstrates that both MCNP and SCALE can be used to model an MSR in developing NMA strategies for a molten salt reactor core design. Results of the fuel-salt depletion were used to determine that the proposed MBPs of one month and three months meet the three IAEA safeguards compliance conditions ($MUF < SQ$, $MUF < 3\sigma_{MUF}$, and $3\sigma_{MUF} < SQ$) for both ^{235}U and plutonium.

DEDICATION

This thesis is dedicated to my parents.

ACKNOWLEDGEMENTS

I would like to thank my committee chair, Dr. Chirayath, and my committee members, Dr. Marianno and Dr. Fuhrmann, for their support.

I would also like to thank Donald Kovacic, Michael Whitaker, Jessica White-Horton, and Logan Scott for welcoming me to the International Safeguards Group at ORNL over the course of my thesis research. Thank you to Dr. Betzler of ORNL who generously aided me in learning SCALE, which was integral to my project. Thanks also go to Robert McElroy, Jr. who gave me a tour of the HKED installed at ORNL.

CONTRIBUTORS AND FUNDING SOURCES

Contributors

This work was supervised by a thesis committee consisting of Dr. Chirayath and Dr. Marianno of the Department Nuclear Engineering and Dr. Fuhrmann of the Department of Political Science.

Assistance in MCNP modeling and design of the reactor core was provided by Dr. Chirayath.

All other work conducted for the thesis was completed by the student independently.

Funding Sources

Graduate study was supported by a graduate research assistantship from Texas A&M University.

This work was also made possible in part by the United States Department of Energy's National Nuclear Security Administration (NNSA). Its contents are solely the responsibility of the author and do not necessarily represent the official views of the NNSA.

NOMENCLATURE

CANDU	Canada Deuterium Uranium
CoK	continuity of knowledge
C/S	containment and surveillance
CSA	Comprehensive Safeguards Agreement
CVD	Cherenkov viewing device
DA	destructive analysis
EURATOM	European Atomic Energy Community
FLiBe	lithium and beryllium fluoride
HEU	high enriched uranium
HKED	hybrid K-edge densitometry
IAEA	International Atomic Energy Agency
IMSBR	Indian molten salt breeder reactor
INFCIRC	Information Circular
ITV	international target value
KED	K-edge densitometry
KMP	key measurement point
LANL	Los Alamos National Laboratory
LEU	low enriched uranium
LWR	light water reactor
MBA	material balance area

MBP	material balance period
MC&A	material control and accountancy
MCNP	Monte Carlo N-Particle
MOX	mixed oxide
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
MUF	material unaccounted for
NDA	non-destructive assay
NMA	nuclear material accountancy
NNSA	National Nuclear Security Administration
NNWS	Non-Nuclear Weapons State
NPT	Treaty on the Non-Proliferation of Nuclear Weapons
NRTA	near-real-time accountancy
NWS	Nuclear Weapons State
OSL	on-site laboratory
ORNL	Oak Ridge National Laboratory
REDC	Radiochemical Engineering Development Complex
SBD	safeguards-by-design
SCALE	Standardized Computer Analyses for Licensing Evaluation
SMR	small modular reactor
SNM	special nuclear material
SQ	significant quantity

SSAC	State System of Accounting for and Control of SNM
UV	ultraviolet
XRF	X-ray fluorescence

TABLE OF CONTENTS

	Page
ABSTRACT	ii
DEDICATION	v
ACKNOWLEDGEMENTS	vi
CONTRIBUTORS AND FUNDING SOURCES.....	vii
NOMENCLATURE.....	viii
TABLE OF CONTENTS	xi
LIST OF FIGURES.....	xiii
LIST OF TABLES	xiv
1. INTRODUCTION.....	1
1.1. Objective	8
1.2. Overview of MSR technology.....	8
1.3. International development of MSRs	10
1.4. Proliferation concerns relating to MSRs	10
1.5. International nuclear safeguards.....	11
1.5.1. Motivations for safeguards.....	11
1.5.2. IAEA safeguards goals.....	12
1.5.3. Need for safeguards technologies.....	12
1.5.4. Challenges facing safeguards	15
1.6. Literature review	17
1.7. Why is this study different?	20
2. METHODOLOGY	21
2.1. Theoretical molten salt reactor core.....	21
2.2. Reactor physics (fuel burnup neutronics) simulations	24
2.2.1. MSR Fuel Burnup Simulations using SCALE	25
2.2.2. MSR fuel burnup simulations using MCNP.....	27
2.3. SNM quantification and NMA analysis for nuclear safeguards	28
2.3.1. MBAs and KMPs	28

- 2.3.2. Reprocessing cycle of blanket loops31
- 2.3.3. Analysis of holdup accumulation and waste31
- 2.3.4. Consideration of multi-module MSR facility.....31
- 2.4. Hybrid K-edge densitometer for MSR safeguards applications.....31
 - 2.4.1. Hybrid K-edge densitometry31
 - 2.4.1.1. K-edge subsystem.....33
 - 2.4.1.2. X-ray fluorescence subsystem.....34
 - 2.4.1.3. Hybrid K-edge technique34
 - 2.4.1.4. Hybrid K-edge accuracy.....34
 - 2.4.2. Hybrid K-edge densitometry applicability to MSRs.....35
 - 2.4.3. NMA analysis using HKED35
 - 2.4.4. Suitability of the detection technique36
- 3. RESULTS.....37
 - 3.1. MCNP and SCALE-based MSR core fuel burnup simulation results37
 - 3.2. MBA inventory and nuclear material accountancy calculations.....37
 - 3.2.1. Core reprocessing loop37
 - 3.2.2. Blanket reprocessing loop39
 - 3.2.3. Analysis of holdup and waste.....40
 - 3.2.4. Analysis of SNM in all three MSR loops.....41
 - 3.2.5. MBA calculations for a multi-module MSR site.....45
- 4. CONCLUSIONS47
 - 4.1. Conclusions47
 - 4.2. Significance of conclusions.....49
 - 4.3. Future work49
- REFERENCES51
- APPENDIX55

LIST OF FIGURES

	Page
Figure 1. The 1960s-era MSR design developed by Oak Ridge National Laboratory [2].....	1
Figure 2. A Generation IV MSR design schematic depicting separate flows of fuel and coolant (blanket) salts [4].....	2
Figure 3. A fork detector produced by ANTECH, used by EURATOM to measure spent fuel assemblies [12].....	6
Figure 4. Schematic for the loop-type Indian molten salt breeder reactor (IMSBR) [20].....	14
Figure 5. (left) top view, and (right) side view of the MSR core model using SCALE. Legend: brown (air), green shades are surrounding materials from outermost position (Hastelloy-N, boron carbide, graphite), blue (graphite), grey hexagon with green core (graphite with blanket-salt channel), navy blue hexagon with cyan core (graphite with fuel-salt channel).....	24
Figure 6. (left) top view, and (right) side view of the MSR core model using MCNP. Legend: green shades are surrounding materials from outermost position (Hastelloy-N, boron carbide), light blue (graphite), light blue hexagon with yellow core (graphite with blanket-salt channel), light blue hexagon with dark blue core (graphite with fuel-salt channel), orange (fuel-salt plena), red (blanket-salt plena).	25
Figure 7. Effective neutron multiplication factor (k-eff) and fuel-salt burnup values vs. time for the MSR depletion (one fuel cycle) using SCALE.	26
Figure 8. Effective neutron multiplication factor (k-eff) and fuel-salt burnup values vs. time for the first fuel cycle using MCNP.	28
Figure 9. Proposed MBA scheme for the MSR under study.....	29
Figure 10. Plutonium production as a function of burnup.	30
Figure 11. Schematic of HKED system combining KED and XRF subsystems [34].....	32
Figure 12. HKED system available from Mirion Technologies [35].....	33

LIST OF TABLES

	Page
Table 1. ^{235}U MUF calculations for one loop of the reprocessing MBA for fuel cycle 1.	38
Table 2. Plutonium MUF calculations for one loop of the reprocessing MBA for fuel cycle 1.	38
Table 3. ^{235}U MBA calculations for one loop of the reprocessing cycle of the blanket loop.	39
Table 4. Plutonium MBA calculations for one loop of the reprocessing cycle of the blanket loop.	39
Table 5. ^{235}U buildup in holdup and waste.	40
Table 6. Plutonium buildup in holdup and waste.	41
Table 7. ^{235}U fuel-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.	42
Table 8. Summary of ^{235}U fuel-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	42
Table 9. Pu fuel-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.	43
Table 10. Summary of Pu fuel-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	43
Table 11. Summary of ^{235}U blanket-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	43
Table 12. Summary of Pu blanket-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	44
Table 13. Summary of ^{235}U holdup and waste NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	44
Table 14. Summary of Pu holdup and waste NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.	45

Table 15. Summary of ^{235}U fuel-salt reprocessing NMA calculations when considering four MSR's at one site.	45
Table 16. Summary of Pu fuel-salt reprocessing NMA calculations when considering four MSR's at one site.	45
Table 17. Summary of ^{235}U holdup and waste NMA calculations when considering four MSR's at one site.	46
Table 18. Summary of Pu holdup and waste NMA calculations when considering four MSR's at one site.	46
Table 19. ^{235}U blanket-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.	55
Table 20. Pu blanket-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.	55
Table 21. ^{235}U fuel-salt MUF calculations for holdup and waste for all three loops of the reprocessing MBA for fuel cycle 1.	56
Table 22. Pu fuel-salt MUF calculations for holdup and waste for all three loops of the reprocessing MBA for fuel cycle 1.	56
Table 23. Summary of ^{235}U blanket-salt reprocessing NMA calculations when considering four MSR's at one site.	56
Table 24. Summary of Pu blanket-salt reprocessing NMA calculations when considering four MSR's at one site.	57

1. INTRODUCTION

The development of the molten salt reactor (MSR) began in the late 1940s at Oak Ridge National Laboratory (ORNL) with the goal of generating low-cost power and extending the fissionable fuel resources of the United States [1]. This MSR design used a molten fuel of UF_4 and ThF_4 mixed with beryllium and lithium fluorides; the fuel circulated in a core surrounded by a graphite moderator, as seen in Figure 1. After 20 years of development of MSR technology, a core design with an output power of 8 MW_{th} , namely, the Molten Salt Reactor Experiment (MSRE), was successfully operated, demonstrating the practicality and stability of MSR operations [2].

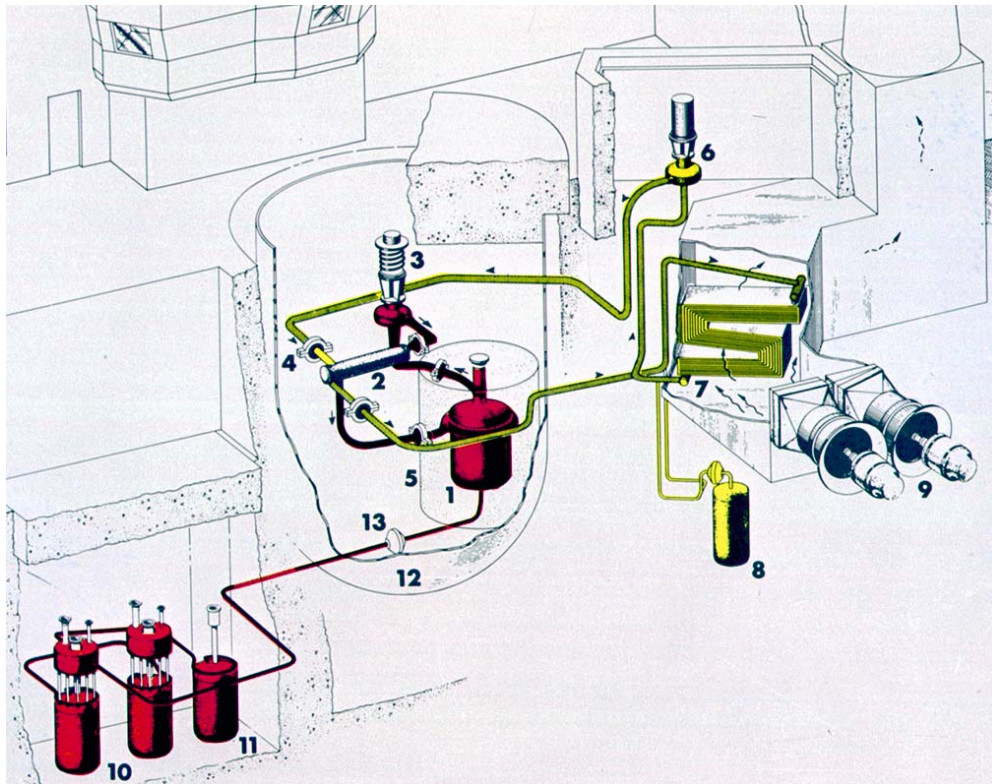


Figure 1. The 1960s-era MSR design developed by Oak Ridge National Laboratory [2].

Increasing energy demands and concerns about global CO₂ emissions have emphasized the need for such advanced nuclear reactors. Interest in MSR was reinvigorated upon their inclusion in the Generation IV reactor types [3]. A schematic depicting a Gen IV MSR design can be seen in Figure 2 [4]. As MSR technology continues to mature, a methodology for implementing special nuclear material control and accountancy (MC&A), but most importantly, nuclear material accountancy (NMA) methods, for an MSR must be developed.

