

# **Feasibility Study of Fresh Fuel Feed Monitoring for Liquid-Fueled Molten Salt Reactor Safeguards**

A Dissertation Presented for the  
Doctor of Philosophy  
Degree  
The University of Tennessee, Knoxville

Karen Koop Hogue

December 2023

© by Karen Koop Hogue, 2023  
All Rights Reserved.

*To my extraordinary trio:*  
*Whatever your hand finds to do, do it with all your might . . .*  
*—Ecclesiastes 9:10*

# Acknowledgments

This research was performed under appointment to the Nuclear Nonproliferation International Safeguards Fellowship Program sponsored by the US Department of Energy, National Nuclear Security Administration's Office of International Nuclear Safeguards (NA-241).

I would like to thank my committee for their support: Dr. Jason Hayward, Dr. Jamie Coble, Dr. Louise Evans, Dr. Ondrej Chvala, and Dr. Steve Skutnik.

I am grateful for the tremendous support of colleagues who have provided input and guidance: Bob McElroy, Ram Venkataraman, and Susan Smith for their input on radiation detection; Richard Reed, Mark Adams, Jason Hite, Greg Westphal, Scott Stewart, Birdy Phathanapirom, and Luke Wagner for their support with my many coding questions; David Holcomb, Jeff Latkowski, Kevin Robb, and Joanna McFarlane for sharing their knowledge on molten salt reactors; and Jill Cooley, Bruce Moran, and George Anzelon for instilling my passion for international safeguards.

Most importantly, thank you to my husband for the gift of time, favorite sister (Jennifer) and parents for their unconditional love, kids for the best hugs in the world, and wonderful friends for supporting me and encouraging me to do hard things. Per request of my children, I'd also like to acknowledge our pup, Pepper, for all the smiles and laughs she's brought.

# Abstract

Liquid-fueled molten salt reactors (MSRs) are an advanced reactor design concept with material accountancy challenges due to continuously circulating fuel in bulk form with online additions of fresh fuel. With more than 24 distinct liquid-fueled MSR designs being pursued by companies around the world, facility operators and the International Atomic Energy Agency need approaches and technologies to quantify nuclear material within MSR facilities for safeguards and security purposes. Safeguards technical objectives were defined across a prospective liquid-fueled MSR. One high-priority objective for facility operators and the IAEA will likely be quantifying nuclear material in fresh initial salt (i.e., salt added to the MSR at the beginning of life) and makeup salt additions to the reactor system (i.e., salt added over time while the MSR is operational), which was selected for further analysis. Computational simulations combining fresh fuel source terms from SCALE/ORIGEN into MCNP models were run for different MSR design parameters as well as different gamma- and neutron-based measurement techniques to determine the expected impact of each parameter on the feasibility of radiation monitoring on the outside of fresh fuel salt in piping for material accountancy. MSR design parameters analyzed were a fluoride-based and chloride-based salt, four uranium enrichments, two pipe materials, several pipe outer diameters and thicknesses, and the optional presence of insulation and aluminum jacketing external to the piping. Measurement modalities including NaI and high-purity germanium gamma detector systems, total passive neutron counting in a moderated  $^3\text{He}$  collar, coincident neutron counting in a

moderated  $^3\text{He}$  collar, and coincident neutron counting in in a moderated  $^3\text{He}$  collar with an interrogating neutron source were assessed for feasibility.

Although no measurement technique worked well for both salt types and all design parameters assessed, passive gamma spectrometry and total passive neutron counting in a moderated neutron detector collar are promising, especially for salts with high-assay low-enriched uranium salt and larger diameters. Active neutron interrogation with a neutron coincidence collar counting thermal neutrons in  $^3\text{He}$  exhibited high relative uncertainties due to high accidental coincidences. A neutron coincidence collar using liquid scintillators counting fast neutrons will likely be a promising alternative for quantifying  $^{235}\text{U}$  within unirradiated, uranium-bearing fuel salts within piping because of the expected lower accidental coincidence rate.