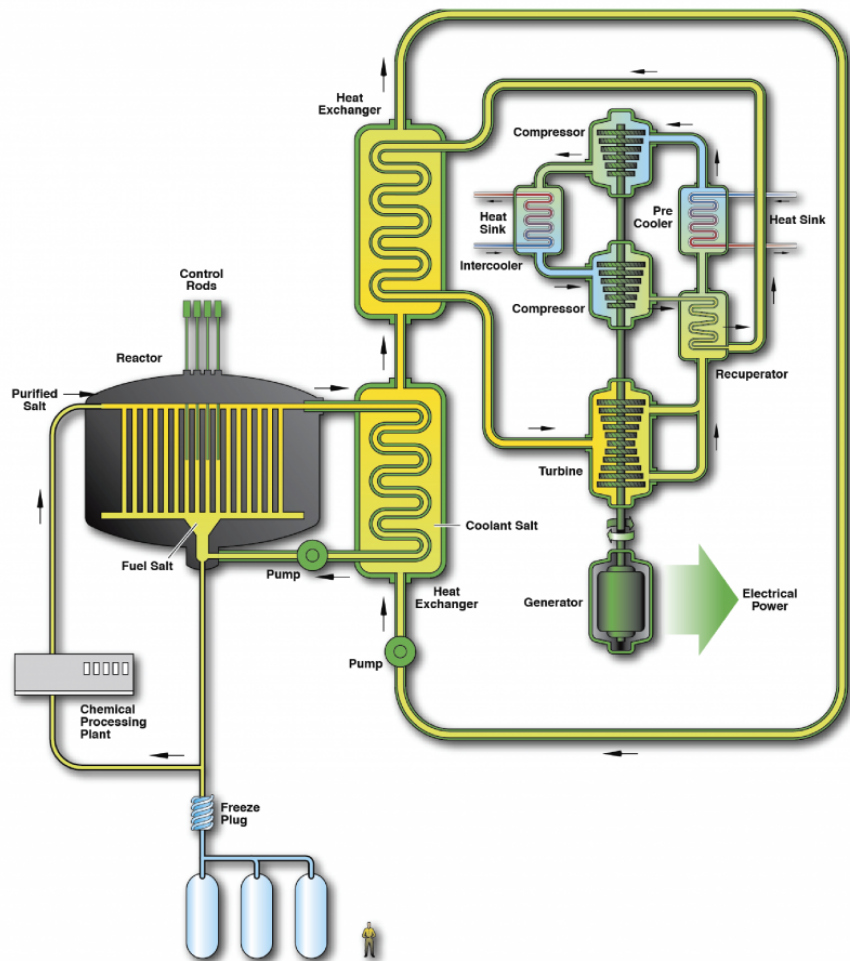


Figure 2. A Generation IV MSR design schematic depicting separate flows of fuel and coolant (blanket) salts [4].

Nuclear safeguards measures contribute to global non-proliferation objectives and are guided by the International Atomic Energy Agency (IAEA), in accordance with the Treaty on the Non-Proliferation of Nuclear Weapons (NPT), under the IAEA's Comprehensive Safeguards Agreement (CSA), Information Circular (INFCIRC)/153 [7] [8]. The nuclear safeguards framework was enhanced by the IAEA Additional Protocol (AP), given in INFCIRC/540 [9]. INFCIRC/153 and INFCIRC/540 ensure correctness and completeness of declarations of special nuclear material (SNM) and nuclear facility operations, respectively.

The IAEA safeguards systems has two objectives: to ensure that SNM is not diverted from its peaceful uses to military purposes; and to ensure that no undeclared nuclear activity is occurring in a State. MC&A is the responsibility of each facility and the State System of Accounting for and Control of SNM (SSAC), specifically for those states who have signed the INFCIRC/153 and potentially INFCIRC/540 [10]. MC&A works in tandem with containment and surveillance (C/S) to verify the correctness of nuclear accountancy as well as to provide continuity of knowledge (CoK) between IAEA inspections. The IAEA definition of a significant quantity (SQ) is the amount of material for which the possibility of manufacturing a nuclear explosive cannot be excluded [10]. The SQ values for low enriched uranium (LEU) and plutonium are 75 kg of ^{235}U and 8 kg, respectively.

Liquid-fueled MSR are distinct from fixed-fuel assembly light water reactors (LWRs). Thus, MSR, particularly those that are liquid-fueled, present unique challenges with respect to nuclear safeguards. Liquid-fueled MSR are tightly coupled systems that

combine the reactor core and fuel cycle (such as the processing of salt and separation of materials) into one facility. Liquid-fueled MSR designs differ from fixed-fuel assembly LWRs in that MSRs utilize online processing, whereby it is possible to remove a portion of the fuel material during reactor operation. Further complicating MSR safeguards is the fact that some designs have unique refueling schemes in which the core might be continuously fed fresh material [5]. Given the uniqueness of liquid-fueled MSR designs, it is also possible for fuel to be present outside the reactor core vessel, which is a further challenge with respect to safeguards.

Liquid-fueled MSRs, and MSRs in general, cannot follow the same approach as fixed-fuel assembly LWRs for IAEA safeguards. Current IAEA safeguards approaches for nuclear reactors focus on item counting, which is only applicable to fixed-fuel assembly reactors. IAEA inspections for bulk material accountancy have focused on the front-end and back-end of the fuel cycle; techniques and instrumentation for bulk NMA have focused on enrichment, fuel fabrication, and aqueous reprocessing, which cannot be applied to liquid-fueled MSRs without further development [5]. The use of liquid fuel means that material diversion could occur in potentially a much easier way than in a fixed-fuel assembly reactor. Liquid-fueled reactors lack discrete fuel elements, which when “combined with continuous transmutation and online processing,” makes traditional item-type NMA methods ineffective [6]. Processing that is done to the liquid fuel-salt mixture in an MSR further creates a pathway for material access. Given that potentially very large volumes of material are present in an MSR core at any one time due to the high material throughput, current safeguards methods must be altered before an MSR can be

successfully safeguarded from material diversion [6]. In order to properly safeguard a liquid-fueled MSR, a safeguards approach must be identified in which nuclear material will be accounted for at each stage of operation, with material balance areas and a detection method at key measurement points to accurately measure SNM.

An MSR is considered as a bulk SNM handling facility as compared to light water or heavy water reactors, which are considered as item facilities as they have fixed fuel assemblies in the core. Safeguards NMA for fixed-fuel reactors rely upon nondestructive analysis techniques. NDA techniques measure SNM content of an item without physically or chemically causing changes to the item. NDA is the most commonly used method for material accountancy. NDA methods can be either passive (no external source present) or active (neutrons or gammas irradiate the source to magnify the signal) [11]. NDA measurements typically measure an item's radiation emission; these data are then compared to a calibration which has been determined through destructive analysis techniques. There are various NDA instruments used to safeguard fixed-fuel reactors, to include: the digital Cherenkov viewing device (CVD) and the fork detector.

The fork detector is widely used as an NDA technique by the IAEA and EURATOM; the EURATOM fork detector can be seen in Figure 3 [12]. The detector system of the fork detector straddles an LWR fuel assembly with a combination of fission chambers (to measure neutrons) and ion chambers (to measure gammas). Fork detectors give information related to cooling time and burnup of fuel assemblies. While fork detectors are easy to use, they may miss fuel pin removal under 50%. Cherenkov viewing devices are sensitive to ultraviolet (UV) radiation in the water surrounding spent fuel. The

UV light is provided by the Cherenkov radiation derived from the intense gamma radiation of the spent fuel. CVDs are able to distinguish the glow patterns above fuel rods to distinguish spent fuel from non-fuel [11].



Figure 3. A fork detector produced by ANTECH, used by EURATOM to measure spent fuel assemblies [12].

For safeguards practices, bulk SNM handling facilities are organized into one or more material balance areas (MBAs) for MC&A purposes [10]. MBAs are areas in which inputs, outputs, and inventories of SNM are quantified and verified in order to ensure that no diversion (abrupt – diversion of an SQ in one attempt or protracted – diversion of an SQ in multiple attempts in a material balance period) of SNM has occurred. Flow and inventory values in MBAs are declared by a facility and verified by the IAEA. Important criteria in determining the MBAs include: whether the form (solid, liquid, or gas) of the

SNM handled in the MBA is same; whether the physical inventory in the MBA can be determined; the flow into and out of the MBA can be determined; and whether the MBA meets the given accuracies of NMA. Key measurement points (KMPs) are the strategic locations at which NMA measurements are taken inside or between the MBAs. For each MBA, there is an associated value of material-unaccounted-for (MUF) over a given material balance period (MBP). MBP is the time between inventory taking; MBPs are usually the same as the timeliness goals, which are: one month for unirradiated direct use material (for example: plutonium separated from fuel-salt of MSR), three months for irradiated direct use material (irradiated fuel-salt containing plutonium), and one year for indirect use material (fresh fuel-salt of MSR) [10]. MUF for a given MBA can be mathematically expressed as

$$\text{MUF} = \text{PB} + \text{X} - \text{Y} - \text{PE}, \quad (\text{Eq. 1})$$

where PB is the beginning inventory in an MBA, X is the inventory increase, Y is the inventory decrease, and PE is the ending inventory. The uncertainty associated with MUF is denoted as σ_{MUF} and is calculated using

$$\sigma_{\text{MUF}} = \sqrt{\sigma_{\text{PB}}^2 + \sigma_{\text{X}}^2 + \sigma_{\text{Y}}^2 + \sigma_{\text{PE}}^2} \quad (\text{Eq. 2})$$

which accounts for uncertainty associated with the terms in Eq. 1 for MUF. MBAs must meet the following IAEA governing conditions:

$$\text{MUF} < \text{SQ}; \quad (\text{Eq. 3})$$

$$\text{MUF} < 3\sigma_{\text{MUF}}; \text{ and} \quad (\text{Eq. 4})$$

$$3\sigma_{\text{MUF}} < \text{SQ}. \quad (\text{Eq. 5})$$

1.1. Objective

The work performed here was to develop a nuclear safeguards approach for a typical MSR. MSRs present a unique challenge from the perspective of applying nuclear safeguards and the corresponding nuclear MC&A. The safeguards challenges of MSRs mainly arise from the fact that they operate with molten nuclear fuel, which circulates within and outside the reactor core. This is distinctly different from light water or heavy water moderated nuclear reactors, which use fixed fuel assemblies or bundles. There are ongoing efforts to identify techniques capable of nuclear NMA measurements in molten salt. One candidate technique identified is hybrid K-edge densitometry (HKED) [13]. HKED can determine elemental concentrations of special nuclear material in molten salt to an uncertainty of less than 1%. HKED could be deployed at strategic points in the MSR system to support implementation of MBAs that are required to determine if any material is unaccounted for. The MUF value must be lower than one SQ in an MBP, to ensure that no diversion, either abrupt or protracted, of an SQ of SNM occurred from the MSR for non-peaceful uses.

1.2. Overview of MSR technology

There are numerous designs for MSRs, both domestic and international; there is large variation in MSR fuel cycles and reactor technologies which further complicates the design and application of safeguards strategies [5]. Each of the proposed designs will “need fresh fuel containing many significant quantities (SQ) of fissile/fertile materials” [5]. Multiple MBAs are required for MSRs and special “attention should be paid to

material in-process and material-unaccounted-for” given that liquid fuels require bulk nuclear material accountancy (NMA) [5].

Salt-fueled MSR designs are challenging because of the “continuous variation of isotopic concentrations in the fuel-salt from both transmutation and online chemical processing” [5]. Safeguards for MSR designs are further challenged by the fact that “there is the potential for online fuel processing whereby some fraction of the inventory can be removed while the reactor is operating” [5]. Additionally, existing safeguards technologies are likely not applicable to salt-fueled MSR designs given that they operate in high-temperature and high-radiation environments that are potentially highly corrosive; this is a challenge for any safeguards monitoring instrumentation as well as measurement techniques.

One interesting facet of MSR technology is that some MSR designs fall under the classification of small modular reactors (SMRs), which are defined as advanced reactors designed to generate power up to 300 MW_e [14]. SMRs are unique in that multiple reactors can be installed at a single site. As an example, the SMR developed by NuScale and the U.S. Department of Energy, generates 50 MW_e; up to 12 NuScale SMRs can be combined at a single site, which would generate up to 600 MW_e [15]. From a proliferation standpoint, this presents an interesting dilemma. A single SMR could fall within IAEA safeguards conditions, but when multiple SMRs at a single site are considered as one overall facility, it is conceivable that the IAEA safeguards conditions may no longer be met. Most of the current research related to multi-unit/multi-module aspects of SMRs focuses on nuclear safety rather than nonproliferation, so further development of SMR safeguards is needed [16].

1.3. International development of MSRs

While much of the early development of MSR technology was based in the US, MSR development evolved into an international endeavor. Following the Gen IV selection process, there was renewed interest in MSRs in Japan, Russia, China, France, and the UK [17]. Many MSR projects are multinational, so it is difficult to pinpoint the geographic spread of MSR development [5] [17]. Much of the development is within the NWSs, and within the European Union (including the Netherlands, Denmark, and Germany), India, South Korea, Indonesia, and Canada [5] [17].

1.4. Proliferation concerns relating to MSRs

The safeguards implications of the vast development of MSR technology cannot be understated. While much of the development of MSR technology has been concentrated within the NWSs, there is significant worldwide development of MSRs in NNWSs. Given the vastness of MSR designs, especially fuel cycles, it is conceivable that some MSR designs may present a substantial safeguards challenge. Thorium-based MSRs in particular will present a significant challenge for safeguards and nonproliferation. China and India are particularly strong in their pursuit of thorium-based fuel cycles and MSR technologies [18]. Given that the international safeguards community is focused on the conventional ^{235}U and ^{239}Pu fuel cycles, the international community must work to develop thorium fuel cycle safeguards approaches.

As stated previously, many MSR designs can be considered under the umbrella of SMRs. This provides a unique economically viable pathway for States to acquire nuclear technology. Many countries will likely pursue the purchase of MSR technologies from the

more advanced nuclear countries. As MSR technology spreads, it may become more difficult to safeguard units, especially when they are deployed in a multi-module configuration, as mentioned previously.

Overall, there are emerging proliferation concerns relating to MSRs. Due to the lack of existing safeguards approaches for MSRs and SMRs in general, there exists a significant need to develop a safeguards framework for MSRs, SMRs, and other Gen IV reactor designs.

1.5. International nuclear safeguards

Since the discovery of nuclear fission in 1938, it has been known that it is critical to control spread of nuclear technology. A November 1945 document called the Three Nation Agreed Declaration between the US, Canada, and the UK was one of the first documents regarding international nuclear energy policy; this document also contained the first instance of the term safeguards in the context of nuclear energy [19]. The US began entering into bilateral agreements in 1954 with respect to nuclear cooperation; these agreements included safeguards provisions to prevent the proliferation of nuclear material and technology for military uses [19]. The IAEA was established in 1957 as an intergovernmental organization affiliated with the United Nations (UN).