# Table of Contents

|          |                                                                     |           |
|----------|---------------------------------------------------------------------|-----------|
| <b>1</b> | <b>Introduction</b>                                                 | <b>1</b>  |
| 1.1      | Molten Salt Reactors . . . . .                                      | 3         |
| 1.2      | Safeguards and Security . . . . .                                   | 8         |
| 1.3      | Original Contributions . . . . .                                    | 10        |
| <b>2</b> | <b>MSR Facility-Level Safeguards Objectives</b>                     | <b>12</b> |
| 2.1      | Safeguards-Relevant Design Features of MSRs . . . . .               | 15        |
| 2.2      | Safeguards Objectives and Prospective MC&A Plan . . . . .           | 15        |
| <b>3</b> | <b>Liquid-Fueled MSR Fresh Fuel Monitoring</b>                      | <b>22</b> |
| <b>4</b> | <b>Simulated Radiation Signatures from Fresh Fuel Salt</b>          | <b>29</b> |
| 4.1      | Modes of Decay . . . . .                                            | 31        |
| 4.2      | Modeling Passive Radiation Emissions from Fresh Fuel Salt . . . . . | 35        |
| 4.2.1    | SCALE/ORIGEN Passive Neutron Emission Results . . . . .             | 36        |
| 4.2.2    | SCALE/ORIGEN Gamma-Ray Emission Results . . . . .                   | 37        |
| <b>5</b> | <b>Radiation Signature Modeling-Methodology</b>                     | <b>45</b> |
| 5.1      | Modeling Assumptions . . . . .                                      | 50        |
| 5.2      | Model Parameters Evaluated . . . . .                                | 50        |
| 5.2.1    | Salt Composition . . . . .                                          | 51        |
| 5.2.2    | Pipe Material . . . . .                                             | 52        |
| 5.2.3    | Pipe Dimensions . . . . .                                           | 52        |

|          |                                                                  |            |
|----------|------------------------------------------------------------------|------------|
| 5.2.4    | Insulation . . . . .                                             | 54         |
| 5.2.5    | Detectors . . . . .                                              | 54         |
| 5.2.6    | Source Definitions . . . . .                                     | 64         |
| <b>6</b> | <b>Radiation Signatures Modeling—Results</b>                     | <b>65</b>  |
| 6.1      | Figures of Merit . . . . .                                       | 67         |
| 6.2      | Gamma Detection Results . . . . .                                | 68         |
| 6.2.1    | Impact of Chemical Composition of the Salt . . . . .             | 70         |
| 6.2.2    | Impact of Uranium Enrichment . . . . .                           | 74         |
| 6.2.3    | Impact of Pipe Material . . . . .                                | 77         |
| 6.2.4    | Impact of Pipe Outer Diameter . . . . .                          | 77         |
| 6.2.5    | Impact of Pipe Thickness . . . . .                               | 77         |
| 6.2.6    | Impact of Insulation . . . . .                                   | 81         |
| 6.3      | Neutron Detection . . . . .                                      | 85         |
| 6.3.1    | Passive Neutron Detection with $^3\text{He}$ Results . . . . .   | 91         |
| 6.3.2    | Active Neutron Detection with $^3\text{He}$ Results . . . . .    | 97         |
| <b>7</b> | <b>Feed Monitoring System Options</b>                            | <b>109</b> |
| 7.1      | Assessment of Radiation Monitoring Measurement Methods . . . . . | 109        |
| <b>8</b> | <b>Conclusions</b>                                               | <b>116</b> |
| 8.1      | Future Work . . . . .                                            | 120        |
|          | <b>Bibliography</b>                                              | <b>122</b> |
|          | <b>Appendix</b>                                                  | <b>135</b> |
| <b>A</b> | <b>SCALE/ORIGEN Input Files</b>                                  | <b>137</b> |
| A.1      | Fluoride-based salt . . . . .                                    | 137        |
| A.2      | Chloride-based salt . . . . .                                    | 143        |
|          | <b>Vita</b>                                                      | <b>149</b> |

# List of Tables

|     |                                                                                                                                                        |    |
|-----|--------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 1.1 | Current MSR design concepts . . . . .                                                                                                                  | 5  |
| 2.1 | Safeguards-relevant design features of MSRs . . . . .                                                                                                  | 17 |
| 2.2 | MC&A technical objectives and elements for a liquid-fueled MSR facility                                                                                | 20 |
| 3.1 | Fuel salts proposed in select current MSR design concepts . . . . .                                                                                    | 26 |
| 4.1 | Salt compositions in select fresh MSR fuel salts . . . . .                                                                                             | 30 |
| 4.2 | Mass fractions of fresh fuel salts . . . . .                                                                                                           | 33 |
| 5.1 | Chemical composition of Hastelloy N and Inconel 625 pipe materials .                                                                                   | 53 |
| 5.2 | Chemical composition of insulation . . . . .                                                                                                           | 55 |
| 6.1 | Comparison of photopeak areas for different chemical compositions .                                                                                    | 72 |
| 6.2 | Mean free paths and infinite thicknesses for 186 keV photons in fluoride-<br>and chloride-based unirradiated uranium-bearing fuel salts . . . . .      | 75 |
| 6.3 | Comparison of photopeak areas for different uranium enrichments . .                                                                                    | 76 |
| 6.4 | Comparison of photopeak areas for different pipe materials . . . . .                                                                                   | 79 |
| 6.5 | Comparison of photopeak areas for different pipe outer diameters . .                                                                                   | 80 |
| 6.6 | Comparison of photopeak areas for different pipe thicknesses . . . . .                                                                                 | 82 |
| 6.7 | Comparison of photopeak areas with and without insulation . . . . .                                                                                    | 83 |
| 6.8 | Singles and doubles rates (and relative uncertainties) for simulated<br>passive neutron counts in $\text{LiF-BeF}_2\text{-UF}_4\text{-Zr}_4$ . . . . . | 87 |

|      |                                                                                                                                                                |     |
|------|----------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| 6.9  | Singles and doubles rates (and relative uncertainties) for simulated passive neutron counts in NaCl–UCl <sub>3</sub> . . . . .                                 | 88  |
| 6.10 | Singles and doubles rates (and relative uncertainties) for simulated active neutron counts in LiF–BeF <sub>2</sub> –UF <sub>4</sub> –Zr <sub>4</sub> . . . . . | 89  |
| 6.11 | Singles and doubles rates (and relative uncertainties) for simulated active neutron counts in NaCl–UCl <sub>3</sub> . . . . .                                  | 90  |
| 6.12 | Comparison of simulated passive $S$ and $D$ for different chemical compositions of salt with all other factors constant . . . . .                              | 92  |
| 6.13 | Comparison of simulated passive $S$ and $D$ for different uranium enrichments with all other factors constant . . . . .                                        | 93  |
| 6.14 | Comparison of simulated passive $S$ and $D$ for different pipe materials with all other factors constant . . . . .                                             | 95  |
| 6.15 | Comparison of simulated passive $S$ and $D$ for different pipe outer diameters with all other factors constant . . . . .                                       | 96  |
| 6.16 | Comparison of simulated passive $S$ and $D$ for different pipe thicknesses with all other factors constant . . . . .                                           | 98  |
| 6.17 | Comparison of simulated passive $S$ and $D$ for pipes with and without insulation . . . . .                                                                    | 99  |
| 6.18 | Comparison of simulated active $S$ and $D$ for different chemical compositions of salt with all other factors constant . . . . .                               | 101 |
| 6.19 | Comparison of simulated active $S$ and $D$ for different uranium enrichments with all other factors constant . . . . .                                         | 102 |
| 6.20 | Comparison of simulated active $S$ and $D$ for different pipe materials with all other factors constant . . . . .                                              | 105 |
| 6.21 | Comparison of simulated active $S$ and $D$ for different pipe outer diameters with all other factors constant . . . . .                                        | 106 |
| 6.22 | Comparison of simulated active $S$ and $D$ for different pipe thicknesses with all other factors constant . . . . .                                            | 107 |

|      |                                                                                                              |     |
|------|--------------------------------------------------------------------------------------------------------------|-----|
| 6.23 | Comparison of simulated active $S$ and $D$ for pipes with and without insulation . . . . .                   | 108 |
| 7.1  | Feasibility of measurement methods analyzed for fresh fuel salt monitoring for safeguards purposes . . . . . | 110 |
| 7.2  | Singles/doubles rates for active neutron with half/full collar . . . . .                                     | 112 |