1.5.1. Motivations for safeguards

Nuclear safeguards schemes, and particularly the IAEA safeguards scheme, can only be effective if sensitive materials and facilities are not widely available in Non-Nuclear Weapon States (NNWSs) [19]. The goal of IAEA safeguards is to ensure that a significant quantity of nuclear material has not been diverted or misused by a State during

a given period of time. Further, IAEA safeguards require that detection of nuclear material diversion is timely.

1.5.2. IAEA safeguards goals

The IAEA has the authority to enter into safeguards agreements with individual States to ensure that nuclear materials, equipment, and facilities available for inspections were not diverted from peaceful uses to military purposes. The IAEA has the obligation to ensure that all nuclear “materials subject to safeguards are in fact safeguarded” [19]. The IAEA also monitors activities, to include: nuclear reactor operation, fuel fabrication, uranium enrichment, and plutonium separation in reprocessing plants [19]. Safeguards under the NPT focus on nuclear material, including weapon-usable plutonium and enriched uranium. Materials subject to safeguards include all plutonium and high enriched uranium (HEU) above gram quantities [19]. The NPT safeguards agreements do not distinguish between declared and undeclared materials, even though the safeguards monitoring of each of them are covered to some extent separately through INFCIRC/153 and INFCIRC/540, respectively. Full-scope safeguards is a term which refers to a comprehensive safeguards agreement, which obligate a State to accept safeguards on all source and special fissionable materials in peaceful nuclear activities [19].

1.5.3. Need for safeguards technologies

For IAEA safeguards, the main method used is NMA, which is supplemented by C/S and verified by inspections. In this sense, NMA is of fundamental importance for safeguards. Safeguards are “essentially an audit system” and can only detect the diversion of nuclear material; safeguards cannot prevent diversion of nuclear material [19]. There

are challenges with respect to existing safeguards technologies used to detect nuclear material diversion from bulk-handling facilities, mainly stemming from the systematic and random errors associated with these measurements.

Emerging technologies such as near-real-time accountancy (NRTA) methods by monitoring process parameters have not been used on a large scale by the IAEA, but will prove to be vitally important for large reprocessing plants and likely MSRMs [19]. Current technologies may still not be accurate enough at measuring material flows and inventories to detect the diversion of an SQ of material using the NRTA, but anomalies or unconventional operations could be observed.

NRTA methods monitor the in-process inventories of SNM in MBAs and flows in and out of MBAs at frequent intervals; the intervals between inventories are much shorter for NRTA than those in conventional NMA methods [19]. NRTA measurements can be “direct measurements from in-process instruments, off-line analysis, and data from computer simulations” [19]. The NRTA method also benefits from the fact that many measurements are taken, which allows for statistical tests to be used to increase the detection of any anomalies or unconventional operations; this should also improve the sensitivity of detection [19]. However, NRTA methods do not solve every problem associated with NMA, but rather may be susceptible to potential misuse. There is still the possibility that NRTA will not detect diversion of small amounts of nuclear materials. Over subsequent intervals or MBPs, it would therefore be possible that NRTA methods would miss the diversion of enough material to constitute an SQ [19].

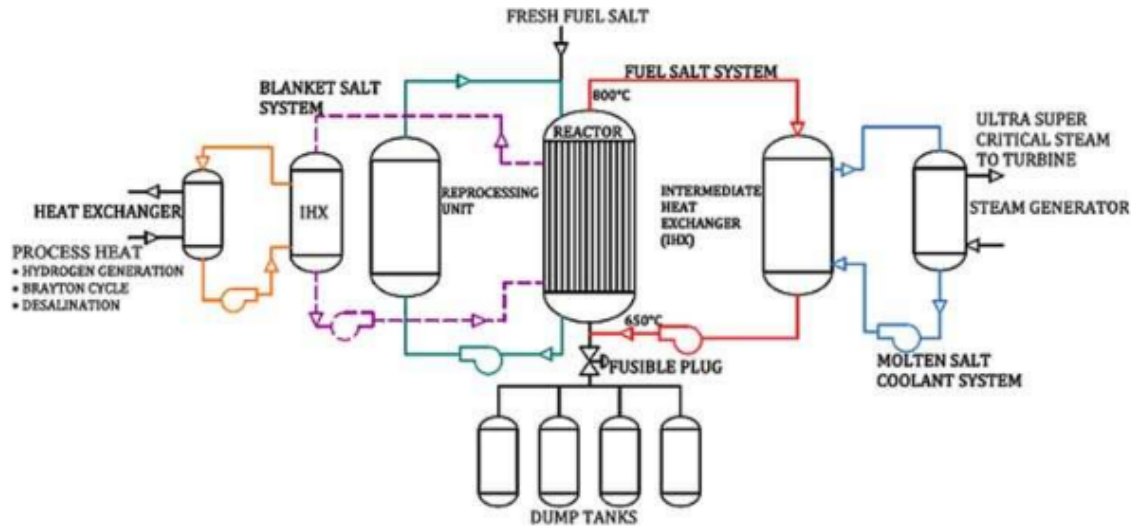


Figure 4. Schematic for the loop-type Indian molten salt breeder reactor (IMSBR) [20].

MSRs are unique and challenging from a safeguards perspective as they are essentially a reactor and fuel cycle combined in a single facility, and, further, because an MSR is a bulk SNM handling facility [6]. This is due to the fact that MSRs have transmutation, a characteristic of reactors, as well as changes in chemical and/or physical forms, a characteristic of reprocessing plants. Liquid-fueled MSRs are complex facilities, with a blanket-salt system, a fuel-salt system, a reprocessing unit, a heat exchanger, among many other systems. A schematic of the design for a loop-type MSR, the Indian molten salt breeder reactor (IMSBR) can be seen in Figure 4 [20].

Given their unique characteristics, MSRs will require nontraditional safeguards approaches. The unique characteristics of MSRs as both a reactor and fuel reprocessing plant are further complicated by the “intense heat and radiation arising from active nuclear fissioning” [6]. Thus, the fuel-salt must be recirculated to remove excess heat. Liquid-fueled MSRs utilize a homogenous mixture of “fuel, coolant, fission products, and

actinides” [6]. The isotopic concentrations of the fuel-salt are varying continuously in an MSR; this variation also includes both passive and active removal of “fission products, rare earth elements, and noble metals” [6]. This demonstrates that liquid-fueled MSRs are much more challenging from a safeguards perspective when compared to other reactors, such as online-refueled CANDU reactors.

The unique features of MSRs imply that their designers should consider safeguards as part of the design [13]. The implementation of safeguards-by-design (SBD) leads to “more effective and efficient safeguards” [21]. Further, SBD “increase the awareness of safeguards goals and requirements among all stakeholders” thereby improving the international safeguards community’s confidence in nuclear nonproliferation efforts [21]. The basic premise of SBD is that process operations are “designed to facilitate the effective and efficient application” while having negligible impact on the operational performance of a facility [21]. With respect to MSR designs, it is likely that a number of the systems used to provide operational data of the reactor, such as monitoring equipment, can also be used to meet IAEA safeguards objectives.

1.5.4. Challenges facing safeguards

Current safeguards approaches are challenged by liquid-fueled MSRs as MSRs are “unique, tightly coupled nuclear energy systems with the reactor core, balance of plant, and fuel cycle [...] combined in a single facility” [5]. The inspection system of the IAEA is based on item counting for nuclear reactors and “bulk material accountancy for the front and back end of the nuclear fuel cycle” [5]. Bulk material accountancy methods have not

yet been applied to liquid-fueled MSRs, nor has it been proven that they are applicable to the required sensitivity.

In order to meet IAEA safeguards goals, MSRs will require high-accuracy measurement systems. IAEA measurement goals are guided by International Target Values (ITVs) for measurement uncertainties [22]. MUF for item accountancy is general zero when there has not been a diversion of material. Bulk handling type facilities, such as MSRs, have expected, general estimates for measurement uncertainties associated with accounting measurements. These relative standard deviation values are multiplied by the material throughput to obtain an uncertainty for an MBA. Given the random and systematic measurement uncertainties associated with destructive analysis (DA) and non-destructive (NDA) measurements performed when performing NMA on bulk material type facilities, a non-zero value for MUF is expected. For bulk measurements, the value of the MUF can never be zero; the MUF value will “ideally be statistically close to zero” [10].

When applying safeguards to MSRs to quantify SNM, the IAEA will likely have to rely on “technologies and approaches such as continuous monitoring of reactor process” [5]. A candidate technique is hybrid K-edge densitometry, which has been successfully applied in commercial aqueous reprocessing facilities in the on-site laboratories (OSL) of Rokkasho used fuel reprocessing plant in Japan and La Hague in France [22] [23]. It is also likely that remote and unattended monitoring will need to be applied to MSRs, given the challenging operating environment.

Currently, the IAEA has not developed a model approach for safeguards measures in MSRs; no current bulk accountancy techniques can be directly applied to MSRs nor salt-fueled MSRs in particular. It is also critically important that MSR developers implement SBD to “maximize [the effectiveness of safeguards] and minimize their burden on reactor operations” [5]. Some argue that the Agency is not looking enough to the future of nuclear reactor technologies and how they will adapt the international safeguards policies, concepts, approaches, and technologies [5].

1.6. Literature review

In September 2018, ORNL reported the results of a study, where they proposed a roadmap for safety and licensing of commercial MSRs. This study was done to identify research and development gaps of MSRs [24]. The authors brought out that, given MSRs are salt-fueled, inspectors will not have the ability to verify the presence of discrete items of SNM because “the material of interest is [...] in the [molten] salt and isotopes are continuously being consumed and created” [24]. Safeguards challenges of MSRs are also discussed given that on-line fuel-salt processing could “potentially provide pathways for the removal of [special] nuclear materials” [24]. The authors also noted that it is necessary to include safeguards while designing an MSR as any necessary measurement instrumentation will be “integral to the plant” as “MSR technology includes a tightly coupled reactor with fuel cycle operations (reprocessing and recycling) housed in a single facility” [24]. This study emphasized the importance of developing new safeguards technologies as well as evaluating current safeguards technologies that may be applied to meet the challenges given the unique aspects of MSRs [24]. Factors that are unique to

MSR safeguards include: the homogeneous mixture of fuel, molten coolant, fission products, and actinides; continuous variation of isotopic concentrations in the fuel-salt, including removal (passive or active) of fission products, rare earth elements, and noble metals; and the presence of fuel outside the vessel [24].

In October 2018, ORNL conducted a study on the regulatory gaps for MSRs; this study analyzed how nuclear regulations for LWRs are not fully applicable to MSRs [25]. However, the scope of this study did not include nuclear safeguards.

A May 2016 American Nuclear Society article titled “Safeguards Considerations for Thorium Fuel Cycles” focused on thorium molten salt reactors [18]. However, the paper also discussed considerations for defining safeguards approaches for MSRs, stating: “In general, MSRs potentially combine the processes of fuel fabrication, reactor irradiation, chemical separations, and waste assay in one facility, depending on the reactor design. Therefore, MSRs present a number of potentially significant safeguards challenges” [18]. One of the main challenges for MSR safeguards is the fact that the “mass of fissile material in the reactor or reprocessing plant [is] not constant and cannot be considered” for item accounting [18]. Furthermore, liquid-fueled MSRs must operate safeguards technology in a very different way than the traditional reactor types. Given the SQ values of SNM, IAEA inspections will need to occur more frequently and/or on an unannounced basis at MSR facilities than at other reactor facilities. This article also discussed the need to define KMPs and to utilize SBD for MSRs. Lastly, this article mentioned that “hybrid destructive and NDA technologies, such as the hybrid K-edge

densitometer (HKED)” should be employed for MSRs as these facilities can be considered as “both a reactor and [a] fuel processing facility” [18].

ORNL has a domestic HKED measurement system, which is located in the Radiochemical Engineering Development Complex (REDC). ORNL has also contributed significantly to the study of HKED and its application to safeguards [26]. A University of Tennessee, Knoxville graduate dissertation has studied the application of HKED to pyroprocessing, though further work is needed to verify that HKED can be used for molten salt [27]. Pyroprocessing is a method of electrochemical reprocessing. Pyroprocessing combines plutonium in spent nuclear fuel with other waste products from nuclear fuel, including: neptunium, americium, and curium. This recycled plutonium can then be used in breeder or mixed oxide (MOX) reactors. The fuel matrix produced by the pyroprocessing method has higher levels of plutonium than traditional fuels; the ratio of concentrations of uranium and plutonium in MOX approach 1:1. Pyroprocessed fuel also has higher levels of minor actinides like americium and neptunium [28]. Contrastingly, MSR fuel-salt has much smaller uranium to plutonium ratios, as well as smaller concentrations of minor actinides. Thus, the method of HKED used in pyroprocessing activities cannot be directly applied to an MSR safeguards approach; further study of HKED with MSR fuel will be required.

David LeBlanc’s 2009 article “Molten salt reactors: A new beginning for an old idea” discusses much of the history of MSR development as well as emerging MSR designs [3]. LeBlanc briefly mentions that “proliferation resistance is a highly contentious subject but one of great importance” [3]. As worldwide governments and industry

continue to develop advanced MSR designs and technology, safeguards will come to the forefront as an important issue before MSR can be used for civilian power needs.

1.7. Why is this study different?

This was a novel work in that it designed, modeled, and simulated the fuel burnup of a theoretical MSR using both MCNP and SCALE neutronics codes. Fuel depletion simulations were done to determine the amount of SNM in the reactor [29] [30]. The results of the fuel depletion simulations were used in an analysis of the suitability of an emerging technology, HKED, to nuclear safeguards applications in MSRs.

2. METHODOLOGY

A theoretical MSR core design was developed and modeled using two neutronics codes: (1) the Los Alamos National Laboratory (LANL)-developed Monte Carlo N-Particle (MCNP version 6.2) radiation transport code and (2) the ORNL-developed Standardized Computer Analyses for Licensing Evaluation (SCALE version 6.2) radiation transport code [29] [30]. MSR fuel-salt burnup simulations were performed using both MCNP and SCALE models to compute the inventory of SNM production and flow rates. An MBA structure was determined for the MSR in which the core, blanket, and reprocessing areas of the MSR are separated into three MBAs. Each of the three core loops and their respective reprocessing loops were considered separately for material balance purposes. The results from the fuel burnup simulations using MCNP and SCALE were then incorporated into the MBA process in order to develop an approach for MSR safeguards.

2.1. Theoretical molten salt reactor core

As part of this study, a theoretical MSR core design was developed at Texas A&M University. The core was a thermal neutron reactor type with graphite as the neutron moderator and molten uranium salt fuel mixed with lithium and beryllium fluoride as the fuel. The reactor was designed for a rated output of 300 MW_{th}. The fuel-salt, composed of uranium tetrafluoride (UF₄) with a ²³⁵U enrichment of 3.5 wt.% UF₄, constitutes 20% of the molten fuel by weight, with the remaining 80% being molten lithium and beryllium fluoride (FLiBe).

The reactor core is cylindrical with a hexagonal lattice design, which consists of 61 molten fuel-salt (enriched UF₄-FLiBe) channels and 108 molten blanket-salt (depleted UF₄-FLiBe) channels in its lattice locations, each connected to a salt plenum at the top and bottom of the core. The core is surrounded by a graphite radial neutron reflector and a boron carbide (B₄C) neutron absorber to prevent neutron leaking outside the core for safety purposes. The reactor core is enclosed in a Hastelloy-N reactor vessel. The UF₄-FLiBe fuel-salt is set to operate at 900 K. The 61 fuel-salt channels are surrounded in three hexagonal layers of blanket-salt channels for fuel breeding purposes; there are 108 depleted UF₄-FLiBe blanket-salt channels. The fuel-salt and blanket-salt both have densities of 2.892 g/cm³.

An important part of the MBA calculations was to determine the flow rate of the fuel-salt. The flow rate was determined by dividing the power in Watts by the product of the change in temperature in degrees Celsius times the heat capacity in units of J/kg-°C times the density.

$$flow\ rate\ \left[\frac{m^3}{s}\right] = \frac{power\ [W]}{\Delta T\ [^{\circ}C] \times heat\ capacity\ \left[\frac{J}{kg - ^{\circ}C}\right] \times density\ \left[\frac{kg}{m^3}\right]} \quad (Eq. 6)$$

For calculation purposes, the heat capacity of FLiBe with 20% UF₄ was 1700 J/kg-°C, which was assumed to be at a temperature of 700°C. The change in temperature, ΔT, was assumed to be 100°C, based on similar reactor design information available in literature. As mentioned previously, the MSR was burned at a power level of 300 MW. Thus, the flow rate was determined to be 1765 kg/s. Using the density of the fuel-salt, which is 2.892 g/cm³, the required core flow rate was determined as 0.6 m³/s using Eq. 6.

The molten fuel-salt flows through the core at $0.6 \text{ m}^3/\text{s}$; this flow is split into three loops with an identical flow rate of $0.2 \text{ m}^3/\text{s}$ in each loop. The molten fuel-salt volume in the MSR core is 1.04 m^3 and the mass of the fuel-salt is 3013.5 kg . Given the FLiBe with 20% UF_4 , the uranium weight fraction in the fuel-salt is 0.33509 , which gives the total uranium in the fuel-salt as 1009.8 kg . Given that the ^{235}U enrichment in the fuel-salt is 3.5%, ^{235}U flow in one loop of the core is $6.78 \text{ kg/s} \pm 0.019 \text{ kg/s}$. The total plutonium in the fuel-salt is 3.74 kg based on the results of the neutronics calculations aforementioned using MCNP. Thus, in one loop of the core, the total plutonium is 1.25 kg . The flow rate of plutonium in one loop is therefore $0.717 \text{ kg/s} \pm 0.00201 \text{ kg/s}$. The uncertainty in the SNM flow values was calculated using the International Target Value (ITV) for the proposed nondestructive assay (NDA) detector, the HKED, which has an ITV value of 0.28% [22]. The HKED is used measuring uranium and plutonium concentrations in the molten salt samples.

There are three reprocessing loops in the MSR, one for each of the three loops of the core, as mentioned above; $0.05 \text{ m}^3/\text{d}$ of the molten fuel-salt flows through each reprocessing loop, which means the fraction of molten fuel-salt in the reprocessing loop compared to the core loop is 2.89×10^{-6} . Thus, the amount of ^{235}U in one reprocessing loop is $1.96 \times 10^{-5} \text{ kg/s} \pm 5.50 \times 10^{-8} \text{ kg/s}$. The total plutonium in one reprocessing loop is $2.076 \times 10^{-6} \text{ kg/s} \pm 5.81 \times 10^{-9} \text{ kg/s}$, which is very small compared to the reactor core loops. The uncertainty in the flow value was calculated again using the IAEA ITV value for HKED, as described in the previous paragraph [22].

The blanket of the MSR also has three loops. The volume of blanket-salt is 5.65 m³ and the total mass of the blanket-salt is 1.63×10^4 kg. Given the uranium weight fraction is 0.33518 in the molten blanket-salt and since ²³⁵U enrichment is only 0.2% (depleted uranium) in the molten blanket-salt, the flow of ²³⁵U in the blanket loop is 0.388 kg/s $\pm 1.09 \times 10^{-3}$ kg/s. The flow of plutonium in the blanket loop is 0.148 kg/s $\pm 4.14 \times 10^{-4}$ kg/s.

2.2. Reactor physics (fuel burnup neutronics) simulations

Two models of the MSR reactor core design were developed: one using the LANL-developed MCNP-6.2 radiation transport code and another one using the ORNL-developed SCALE-6.2 radiation transport code.

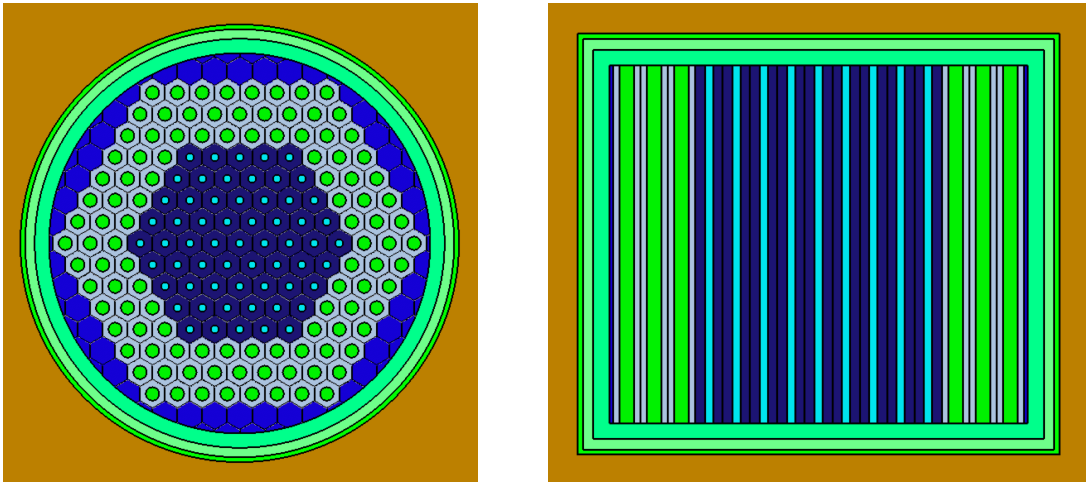


Figure 5. (left) top view, and (right) side view of the MSR core model using SCALE. Legend: brown (air), green shades are surrounding materials from outermost position (Hastelloy-N, boron carbide, graphite), blue (graphite), grey hexagon with green core (graphite with blanket-salt channel), navy blue hexagon with cyan core (graphite with fuel-salt channel).

Figure 5 and Figure 6 show the planar and elevation view of the reactor core obtained using SCALE and MCNP codes, respectively. Using both reactor code models,

neutronics simulations were carried out first to compare the values of effective neutron multiplication factor (k -effective) for the steady-state condition of the reactor. Reactor physics simulations were conducted, which included fuel burnup to quantify and analyze the neutronics and SNM evolution as a function of time for the proposed MSR core design.

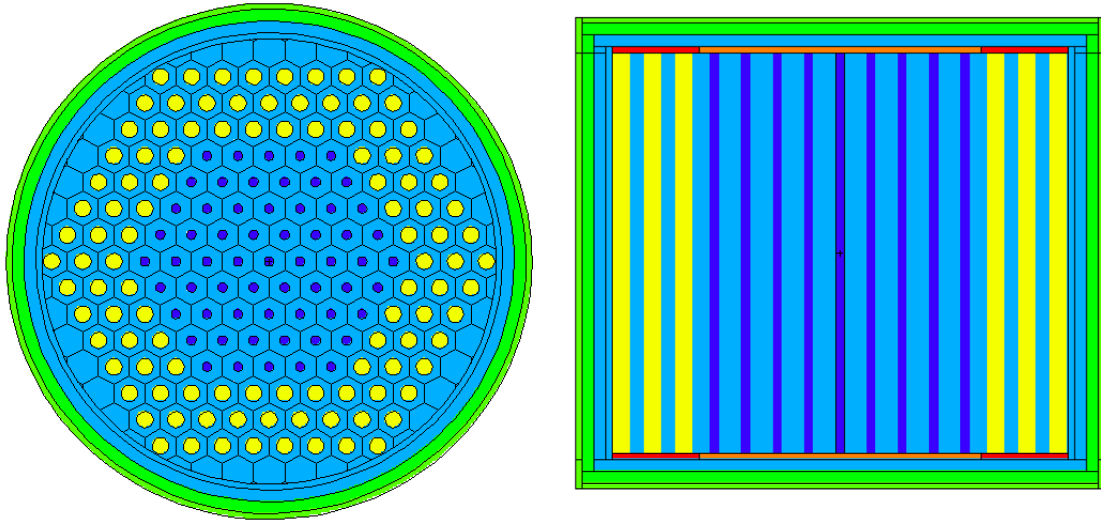


Figure 6. (left) top view, and (right) side view of the MSR core model using MCNP. Legend: green shades are surrounding materials from outermost position (Hastelloy-N, boron carbide), light blue (graphite), light blue hexagon with yellow core (graphite with blanket-salt channel), light blue hexagon with dark blue core (graphite with fuel-salt channel), orange (fuel-salt plena), red (blanket-salt plena).

2.2.1. MSR Fuel Burnup Simulations using SCALE

The fuel-salt and blanket-salt were both burned using SCALE code; however, the assembly-averaged power was normalized only to the fuel-salt. The molten fuel-salt and blanket-salt depletion simulation was carried out at a specific power of $260 \text{ MW}_{\text{th}}$, which is equivalent to a reactor core power of $300 \text{ MW}_{\text{th}}$. This was done as this corresponds to the method in which depletions occur in the MCNP model.

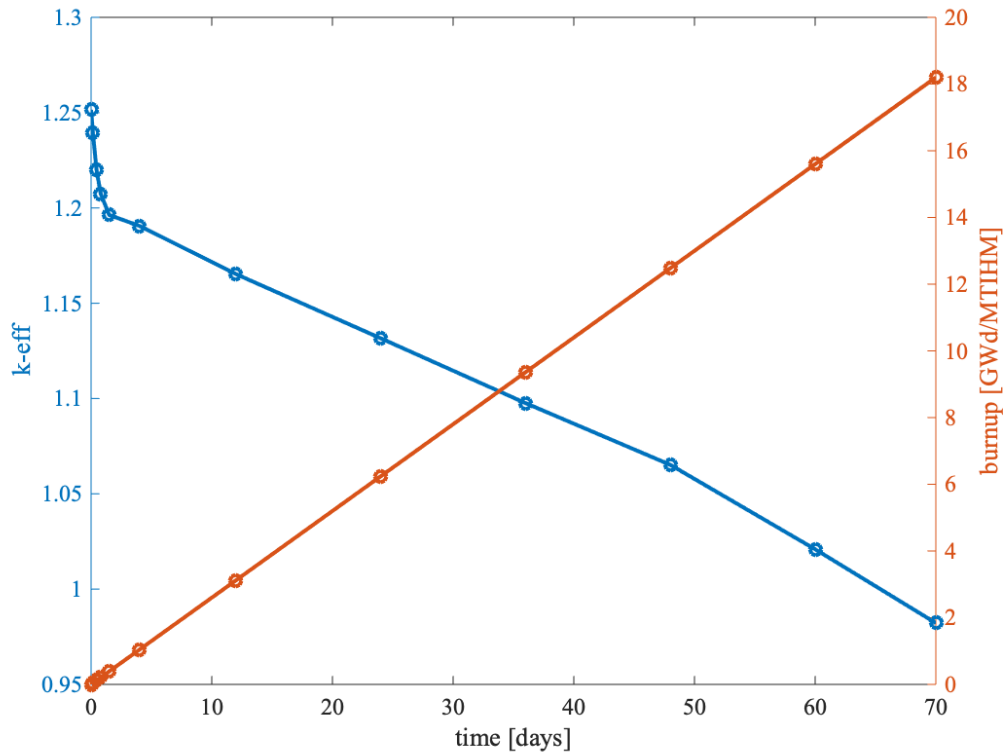


Figure 7. Effective neutron multiplication factor (k-eff) and fuel-salt burnup values vs. time for the MSR depletion (one fuel cycle) using SCALE.

The initial neutron multiplication (k-effective) value was 1.2515, which reduced to a value of 1.0209 in 60 days of fuel burnup. The burnup obtained was 15.6 GWd/MTIHM for the 60-day burnup. Figure 7 shows the change in effective neutron multiplication factor, k-eff (a neutron reactivity measure), as a function of fuel burnup time in days. An initial drop in k-effective, as seen in Figure 7, is due to the initial fission product buildup (such as ^{135}Xe , a neutron poison). Subsequently, after a four-day fuel burnup time, the k-eff value trended linearly downward. The corresponding fuel-salt burnup in gigawatts-day per metric ton of uranium heavy metal (GWd/MTHM) increased linearly over time, as expected.