# List of Figures

|     |                                                                                                                                                                                      |    |
|-----|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 1.1 | Prospective control areas in a liquid-fueled MSR facility . . . . .                                                                                                                  | 2  |
| 1.2 | Nuclear material types seen in MSR design concepts . . . . .                                                                                                                         | 7  |
| 2.1 | Prospective control areas in a liquid-fueled MSR facility . . . . .                                                                                                                  | 19 |
| 4.1 | $(\alpha, n)$ cross sections for low-Z elements in FLiBe-based salt . . . . .                                                                                                        | 34 |
| 4.2 | $(\alpha, n)$ cross sections for low-Z elements in chloride-based salt . . . . .                                                                                                     | 34 |
| 4.3 | Calculated passive total neutron emissions from fluoride- and chloride-based salts over different enrichments using SCALE/ORIGEN . . . . .                                           | 39 |
| 4.4 | Calculated passive total neutron emissions from fluoride- and chloride-based salts over different times since synthesis using SCALE/ORIGEN . . . . .                                 | 40 |
| 4.5 | Comparison of calculated passive total neutron emissions and $(\alpha, n)$ emissions from fluoride- and chloride-based salts with different enrichments using SCALE/ORIGEN . . . . . | 41 |
| 4.6 | Calculated gamma-ray emissions from fluoride- and chloride-based salt over different enrichments from SCALE/ORIGEN . . . . .                                                         | 42 |
| 4.7 | Calculated gamma-ray emissions from fluoride-based salt over different enrichments and times since salt synthesis from SCALE/ORIGEN . . . . .                                        | 43 |
| 4.8 | Calculated gamma-ray emissions from chloride-based salt over different enrichments and times since salt synthesis from SCALE/ORIGEN . . . . .                                        | 44 |
| 5.1 | Flow between different software packages to generate and analyze data for this research . . . . .                                                                                    | 49 |

|     |                                                                                                                                     |     |
|-----|-------------------------------------------------------------------------------------------------------------------------------------|-----|
| 5.2 | NaI and HPGe detectors modeled on the outside of a salt-filled pipe .                                                               | 57  |
| 5.3 | NaI detector modeled outside of a salt-filled pipe surrounded by insulation                                                         | 59  |
| 5.4 | HPGe detector modeled outside of a salt-filled pipe surrounded by<br>insulation . . . . .                                           | 60  |
| 5.5 | Helium-3 collar detector for passive neutron detection . . . . .                                                                    | 61  |
| 5.6 | Helium-3 half-collar detector for active neutron detection . . . . .                                                                | 62  |
| 6.1 | Simulated 186 keV net photopeak areas in HPGe for different salt<br>compositions, uranium enrichments, and pipe materials . . . . . | 71  |
| 6.2 | Simulated counts for different salt chemical compositions in NaI . . .                                                              | 73  |
| 6.3 | Total mass attenuation coefficients for molten salts . . . . .                                                                      | 75  |
| 6.4 | Simulated counts for different uranium enrichments within salts in NaI                                                              | 78  |
| 6.5 | Simulated counts with and without insulation in NaI . . . . .                                                                       | 84  |
| 7.1 | IAEA fast neutron coincidence collar . . . . .                                                                                      | 115 |