A two-part recycling scheme is required as part of the MSR refueling: the first one is for the gaseous extraction to remove non-soluble fission products; and the second one is to remove fission products in the offline salt reprocessing [31]. Gaseous extraction is designed to remove the following elements: with atomic number (Z) = 1, 7, 8, 36, 41, 42, 43, 44, 45, 46, 47, and 54. The offline extraction removed the following fission products with $Z = 40, 48, 49, 50, 53, 55, 56, 57, 58, 60, 61, 62, 63,$ and 64. The fission product removal was not implemented in the SCALE simulations, which can be reserved as future work and would provide the most realistic modeling of the MSR using SCALE code. Subsequent refueling cycles using SCALE model were not successful due to the limitations of the code in reconstituting the fuel-salt. It is necessary to implement refueling of the MSR in SCALE to determine the effectiveness of using SCALE for SNM quantification purposes and also to compare SCALE code results with that obtained using MCNP simulations.

2.2.2. MSR fuel burnup simulations using MCNP

The molten fuel-salt and blanket-salt were burned at a reactor power level of 300 MW_{th}. The depletion of the fuel-salt was done until the neutron reactivity (k-effective value) reached approximately 1.0, as shown in Figure 8; at that point, when considered neutronically, the reactor core is no more reactive or critical produce the rated power output. The fuel-salt reached a burnup of 3.238 GWd/MTHM in 70 days with a corresponding value of k-effective as 1.01351. At this point, the fuel-salt was reconstituted and depleted again in the next fuel cycle. The fuel-salt was reconstituted such that the

amount of uranium in the reconstituted fuel was the same as the amount of uranium in the fresh UF₄-FLiBe fuel.

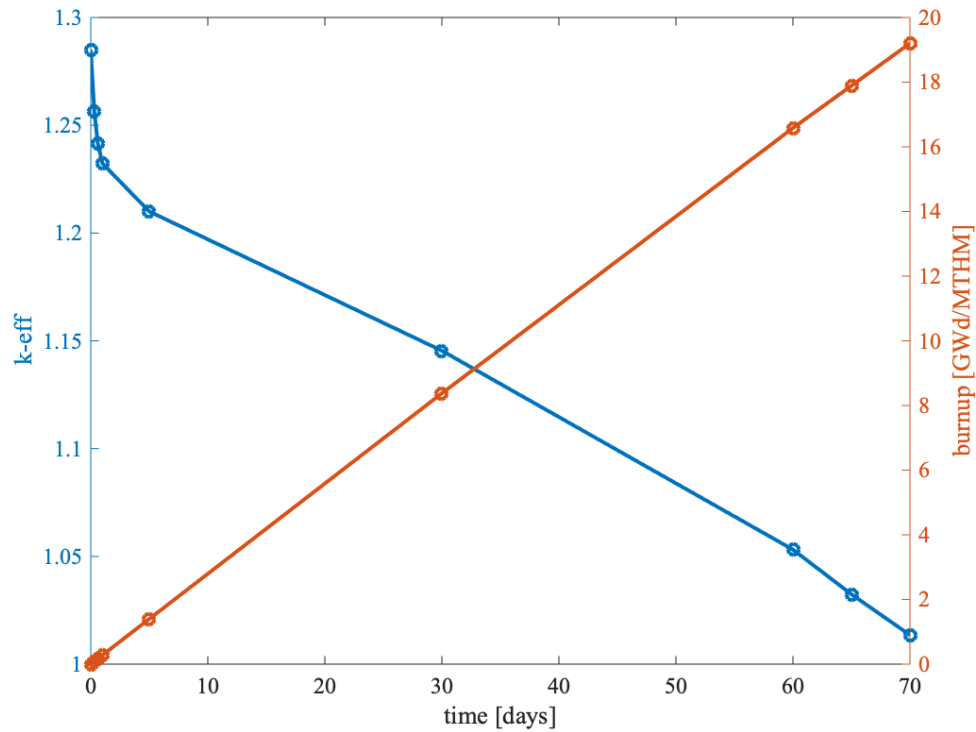


Figure 8. Effective neutron multiplication factor (k-eff) and fuel-salt burnup values vs. time for the first fuel cycle using MCNP.

2.3. SNM quantification and NMA analysis for nuclear safeguards

2.3.1. MBAs and KMPs

Designation of MBAs and strategic points for KMPs was necessary to begin the safeguards analyses. The proposed MBA scheme for the MSR under study was to separate the core, reprocessing, and blanket into three separate MBAs, which is shown in Figure 9. There are KMPs between each of these MBAs. These KMPs are the points at which the proposed NDA detector (HKED) measurements take place to determine if any SNM

diversion occurred. The IAEA recommended international target value (ITV) uncertainty of 0.28% for HKED is used in this study [32] [22].

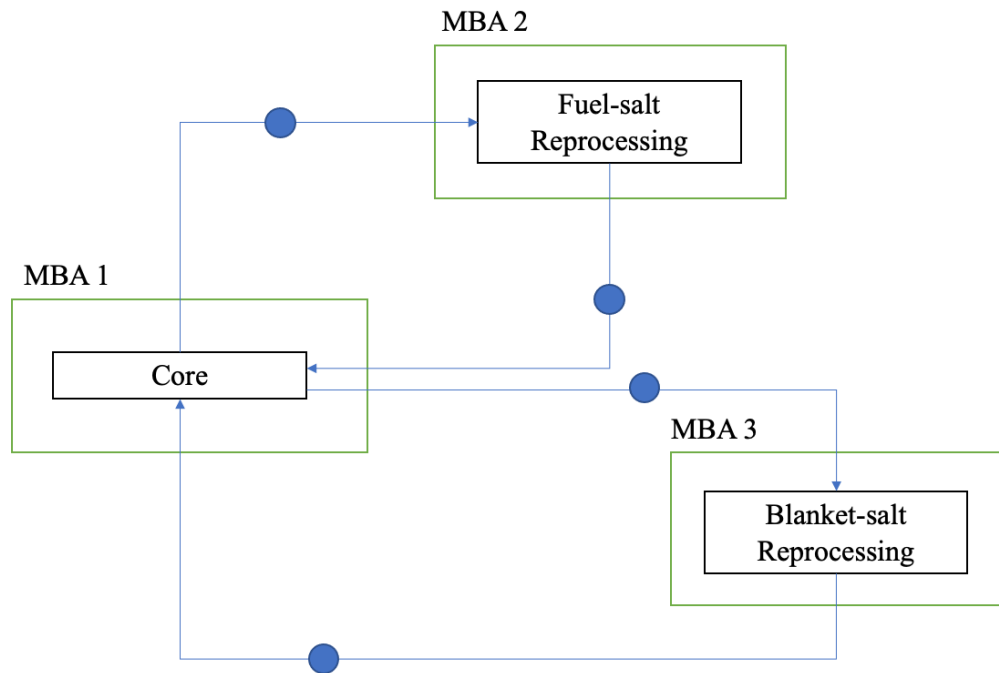


Figure 9. Proposed MBA scheme for the MSR under study.

Only one loop of the three reprocessing loops is considered for the inventory calculations in the current study; subsequent analyses were performed considering all three loops, as detailed in Section 3.2.4. The values for physical inventory at the beginning of the MBP, denoted as PB, and at the end of the MBP, denoted as PE, are set to zero for inventory calculations; this convention is commonly practiced in bulk material handling facilities.

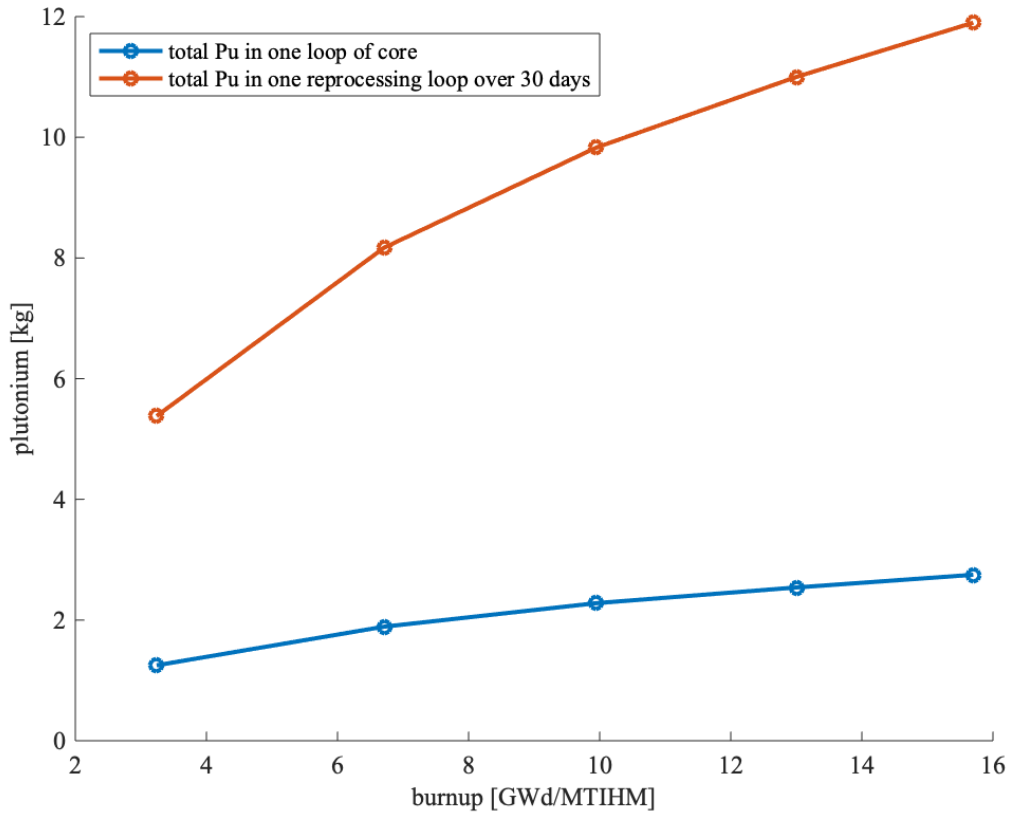


Figure 10. Plutonium production as a function of burnup.

There are three reprocessing loops in the MSR under study, one for each of the three loops of the core; 0.05 m³/d of the molten fuel-salt flows through each reprocessing loop, which means the fraction of molten fuel-salt in the reprocessing loop compared to the core loop is 2.89×10^{-6} . Thus, the amount of ²³⁵U in one reprocessing loop is 1.96×10^{-5} kg/s $\pm 5.50 \times 10^{-8}$ kg/s. The amount plutonium in one reprocessing loop is 2.07×10^{-6} kg/s $\pm 5.81 \times 10^{-9}$ kg/s. Figure 10 shows the amount of plutonium as a function of burnup. The uncertainties in all flow values were calculated using the International Target Value (ITV) for the proposed detector, HKED, which is 0.28%.

2.3.2. Reprocessing cycle of blanket loops

As was done with the core, the safeguards analysis was also applied to the reprocessing cycle of the blanket loop. The blanket loop is also arranged in three loops in this MSR design.

2.3.3. Analysis of holdup accumulation and waste

It was necessary to determine the amount of SNM that constituted holdup accumulation and waste given that, at some point, these amounts may exceed an SQ. Thus, an analysis of holdup accumulation and waste was also conducted to determine the point at which the SNM content in either the holdup or waste exceeds an SQ. For holdup, this required assuming a holdup value, as previously described. For waste, it was assumed that 0.5% of the fuel-salt would be lost to waste given the fuel-salt reprocessing efficiency.

2.3.4. Consideration of multi-module MSR facility

Further consideration of the safeguards approach for the MSR design was done for a multi-module MSR facility. Given that a single SMR site is likely to generate around 1500 MW_{th}, the MBA analysis was applied to a facility with four MSRs, each generating 300 MW_{th} for a total facility electricity generation of 1200 MW_{th} [15]. This analysis can be scaled up or down as needed to fit within a proposed multi-module MSR facility design.

2.4. Hybrid K-edge densitometer for MSR safeguards applications

2.4.1. Hybrid K-edge densitometry

Los Alamos National Laboratory developed the hybrid K-edge densitometer (HKED) [33]. The HKED is a system that integrates analyses from K-edge densitometry (KED) and X-ray fluorescence (XRF), as shown in Figure 11 and Figure 12 [34] [35]. The

HKED system has an X-ray tube to produce photons needed for KED measurements and to serve as “the excitation source” for XRF measurements [27]. The sample is held adjacent to this X-ray tube; this is the point at which the two measurements are made. The beamline for KED measurements is opposite the sample holder and X-ray tube [27].

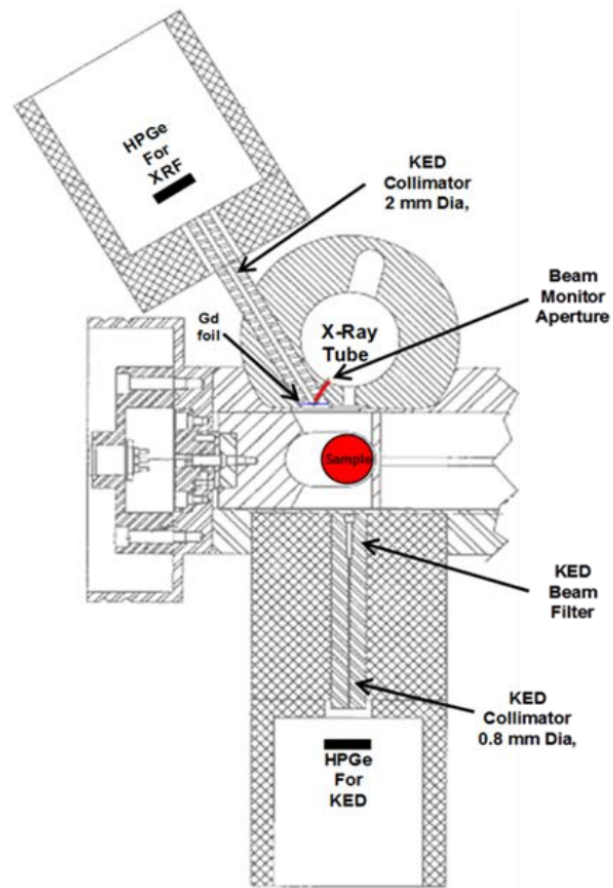


Figure 11. Schematic of HKED system combining KED and XRF subsystems [34].

The KED method is used to find the major actinide density in a sample of material, but cannot determine the elemental composition of the sample. KED measures binding energy of electrons in the element’s K-shell. Photons are “preferentially absorbed at this energy” as the X-ray beam passes through the sample creates a vacancy, which is then filled by a de-excited electron from a higher shell; this process emits a characteristic X-

ray which is measured by the XRF [27]. XRF measurements are able to indicate the identity and concentration of the element, using the energy and intensity of the emitted X-ray, respectively [27].



Figure 12. HKED system available from Mirion Technologies [35].

2.4.1.1. K-edge subsystem

Once calibrated, the KED subsystem measures the “transmission differential from a Bremsstrahlung X-ray source [...] across the K-shell absorption edge of the various elemental constituents of the solution,” thereby determining actinide concentrations [36].

Each element has a unique K-shell transition energy, so measurement of the K-edge is a convenient method to “identify and distinguish” materials.

2.4.1.2. X-ray fluorescence subsystem

In the XRF subsystem, incident X-rays are used to stimulate the sample and produce fluoresced X-rays, which are then measured using X-ray fluorescence. Each actinide has a characteristic fluoresced spectrum, which further increases the accuracy of characterizing samples.

2.4.1.3. Hybrid K-edge technique

HKED systems “provide accurate, rapid assay results” of plutonium and uranium content in sample solutions; HKED results are produced near real time [36]. The HKED provides high-precision and high-accuracy NDA and is a “long-established technique” in IAEA and European Atomic Energy Community (EURATOM) safeguards schemes [23] [37].

2.4.1.4. Hybrid K-edge accuracy

The HKED is commended for its accuracy, with the KED being “nearly absolute” and the XRF technique having “the character of a relative measurement;” calibration is done for correction factors and is not a major concern [36]. A typical assay of one hour in duration using HKED is done on a vial containing a few milliliters of the sample solution. The KED is used to determine the concentration of uranium to “<0.2% accuracy” and the XRF provides the concentration of minor constituents, such as “plutonium to an accuracy of <0.8%” [36]. HKED is an “optimized blend of both techniques” and has already been successfully implemented in some nuclear safeguards applications; HKEDs are currently

installed at reprocessing facilities in La Hague, Sellafield, Rokkasho-mura, Lanzhou, and Mayak [23] [37] [28].

2.4.2. Hybrid K-edge densitometry applicability to MSRs

HKED can be used to determine the elemental concentration of SNM (U/Pu) in sample solutions, without chemical separations. Hybrid densitometers have traditionally been used in safeguards for reprocessing plants. Based on previous MSR safeguards studies, HKED is thought to be the most promising detection technique for MSR NMA applications.

2.4.3. NMA analysis using HKED

For this study, it was necessary to propose a detection technique for NMA purposes and conduct NMA analysis using the detection technique at designated strategic points. The reprocessing flow assumed in this study is 50 L/d given the flow rate in each of the three core loops is 200 L/s. A holdup value of 0.0003% is assumed, which is the ratio of fuel-salt flow rate in a single reprocessing loop (50 L/d) to a single core loop (1.728×10^7 L/d) flow rate. It was assumed that 0.5% of the fuel-salt would be lost to waste given the fuel-salt reprocessing efficiency. An MBP of three months was designated for ^{235}U and plutonium because, per the IAEA definition of SNM, this material of the fuel-salt falls under the category of irradiated direct use material and the timeliness goal for diversion detection for such a material is three months. Fresh fuel-salt of the MSR would fall under indirect use material, with a timeliness goal of 12 months. Uncertainties in the MUF are calculated using the IAEA recommended international target value (ITV) uncertainty of 0.28% for HKED.

2.4.4. Suitability of the detection technique

It was necessary to determine the suitability of the detection technique as applied to nuclear safeguards and determine the efficacy of the proposed MBAs and MBPs for ^{235}U and plutonium. The HKED has a very low uncertainty value, which makes it attractive for safeguards purposes. The ability of HKED to be an online process cannot be established at this time, however, ORNL is working to develop this technology as an online NMA method.

It is important to note that the use of HKED for molten salt applications will likely be a destructive process to measure the SNM in the sample. This is because molten salt samples will need to be removed from the fuel-salt loop and will likely not be able to be returned into the MSR. Further, due to the fact that molten salt has a much higher density as compared to typical pyroprocessing samples, molten salt samples will likely need to be diluted before they can be measured using HKED. Overall, there are outstanding questions about whether the sample will need to be diluted so that its density is within the capability of HKED measurements. These questions were deemed to be outside the scope of this work as many have agreed that HKED will likely emerge as the detection for molten salt in the future.

3. RESULTS

3.1. MCNP and SCALE-based MSR core fuel burnup simulation results

MSR fuel burnup simulations were done in both MCNP and SCALE. The effective neutron multiplication values (k-eff values) from MCNP and SCALE simulations were compared, as described previously in METHODOLOGY. These results showed that the two models were effectively equivalent in terms of neutron multiplication factor and burnup over the first fuel cycle. Refueling cycles were not successful in SCALE due to limitations of the code with respect to reconstituting the fuel-salt for simulating a number of fuel cycles. Thus, the SCALE model was only analyzed for one fuel cycle.

MCNP was used to burn the core of the MSR until a k-eff value of approximately 1.0 was reached. At that point, the fuel-salt was reconstituted and depleted again. The results of the MCNP fuel depletion simulations were used to determine the amount of SNM present in the MSR.

3.2. MBA inventory and nuclear material accountancy calculations

3.2.1. Core reprocessing loop

For the MBP of three months (SNM in the reprocessing loop being irradiated direct use material), the MUF value for ^{235}U is found to be 0.764 kg, which is well within the SQ value of 75 kg. This demonstrates that the MBP is adequate for an effective NMA scheme for MBA-2 as the MUF value is within the IAEA requirements $\text{MUF} < \text{SQ}$, $\text{MUF} < 3\sigma_{\text{MUF}}$, and $3\sigma_{\text{MUF}} < \text{SQ}$. These results are summarized below in Table 1.

Table 1. ²³⁵U MUF calculations for one loop of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months
PB (kg)	0	0	0
X (kg)	5.09E+01	1.53E+02	6.19E+02
σ X (kg)	1.42E-01	4.27E-01	1.73E+00
Y (kg)	5.06E+01	1.52E+02	6.16E+02
σ Y (kg)	1.42E-01	4.25E-01	1.72E+00
Holdup (kg)	0	0	0
Waste (kg)	2.54E-01	7.63E-01	3.10E+00
PE (kg)	0	0	0
MUF (kg)	2.55E-01	7.64E-01	3.10E+00
σ MUF (kg)	2.01E-01	6.03E-01	2.44E+00
3σ MUF (kg)	6.03E-01	1.81E+00	7.33E+00

Analysis of the Pu in the reprocessing MBA showed that the MUF value for Pu was 0.0269 kg for an MBP of 30 days. This is within the IAEA SQ value for Pu of 8 kg.

The MUF calculations for Pu for one loop of the reprocessing MBA are shown in Table

2.

Table 2. Plutonium MUF calculations for one loop of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months
PB (kg)	0	0	0
X (kg)	5.38E+00	1.61E+01	6.54E+01
σ X (kg)	1.51E-02	4.52E-02	1.83E-01
Y (kg)	5.35E+00	1.61E+01	6.51E+01
σ Y (kg)	1.50E-02	4.49E-02	1.82E-01
Holdup (kg)	0	0	0
Waste (kg)	2.69E-02	8.07E-02	3.27E-01
PE (kg)	0	0	0
MUF (kg)	2.69E-02	8.07E-02	3.27E-01
σ MUF (kg)	2.12E-02	6.37E-02	2.58E-01
3σ MUF (kg)	6.37E-02	1.91E-01	7.75E-01

3.2.2. Blanket reprocessing loop

The safeguards analyses were applied to the reprocessing cycle of the blanket loop.

As discussed previously, the blanket was separated into three loops.

Table 3. ²³⁵U MBA calculations for one loop of the reprocessing cycle of the blanket loop.

MBP	1 month	3 months	12 months	24 months	48 months	123 months	126 months	132 months
PB (kg)	0	0	0	0	0	0	0	0
X (kg)	9.69E-01	2.91E+00	1.18E+01	2.36E+01	4.72E+01	1.21E+02	1.24E+02	1.30E+02
σ X (kg)	2.71E-03	8.14E-03	3.30E-02	6.60E-02	1.32E-01	3.38E-01	3.47E-01	3.63E-01
Y (kg)	9.64E-01	2.89E+00	1.17E+01	2.35E+01	4.69E+01	1.20E+02	1.23E+02	1.29E+02
σ Y (kg)	2.70E-03	8.10E-03	3.29E-02	6.57E-02	1.31E-01	3.37E-01	3.45E-01	3.61E-01
PE (kg)	0	0	0	0	0	0	0	0
MUF (kg)	4.85E-03	1.45E-02	5.90E-02	1.18E-01	2.36E-01	6.05E-01	6.20E-01	6.49E-01
σ MUF (kg)	3.83E-03	1.15E-02	4.66E-02	9.32E-02	1.86E-01	4.77E-01	4.89E-01	5.12E-01
3 σ MUF (kg)	1.15E-02	3.45E-02	1.40E-01	2.80E-01	5.59E-01	1.43E+00	1.47E+00	1.54E+00

Table 3 shows the ²³⁵U MBA calculations for one loop of the reprocessing cycle of the blanket loop. These results show that the reprocessing cycle of the blanket loop will be within IAEA limits of ²³⁵U for at least 132 months. Table 4 shows the plutonium MBA calculations for one loop of the reprocessing cycle of the blanket loop. These results show that the reprocessing cycle of the blanket loop will be within IAEA limits of plutonium for at least 132 months.

Table 4. Plutonium MBA calculations for one loop of the reprocessing cycle of the blanket loop.

MBP	1 month	3 months	12 months	24 months	48 months	123 months	126 months	132 months
PB (kg)	0	0	0	0	0	0	0	0
X (kg)	3.69E-01	1.11E+00	4.49E+00	8.99E+00	1.80E+01	4.61E+01	4.72E+01	4.94E+01
σ X (kg)	1.03E-03	3.10E-03	1.26E-02	2.52E-02	5.03E-02	1.29E-01	1.32E-01	1.38E-01
Y (kg)	3.67E-01	1.10E+00	4.47E+00	8.94E+00	1.79E+01	4.58E+01	4.69E+01	4.92E+01
σ Y (kg)	1.03E-03	3.09E-03	1.25E-02	2.50E-02	5.01E-02	1.28E-01	1.31E-01	1.38E-01
PE (kg)	0	0	0	0	0	0	0	0
MUF (kg)	1.85E-03	5.54E-03	2.25E-02	4.50E-02	8.99E-02	2.30E-01	2.36E-01	2.47E-01
σ MUF (kg)	1.46E-03	4.38E-03	1.77E-02	3.55E-02	7.10E-02	1.82E-01	1.86E-01	1.95E-01
3 σ MUF (kg)	4.38E-03	1.31E-02	5.32E-02	1.06E-01	2.13E-01	5.46E-01	5.59E-01	5.86E-01

3.2.3. Analysis of holdup and waste

An analysis was conducted regarding the SNM buildup due to holdup in pipes and waste streams emanating from the MSR, specifically from the reprocessing and fuel reconstituting operations. This analysis is necessary to determine the point at which the amount of SNM in holdup or waste exceeds the SQ limits set forth by the IAEA. At that point, the MSR would need to be shut down and cleaned.

Calculations for the accumulation of holdup and waste for ^{235}U are shown below in Table 5. The analysis showed that the $3\sigma_{\text{MUF}} < \text{SQ}$ condition would not be met beyond 120 months. However, the holdup and waste at that point would still be within the IAEA limit of 75 kg ^{235}U . This result shows that the accumulation of ^{235}U in holdup and waste is not a significant factor in the safeguarding of the MSR; rather, the safeguards analysis is dominated by SNM buildup due to reactor operation.

Table 5. ^{235}U buildup in holdup and waste.

MBP	1 month	3 months	12 months	24 months	48 months	123 months
PB (kg)	0	0	0	0	0	0
X (kg)	5.09E+01	1.53E+02	6.19E+02	1.24E+03	2.48E+03	6.34E+03
σ X (kg)	1.42E-01	4.27E-01	1.73E+00	3.47E+00	6.93E+00	1.78E+01
Y (kg)	5.06E+01	1.52E+02	6.16E+02	1.23E+03	2.46E+03	6.31E+03
Holdup (kg)	1.53E-04	4.58E-04	1.86E-03	3.71E-03	7.43E-03	1.90E-02
Waste (kg)	2.54E-01	7.63E-01	3.10E+00	6.19E+00	1.24E+01	3.17E+01
σ Y (kg)	1.42E-01	4.25E-01	1.72E+00	3.45E+00	6.90E+00	1.77E+01
PE (kg)	0	0	0	0	0	0
MUF (kg)	2.55E-01	7.64E-01	3.10E+00	6.19E+00	1.24E+01	3.17E+01
σ MUF (kg)	2.01E-01	6.03E-01	2.44E+00	4.89E+00	9.78E+00	2.51E+01
3σ MUF (kg)	6.03E-01	1.81E+00	7.33E+00	1.47E+01	2.93E+01	7.52E+01

Similar results were obtained for the buildup of plutonium in holdup and waste. The $3\sigma_{\text{MUF}} < \text{SQ}$ would not be met beyond 123 months; the amount of plutonium in holdup

and waste would still be within the IAEA limit of 8 kg plutonium at that point. These results are summarized below in Table 6.

Table 6. Plutonium buildup in holdup and waste.