# List of Acronyms

|                |                                                          |
|----------------|----------------------------------------------------------|
| <b>AWCC</b>    | active well coincidence counter                          |
| <b>ARIS</b>    | Advanced Reactor Information System                      |
| <b>ASME</b>    | American Society of Mechanical Engineers                 |
| <b>BARC</b>    | Bhabha Atomic Research Centre                            |
| <b>CANDU</b>   | Canadian deuterium uranium                               |
| <b>CNRS</b>    | Centre National de la Recherche Scientifique             |
| <b>CMSR</b>    | compact molten salt reactor                              |
| <b>CEMO</b>    | continuous enrichment monitor                            |
| <b>CHEM</b>    | cascade header enrichment monitor                        |
| <b>DOE</b>     | Department of Energy                                     |
| <b>DU</b>      | depleted uranium                                         |
| <b>ECGS</b>    | electrically cooled high-purity germanium system         |
| <b>Euratom</b> | European Atomic Energy Community                         |
| <b>ESARDA</b>  | European Safeguards Research and Development Association |
| <b>FASTR</b>   | Facility to Alleviate Salt Technology Risks              |

|                 |                                            |
|-----------------|--------------------------------------------|
| <b>FNCL</b>     | fast neutron coincidence collar            |
| <b>FWHM</b>     | full width at half maximum                 |
| <b>GEB</b>      | Gaussian energy broadening                 |
| <b>GEM</b>      | general enrichment meter                   |
| <b>GIF</b>      | Generation IV International Forum          |
| <b>HALEU</b>    | high-assay low-enriched uranium            |
| <b>HDPE</b>     | high-density polyethylene                  |
| <b>HPGe</b>     | high-purity germanium                      |
| <b>HKED</b>     | hybrid K-edge densitometry                 |
| <b>ISOCS</b>    | In Situ Object Counting System             |
| <b>IMSBR</b>    | Indian Molten Salt Breeder Reactor         |
| <b>IHTR</b>     | Innovative High Temperature Reactor        |
| <b>IMSR</b>     | integral molten salt reactor               |
| <b>IAEA</b>     | International Atomic Energy Agency         |
| <b>INMM</b>     | International Nuclear Materials Management |
| <b>LEU</b>      | low-enriched uranium                       |
| <b>LFTR</b>     | Liquid Fluoride Thorium Reactor            |
| <b>LWR</b>      | light water reactor                        |
| <b>MBA</b>      | material balance area                      |
| <b>MC&amp;A</b> | material control and accountancy           |

**MCFR** Molten Chloride Fast Reactor

**MCRE** Molten Chloride Reactor Experiment

**MSRR** Molten Salt Research Reactor

**MSR** molten salt reactor

**MSRE** Molten Salt Reactor Experiment

**MCNP** Monte Carlo N-Particle

**NNSA** National Nuclear Security Administration

**NU** natural uranium

**NRC** Nuclear Regulatory Commission

**ORIGEN** Oak Ridge Isotope Generation code

**OLEM** On-line Enrichment Monitor

**ORNL** Oak Ridge National Laboratory

**PNEM** passive neutron enrichment monitor

**PHWR** pressurized heavy water reactor

**R&D** research and development

**ROI** region of interest OIs]roiregions of interest

**SINAP** Shanghai Institute of Applied Physics

**SNM** special nuclear material

**TMSR-LF1** Thorium Molten Salt Reactor–Liquid Fuel

**TMSR-SF1** Thorium Molten Salt Reactor–Solid Fuel

**TRISO** TRi-structural ISOtropic

**UCVS** unattended cylinder verification station

**US** United States

**WISSR** Wielenga Innovative Static Salt Reactor

# Chapter 1

## Introduction

Advanced reactor design concepts have gained significant traction as countries around the world consider nuclear power plants to meet increased energy demand using carbon-free sources [1, 2]. Many of these reactor concepts include significant safety benefits (e.g., passive safety features) and use fuel more efficiently than the light water reactors that constitute the overwhelming majority of the world’s current commercial reactor fleet. In 2002, experts at the Generation IV International Forum (GIF) evaluated more than 130 reactor concepts and recommended 6 promising reactor technologies for further research and development (R&D). GIF recommended the following reactor concepts (Figure 1.1):

- Gas-cooled fast reactors
- Lead-cooled fast reactors
- Molten salt reactors
- Sodium-cooled fast reactors
- Supercritical water-cooled reactors
- Very-high-temperature reactors [3]