MBP	1 month	3 months	12 months	24 months	48 months	123 months	126 months
PB (kg)	0	0	0	0	0	0	0
X (kg)	5.38E+00	1.61E+01	6.54E+01	1.31E+02	2.62E+02	6.71E+02	6.87E+02
σ X (kg)	1.51E-02	4.52E-02	1.83E-01	3.66E-01	7.33E-01	1.88E+00	1.92E+00
Y (kg)	5.35E+00	1.61E+01	6.51E+01	1.30E+02	2.60E+02	6.67E+02	6.84E+02
Holdup (kg)	1.61E-05	4.84E-05	1.96E-04	3.93E-04	7.85E-04	2.01E-03	2.06E-03
Waste (kg)	2.69E-02	8.07E-02	3.27E-01	6.54E-01	1.31E+00	3.35E+00	3.44E+00
σ Y (kg)	1.50E-02	4.49E-02	1.82E-01	3.65E-01	7.29E-01	1.87E+00	1.91E+00
PE (kg)	0	0	0	0	0	0	0
MUF (kg)	2.69E-02	8.07E-02	3.27E-01	6.55E-01	1.31E+00	3.35E+00	3.44E+00
σ MUF (kg)	2.12E-02	6.37E-02	2.58E-01	5.17E-01	1.03E+00	2.65E+00	2.71E+00
3 σ MUF (kg)	6.37E-02	1.91E-01	7.75E-01	1.55E+00	3.10E+00	7.95E+00	8.14E+00

3.2.4. Analysis of SNM in all three MSR loops

Sections 3.2.1., 3.2.2., and 3.2.3. detail the results of the NMA analysis for a consideration of a single loop within the MSR. Given that the MSR has three loops, it was necessary to consider the total SNM within all three loops.

MUF calculations were performed for ^{235}U in the fuel-salt for all three loops of the reprocessing MBA. Table 7 shows the results ^{235}U in the fuel-salt for fuel cycle 1.

Once the MUF values were calculated for all three loops, it could be determined if the MSR unit as a whole would meet the IAEA conditions. A summary of these results is shown below in Table 8 for ^{235}U in the fuel-salt. It can be seen that all of the IAEA conditions are met for ^{235}U in the fuel-salt for the MBPs of 1 month, 3 months, and 12 months.

Table 7. ²³⁵U fuel-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months
PB (kg)	0	0	0
X (kg)	1.53E+02	4.58E+02	1.86E+03
σ X (kg)	4.27E-01	1.28E+00	5.20E+00
Y (kg)	1.52E+02	4.56E+02	1.85E+03
σ Y (kg)	4.25E-01	1.28E+00	5.17E+00
Holdup (kg)	0	0	0
Waste (kg)	7.63E-01	2.29E+00	9.29E+00
PE (kg)	0	0	0
MUF (kg)	7.64E-01	2.29E+00	9.29E+00
σ MUF (kg)	6.03E-01	1.81E+00	7.33E+00
3σ MUF (kg)	1.81E+00	5.43E+00	2.20E+01

Table 8. Summary of ²³⁵U fuel-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < $3\sigma_{\text{MUF}}$	3σ MUF < SQ
1 month	7.64E-01	1.81E+00	MET	MET	MET
3 months	2.29E+00	5.43E+00	MET	MET	MET
12 months	9.29E+00	2.20E+01	MET	MET	MET

This same process was repeated for Pu content in the fuel-salt for all three loops of the reprocessing MBA for fuel cycle 1. These results can be seen in Table 9. For the designated MBPs, all IAEA conditions were met for Pu in the fuel-salt, as seen in Table 10.

Table 9. Pu fuel-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months
PB (kg)	0	0	0
X (kg)	1.61E+01	4.84E+01	1.96E+02
σ X (kg)	4.52E-02	1.36E-01	5.50E-01
Y (kg)	1.61E+01	4.82E+01	1.95E+02
σ Y (kg)	4.49E-02	1.35E-01	5.47E-01
Holdup (kg)	0	0	0
Waste (kg)	8.07E-02	2.42E-01	9.81E-01
PE (kg)	0	0	0
MUF (kg)	8.07E-02	2.42E-01	9.82E-01
σ MUF (kg)	6.37E-02	1.91E-01	7.75E-01
3σ MUF (kg)	1.91E-01	5.73E-01	2.33E+00

Table 10. Summary of Pu fuel-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < $3\sigma_{\text{MUF}}$	3σ MUF < SQ
1 month	8.07E-02	1.91E-01	MET	MET	MET
3 months	2.42E-01	5.73E-01	MET	MET	MET
12 months	9.82E-01	2.33E+00	MET	MET	MET

The detailed MUF calculations for ^{235}U in the blanket-salt can be found in APPENDIX. Table 11 shows that the IAEA conditions for ^{235}U in the blanket-salt were met for the proposed MBPs of 1 month, 3 months, and 12 months.

Table 11. Summary of ^{235}U blanket-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < $3\sigma_{\text{MUF}}$	3σ MUF < SQ
1 month	1.45E-02	3.45E-02	MET	MET	MET
3 months	4.36E-02	1.03E-01	MET	MET	MET
12 months	1.77E-01	4.19E-01	MET	MET	MET

Pu MUF calculations for all three loops of the blanket-salt can be found in APPENDIX. The summary of these results can be seen in Table 12, which shows that IAEA conditions for Pu in the blanket-salt are satisfied.

Table 12. Summary of Pu blanket-salt NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	5.54E-03	1.31E-02	MET	MET	MET
3 months	1.66E-02	3.94E-02	MET	MET	MET
12 months	6.74E-02	1.60E-01	MET	MET	MET

An analysis was conducted for ²³⁵U in holdup and waste for all three loops of the reprocessing MBA for fuel cycle 1; detailed results of the NMA calculations are shown in APPENDIX. The summary of these results, shown in Table 13 shows that IAEA conditions are met for ²³⁵U in holdup and waste.

Table 13. Summary of ²³⁵U holdup and waste NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	7.64E-01	1.81E+00	MET	MET	MET
3 months	2.29E+00	5.43E+00	MET	MET	MET
12 months	9.29E+00	2.20E+01	MET	MET	MET

Finally, Pu content in holdup and waste was calculated, as seen in APPENDIX. The summary of these calculations can be seen below in Table 14. IAEA conditions for Pu were met for the proposed MBPs.

Table 14. Summary of Pu holdup and waste NMA calculations all three loops of the reprocessing MBA for fuel cycle 1 as compared to IAEA conditions.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	8.07E-02	1.91E-01	MET	MET	MET
3 months	2.42E-01	5.73E-01	MET	MET	MET
12 months	9.82E-01	2.33E+00	MET	MET	MET

3.2.5. MBA calculations for a multi-module MSR site

As discussed in Section 2.3.4., given that MSRs may be deployed as SMRs in a multi-module facility, the NMA analysis described in Section 3.2.4. was applied to a facility with four MSRs.

Table 15 and Table 16 show the summary of ²³⁵U and Pu content in the fuel-salt reprocessing MBA, respectively. For a MSR site with four MSRs, the IAEA conditions would not be met for the 12-month MBP.

Table 15. Summary of ²³⁵U fuel-salt reprocessing NMA calculations when considering four MSRs at one site.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	3.05E+00	7.23E+00	MET	MET	MET
3 months	9.16E+00	2.17E+01	MET	MET	MET
12 months	3.72E+01	8.80E+01	MET	MET	NOT MET

Table 16. Summary of Pu fuel-salt reprocessing NMA calculations when considering four MSRs at one site.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	3.23E-01	7.65E-01	MET	MET	MET
3 months	9.68E-01	2.29E+00	MET	MET	MET
12 months	3.93E+00	9.30E+00	MET	MET	NOT MET

Results for the ^{235}U and Pu content in the blanket-salt for a four-MSR site can be found in APPENDIX. All IAEA conditions were met for the MBPs of 1 months, 3 months, and 12 months for both ^{235}U and Pu in the blanket-salt.

Finally, an analysis of ^{235}U and Pu accumulation in holdup and waste was considered for a single site with four MSRs. For both ^{235}U and Pu, the IAEA condition of $3\sigma_{\text{MUF}} < \text{SQ}$ would not be met for the 12-month MBP. These results are summarized below in Table 22 and Table 18.

Table 17. Summary of ^{235}U holdup and waste NMA calculations when considering four MSRs at one site.

MBP	MUF (kg)	$3\sigma_{\text{MUF}}$ (kg)	IAEA Conditions		
			MUF < SQ	MUF < $3\sigma_{\text{MUF}}$	$3\sigma_{\text{MUF}}$ < SQ
1 month	3.05E+00	7.23E+00	MET	MET	MET
3 months	9.16E+00	2.17E+01	MET	MET	MET
12 months	3.72E+01	8.80E+01	MET	MET	NOT MET

Table 18. Summary of Pu holdup and waste NMA calculations when considering four MSRs at one site.

MBP	MUF (kg)	$3\sigma_{\text{MUF}}$ (kg)	IAEA Conditions		
			MUF < SQ	MUF < $3\sigma_{\text{MUF}}$	$3\sigma_{\text{MUF}}$ < SQ
1 month	3.23E-01	7.65E-01	MET	MET	MET
3 months	9.68E-01	2.29E+00	MET	MET	MET
12 months	3.93E+00	9.30E+00	MET	MET	NOT MET

4. CONCLUSIONS

4.1. Conclusions

This study was carried out to demonstrate that both MCNP and SCALE could be used to model an MSR in developing nuclear safeguards strategies for a core design developed at Texas A&M University. The MCNP portion of this work was most effective. SCALE was used to successfully model the MSR, however, refueling of the fuel-salt was not possible in the given time frame of the project. However, neutronic parametric results, from SCALE and MCNP, including neutron multiplication factor and burnup for the first fuel cycle, were compared to verify efficacy of the MSR core model and was found to be satisfactory. Further work is required to refuel the MSR in SCALE and to analyze and develop effective NMA strategies. Results of the MCNP fuel burnup simulations were used to determine SNM quantities over time in each of the three designated MBAs for the MSR by utilizing HKED as an SNM assay methodology at the KMPs. A combined (systematic and random) measurement uncertainty of 0.28% provided for HKED by the IAEA ITV document was assumed.

The MBA analysis for the core reprocessing loop showed that the MUF value for ^{235}U was well within the SQ value of 75 kg for the MBP of three months (SNM in the reprocessing loop). Thus, the MBP is adequate for an effective NMA scheme for MBA-2 as the MUF value is within the IAEA requirements $\text{MUF} < \text{SQ}$, $\text{MUF} < 3\sigma_{\text{MUF}}$, and $3\sigma_{\text{MUF}} < \text{SQ}$. Analysis of the Pu in the core reprocessing MBA found that, for an MBP of 30 days, the MUF value for Pu was within the IAEA SQ value for Pu of 8 kg.

The safeguards analyses were applied to the reprocessing cycle of the blanket loop; the blanket was separated into three loops. The results demonstrated that the reprocessing cycle of the blanket loop will be within IAEA limits of ^{235}U for at least 132 months. MBA calculations for plutonium show that the reprocessing cycle of the blanket loop will be within IAEA limits of plutonium for at least 132 months.

An analysis of SNM buildup in the holdup and waste of the MSR was conducted. This analysis was necessary to determine the point at which the amount of SNM in holdup or waste exceeds the SQ limits set forth by the IAEA. At that point, the MSR would need to be shut down and cleaned. The analysis showed that the $3\sigma_{\text{MUF}} < \text{SQ}$ condition would not be met beyond 120 months. However, the holdup and waste at that point would still be within the IAEA limit of 75 kg ^{235}U . This result showed that the accumulation of ^{235}U in holdup and waste is not a significant factor in the safeguarding of the MSR; rather, the safeguards analysis is dominated by SNM buildup due to reactor operation. Calculations for the buildup of plutonium in holdup and waste gave similar results. The $3\sigma_{\text{MUF}} < \text{SQ}$ would not be met beyond 123 months; the amount of plutonium in holdup and waste would still be within the IAEA limit of 8 kg plutonium at that point.

This analysis was expanded to consider the MSR as a whole, with MUF calculations from all three loops combined into a single value. Results showed that the MSR design would be within the required IAEA conditions for the designated MBPs.

Given that some MSR designs can be considered SMRs, it was necessary to consider a safeguards approach for a multi-module MSR site. For analysis purposes, a

four-module site was considered. Results from this analysis showed that the $3\sigma_{\text{MUF}} < \text{SQ}$ would not be met for the 12-month MBP.

4.2. Significance of conclusions

This thesis can contribute to the development of MSR safeguards. This work is necessary due to the increasing international development of MSRs and the associated proliferation concerns. In particular, due to the low economic cost of SMR designs and the difficulty in safeguarding a multi-module site where small amounts of SNM may be taken from multiple modules. The safeguards field of study is lacking MC&A analyses for detection techniques for molten salt. This work was done to demonstrate that both MCNP and SCALE could be used to model an MSR in developing NMA strategies for a molten salt reactor core design developed at Texas A&M University. Results of the fuel-salt depletion were used to determine that the proposed MBPs meet the three IAEA safeguards compliance conditions ($\text{MUF} < \text{SQ}$, $\text{MUF} < 3\sigma_{\text{MUF}}$, and $3\sigma_{\text{MUF}} < \text{SQ}$) for both ^{235}U and plutonium.

4.3. Future work

Future work should be done with respect to the fuel depletion in SCALE. Once results are obtained, they can be compared to those obtained from the MCNP analysis. This will contribute to a more comprehensive safeguards approach.

SCALE was used to successfully model the MSR, however, refueling of the fuel-salt was not possible in the given time frame of the project. Neutronic parametric results from SCALE and MCNP were compared to verify efficacy of the MSR core model and was found to be satisfactory. Further work is required to refuel the MSR in SCALE and

to analyze and develop effective NMA strategies. Implementation of the ORNL-developed ChemTriton Python script is the best course of action for the most realistic modeling of an MSR for neutron transport and depletion calculations [38].

Further work should also be done to analyze the efficacy of HKED as the technology continues to develop for application to MSR fuel-salt. It would be a useful endeavor to experimentally verify that HKED can measure samples of densities similar to molten salt; an experiment measuring molten salt using HKED would be most beneficial to demonstrating it and to determine whether it can be, in fact, be used for MSR safeguards purposes.

REFERENCES

- [1] M. W. Rosenthal, P. R. Kasten and R. B. Briggs, “Molten-Salt Reactors – History, Status, and Potential,” Oak Ridge National Laboratory, 1969.
- [2] C. Forsberg, “Safety and Licensing Aspects of the Molten Salt Reactor,” in *2004 American Nuclear Society Annual Meeting*, 2004.
- [3] D. LeBlanc, “Molten salt reactors: A new beginning for an old idea,” *Nuclear Engineering and Design*, 2009.
- [4] U.S. DOE, Office of Nuclear Energy, “3 Advanced Reactor Systems to Watch by 2030,” 7 March 2018. [Online]. Available: <https://www.energy.gov/ne/articles/3-advanced-reactor-systems-watch-2030>.
- [5] D. N. Kovacic, L. G. Worrall, A. Worrall, G. F. Flanagan, D. E. Holcomb, R. Bari, L. Cheng, D. Farley and M. Sternat, “Safeguards Challenges for Molten Salt Reactors,” 2018.
- [6] G. Flanagan, “Fuel Cycle and Safeguards - Presentation on Molten Salt Reactor Technology,” 2017.
- [7] International Atomic Energy Agency, “Treaty on the Non-Proliferation of Nuclear Weapons (NPT),” 1968.
- [8] International Atomic Energy Agency, “The Structure and Content of Agreements Between the Agency and States Required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons (INFCIRC/153),” 1972.
- [9] International Atomic Energy Agency, “Model Protocol Additional to the Agreement(s) Between State(s) and the International Atomic Energy Agency for the Application of Safeguards (INFCIRC/540),” 1997.
- [10] International Atomic Energy Agency, “IAEA Safeguards Glossary 2001 Edition,” 2002.
- [11] K. Mummah, D. I. Poston, M. A. Griffin, R. M. Bahran and C. J. Verschuren, “Integrating Nonproliferation and Safeguards Concepts into Reactor Lab Courses,” 2017.
- [12] ANTECH, “Fork Detectors (EURATOM) B2104 Series,” 2021. [Online]. Available: <https://www.antech-inc.com/products/b2104/>.

- [13] J. B. Coble, S. E. Skutnik, S. N. Gilliam and M. P. Cooper, “Review of Candidate Techniques for Material Accountancy Measurements in Electrochemical Separations Facilities,” *Nuclear Technology*, vol. 206, no. 12, pp. 1803-1826, 2020.
- [14] International Atomic Energy Agency, “Small modular reactors,” 2021. [Online]. Available: <https://www.iaea.org/topics/small-modular-reactors>.
- [15] NuScale Power, “Technology Overview: How the NuScale Module Works,” 2021. [Online]. Available: <https://www.nuscalepower.com/technology/technology-overview>.
- [16] Small Modular Reactors Regulators' Forum, “Report on Multi-unit/Multi-module aspects specific to SMRs,” 2019.
- [17] World Nuclear Association, “Molten Salt Reactors,” December 2020. [Online]. Available: <https://www.world-nuclear.org/information-library/current-and-future-generation/molten-salt-reactors.aspx>.
- [18] L. G. Worrall, A. Worrall, G. F. Flanagan, S. Croft, A. M. Krichinsky, C. A. Pickett, R. D. McElroay Jr., S. L. Cleveland, D. N. Kovacic, J. M. Whitaker and J. L. White-Horton, “Safeguards Considerations for Thorium Fuel Cycles,” *Nuclear Technology*, vol. 194, no. 2, pp. 281-293, 2016.
- [19] F. Barnaby and S. Burnie, “Planning for Failure – International Nuclear Safeguards and the Rokkasho-Mura Reprocessing Plant,” 2002.
- [20] C. H. Yamaguchi, G. L. Stefani and T. A. Santos, “A general overview of generation IV molten salt reactor (MSR) and the use of thorium as fuel,” in *2017 International Nuclear Atlantic Conference - INAC 2017*, 2017.
- [21] International Atomic Energy Agency, “International Safeguards in Nuclear Facility Design and Construction,” 2013.
- [22] International Atomic Energy Agency, “International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials,” 2010.
- [23] R. Bean, “Aqueous processing material accountability instrumentation.” Idaho National Laboratory, Idaho National Laboratory, 2007.
- [24] R. J. Bellas, G. F. Flanagan, D. E. Holcomb and W. P. Poore, “A Safety and Licensing Roadmap to Identify the Research and Development Gaps of Commercial Molten Salt Reactors,” Oak Ridge National Laboratory, 2018.

- [25] R. J. Bellas and G. F. Flanagan, “Regulatory Gap Analysis of Select NUREG-0800 Chapters for Applicability to Molten Salt Reactors,” Oak Ridge National Laboratory, 2018.
- [26] T. F. Guzzardo, R. D. McElroy, S. Croft, J. Garrison, R. Venkataraman and C. A. Pickett, “Stability of Working Reference Standards for Hybrid K-Edge Densitometer Quality Assurance,” 2014.
- [27] M. T. Cook, “Hybrid K-edge Densitometry as a Method for Materials Accountancy Measurements in Pyrochemical Reprocessing,” University of Tennessee, 2015.
- [28] G. S. Mickum, “Development of a dedicated hybrid K-edge densitometer for pyroprocessing safeguards measurements using Monte Carlo simulation models,” Georgia Institute of Technology, 2015.
- [29] J. Werner, “MCNP User’s Manual – Code Version 6.2,” Los Alamos National Laboratory, 2017.
- [30] B. T. Readen and M. A. Jessee, “SCALE Code System – Version 6.2,” Oak Ridge National Laboratory, 2016.
- [31] M. Brovchenko, J. Kloosterman, L. Luzzi, E. Merle, D. Heuer, A. Laureau, O. Feynberg, V. Ignatiev, M. Aufiero, A. Cammi, C. Fiorina, F. Alcaro, S. Dulla, P. Ravetto, L. Frima, D. Lathouwers and B. Merk, “Neutronic benchmark of the molten salt fast reactor in the frame of the EVOL and MARS collaborative projects,” *EPJ Nuclear Sciences & Technology*, vol. 5, no. 2, 2019.
- [32] European Research and Safeguards Development Association, “Bulletin No. 48,” 2012.
- [33] A. Gavron and S. T. Hsue, “Application Note: KED/KXRF Hybrid Densitometer,” Los Alamos National Laboratory, 1996.
- [34] Oak Ridge National Laboratory, “Hybrid K-Edge Densitometer Measurements at ORNL: HKED Calibration and Quality Control Standards,” in *NDA User's Group Meeting*, 2014.
- [35] Mirion Technologies, “Hybrid K-edge/XRF Analyzer,” 2021. [Online]. Available: <https://www.mirion.com/products/hybrid-k-edge-xrf-analyzer-special-application-systems>.
- [36] R. D. McElroy Jr., “Performance Evaluation of the ORNL Multi-Elemental KED (MKED) Analysis Algorithms,” Oak Ridge National Laboratory, 2018.

- [37] S. J. Johnson, R. Abedin-Zadeh, C. Pearsall, K. Hiruta, C. Creusot, M. Ehinger, E. Kuhn, B. Chesnay, N. Robson, H. Higuchi, S. Takeda, K. Fujimaki, H. Ai, S. Uehara, H. Amano and K. Hoshi, "Development of the safeguards approach for the Rokkasho reprocessing plant," International Atomic Energy Agency, 2001.
- [38] B. R. Betzler, J. J. Powers and A. Worrall, "Molten Salt Neutronics and Fuel Cycle Modeling and Simulation with SCALE," *Annals of Nuclear Energy*, vol. 101, 2017.

APPENDIX

Summary of MUF Calculations

Table 19. ²³⁵U blanket-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months	24 months	48 months	123 months	126 months	132 months
PB (kg)	0	0	0	0	0	0	0	0
X (kg)	2.91E+00	8.72E+00	3.54E+01	7.08E+01	1.42E+02	3.63E+02	3.72E+02	3.89E+02
σ X (kg)	8.14E-03	2.44E-02	9.91E-02	1.98E-01	3.96E-01	1.02E+00	1.04E+00	1.09E+00
Y (kg)	2.89E+00	8.68E+00	3.52E+01	7.04E+01	1.41E+02	3.61E+02	3.70E+02	3.87E+02
σ Y (kg)	8.10E-03	2.43E-02	9.86E-02	1.97E-01	3.94E-01	1.01E+00	1.04E+00	1.08E+00
PE (kg)	0	0	0	0	0	0	0	0
MUF (kg)	1.45E-02	4.36E-02	1.77E-01	3.54E-01	7.08E-01	1.81E+00	1.86E+00	1.95E+00
σ MUF (kg)	1.15E-02	3.45E-02	1.40E-01	2.80E-01	5.59E-01	1.43E+00	1.47E+00	1.54E+00
3 σ MUF (kg)	3.45E-02	1.03E-01	4.19E-01	8.39E-01	1.68E+00	4.30E+00	4.40E+00	4.61E+00

Table 20. Pu blanket-salt MUF calculations for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months	24 months	48 months	123 months	126 months	132 months
PB (kg)	0	0	0	0	0	0	0	0
X (kg)	1.11E+00	3.32E+00	1.35E+01	2.70E+01	5.39E+01	1.38E+02	1.42E+02	1.48E+02
σ X (kg)	3.10E-03	9.31E-03	3.77E-02	7.55E-02	1.51E-01	3.87E-01	3.96E-01	4.15E-01
Y (kg)	1.10E+00	3.31E+00	1.34E+01	2.68E+01	5.36E+01	1.37E+02	1.41E+02	1.48E+02
σ Y (kg)	3.09E-03	9.26E-03	3.76E-02	7.51E-02	1.50E-01	3.85E-01	3.94E-01	4.13E-01
PE (kg)	0	0	0	0	0	0	0	0
MUF (kg)	5.54E-03	1.66E-02	6.74E-02	1.35E-01	2.70E-01	6.91E-01	7.08E-01	7.42E-01
σ MUF (kg)	4.38E-03	1.31E-02	5.32E-02	1.06E-01	2.13E-01	5.46E-01	5.59E-01	5.86E-01
3 σ MUF (kg)	1.31E-02	3.94E-02	1.60E-01	3.19E-01	6.39E-01	1.64E+00	1.68E+00	1.76E+00

Table 21. ²³⁵U fuel-salt MUF calculations for holdup and waste for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months	24 months	48 months	123 months
PB (kg)	0	0	0	0	0	0
X (kg)	1.53E+02	4.58E+02	1.86E+03	3.71E+03	7.43E+03	1.90E+04
σ X (kg)	4.27E-01	1.28E+00	5.20E+00	1.04E+01	2.08E+01	5.33E+01
Y (kg)	1.52E+02	4.56E+02	1.85E+03	3.70E+03	7.39E+03	1.89E+04
Holdup (kg)	4.58E-04	1.37E-03	5.57E-03	1.11E-02	2.23E-02	5.71E-02
Waste (kg)	7.63E-01	2.29E+00	9.29E+00	1.86E+01	3.71E+01	9.52E+01
σ Y (kg)	4.25E-01	1.28E+00	5.17E+00	1.03E+01	2.07E+01	5.30E+01
PE (kg)	0	0	0	0	0	0
MUF (kg)	7.64E-01	2.29E+00	9.29E+00	1.86E+01	3.72E+01	9.52E+01
σ MUF (kg)	6.03E-01	1.81E+00	7.33E+00	1.47E+01	2.93E+01	7.52E+01
3 σ MUF (kg)	1.81E+00	5.43E+00	2.20E+01	4.40E+01	8.80E+01	2.26E+02

Table 22. Pu fuel-salt MUF calculations for holdup and waste for all three loops of the reprocessing MBA for fuel cycle 1.

MBP	1 month	3 months	12 months	24 months	48 months	123 months
PB (kg)	0	0	0	0	0	0
X (kg)	1.61E+01	4.84E+01	1.96E+02	3.93E+02	7.85E+02	2.01E+03
σ X (kg)	4.52E-02	1.36E-01	5.50E-01	1.10E+00	2.20E+00	5.63E+00
Y (kg)	1.61E+01	4.82E+01	1.95E+02	3.91E+02	7.81E+02	2.00E+03
Holdup (kg)	4.84E-05	1.45E-04	5.89E-04	1.18E-03	2.36E-03	6.03E-03
Waste (kg)	8.07E-02	2.42E-01	9.81E-01	1.96E+00	3.93E+00	1.01E+01
σ Y (kg)	4.49E-02	1.35E-01	5.47E-01	1.09E+00	2.19E+00	5.60E+00
PE (kg)	0	0	0	0	0	0
MUF (kg)	8.07E-02	2.42E-01	9.82E-01	1.96E+00	3.93E+00	1.01E+01
σ MUF (kg)	6.37E-02	1.91E-01	7.75E-01	1.55E+00	3.10E+00	7.95E+00
3 σ MUF (kg)	1.91E-01	5.73E-01	2.33E+00	4.65E+00	9.30E+00	2.38E+01

Table 23. Summary of ²³⁵U blanket-salt reprocessing NMA calculations when considering four MSR at one site.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ _{MUF}	3 σ MUF < SQ
1 month	5.82E-02	1.38E-01	MET	MET	MET
3 months	1.75E-01	4.14E-01	MET	MET	MET
12 months	7.08E-01	1.68E+00	MET	MET	MET

Table 24. Summary of Pu blanket-salt reprocessing NMA calculations when considering four MSRs at one site.

MBP	MUF (kg)	3 σ MUF (kg)	IAEA Conditions		
			MUF < SQ	MUF < 3 σ_{MUF}	3 σ MUF < SQ
1 month	2.22E-02	5.25E-02	MET	MET	MET
3 months	6.65E-02	1.58E-01	MET	MET	MET
12 months	2.70E-01	6.39E-01	MET	MET	MET