Some of these reactor concepts differ from existing reactors (e.g., light water reactors (LWRs) and pressurized heavy water reactors (PHWRs)) in ways that make

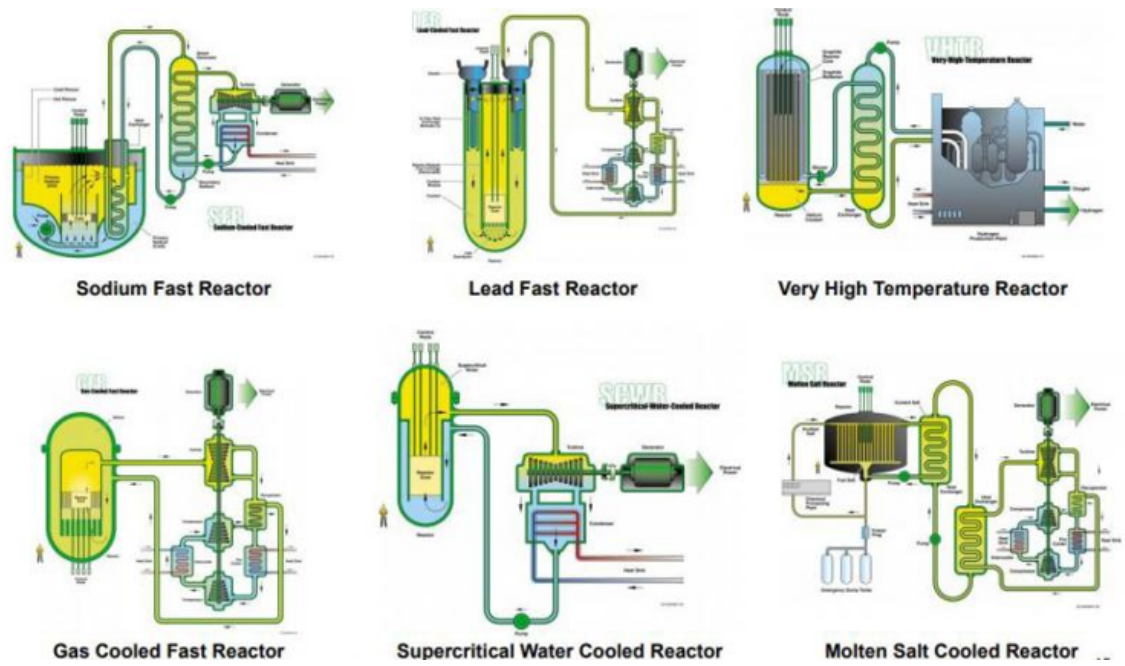


Figure 1.1: Prospective control areas in a liquid-fueled MSR facility [4]

accounting for nuclear material using traditional approaches and measurement technologies challenging. In particular, advanced reactor concepts that use continuously circulating fuel require material accountancy approaches that are distinct from (and more complex than) LWRs with fixed fuel assemblies that can be counted. Liquid-fueled (i.e., salt-fueled) molten salt reactors (MSRs) and pebble bed reactors are both advanced reactor concepts that use circulating fuel.

## 1.1 Molten Salt Reactors

Currently, there are 8 distinct MSR designs included in the International Atomic Energy Agency (IAEA)’s Advanced Reactor Information System (ARIS) database, and open sources indicate at least 16 other MSR designs are currently being developed (e.g., by vendors or national organizations) [5–7]. In alphabetical order by the design organization, the 24 (combined) designs are listed in Table 1.1. Other design organizations have previously developed MSR design concepts. For example, the Molten Salt Reactor Experiment (MSRE) was a research-scale MSR that went critical in 1965 and operated for 4 years at Oak Ridge National Laboratory (ORNL) in Oak Ridge, Tennessee [8]. Since then, several other MSR design concepts have been generated; however, Table 1.1 only includes concepts currently listed in ARIS and those that open-source publications indicate are currently being pursued [5, 9–27]. These MSR design concepts represent designers in Canada, China, Denmark, France, Indonesia, Japan, Russia, South Korea, the United Kingdom, and the United States (US), as well as potential operational reactors around the world [5, 11, 14, 20–22, 24–26]. The table includes the design concept, developer, and type of MSR.

In December 2020, the US Department of Energy (DOE) announced that two of its five awards for risk reduction for future nuclear reactor demonstration projects were awarded to MSRs. The two MSR recipient teams were

1. Hermes Test Reactor: the Kairos Power LLC TRi-structural ISOtropic (TRISO) pebble-fueled reactor that uses a low-pressure fluoride salt coolant

2. Molten Chloride Reactor Experiment (MCRE): the Southern Company Services Inc. fast-spectrum salt reactor experiment related to TerraPower’s Molten Chloride Fast Reactor (MCFR), a future commercial MSR design in which fissile material is dissolved in chloride salt [28]

These are examples of two distinct types of MSRs. The first is a solid-fueled, molten salt-cooled MSR, with no fissile material in the salt coolant. The second is a liquid salt-fueled MSR, in which the primary loop consists of fissile material dissolved in molten-salt coolant. A third type of MSR contains liquid salt in distinct fuel pins (or tubes) [6].

Proposed MSR concepts vary widely across design features, including the physical, chemical, and isotopic composition of fresh and irradiated fuel. Figure 1.2 depicts the various types of nuclear material proposed for use as fresh fuel in current MSR design concepts. Operational neutron energy spectra (e.g., ranging from thermal to fast, including hybrid) and breeding ratios also vary significantly across design concepts. Some concepts are burner reactors designed to transmute the spent nuclear fuel from LWRs or PHWRs, whereas others are breeder reactors designed to breed fissile  $^{233}\text{U}$  or  $^{239}\text{Pu}$  from naturally occurring, fertile  $^{232}\text{Th}$  or  $^{238}\text{U}$ , respectively. Liquid-fueled MSR design concepts typically have low excess reactivities and therefore require online fuel additions (usually small quantities), either through fresh fuel additions or through recycling of fissile material bred during operation. Some MSR concepts are designed to be a part of a once-through fuel cycle, but others involve chemical separation of fissile materials. Those that include plans to recycle the fuel differ by whether the chemical processing would be done on-site, as a process integrated into the reactor system, or in a separate process stream (e.g., at a reprocessing facility), similar to how LWR or PHWR reactor spent fuel is reprocessed in some countries. Because of these broad design variations, the nuclear material accountancy and control approaches for this class of reactors will vary significantly [6].

Table 1.1: Current MSR design concepts

| Design Organization                                                           | Design Name                                       | MSR Type     |
|-------------------------------------------------------------------------------|---------------------------------------------------|--------------|
| Alpha Tech Research Corp.                                                     | Micro Molten Salt Reactor (ARC Generator)         | salt-fueled  |
| Bhabha Atomic Research Centre (BARC)                                          | Innovative High Temperature Reactor (IHTR)        | salt-cooled  |
| BARC                                                                          | Indian Molten Salt Breeder Reactor (IMSBR)        | salt-fueled  |
| Copenhagen Atomics                                                            | Waste Burner                                      | salt-fueled  |
| Exodys Energy                                                                 | Fast Chloride Molten Salt Reactor (FC-MSR)        | salt-fueled  |
| Flibe Energy                                                                  | Liquid Fluoride Thorium Reactor (LFTR)            | salt-fueled  |
| Centre National de la Recherche Scientifique (CNRS)                           | Molten Salt Fast Reactor (MSFR) <sup>1</sup>      | salt-fueled  |
| International Thorium Molten-Salt Forum                                       | Molten Salt Reactor–FUJI (MSR-FUJI) <sup>1</sup>  | salt-fueled  |
| Kairos Power                                                                  | FHR (KP-FHR)                                      | salt-cooled  |
| Moltex Energy                                                                 | FLEX reactor                                      | salt in pins |
| Moltex Energy                                                                 | Stable Salt Reactor–Thorium (SSR-Th) <sup>1</sup> | salt in pins |
| Moltex Energy                                                                 | Stable Salt Reactor–Uranium (SSR-U) <sup>1</sup>  | salt in pins |
| Moltex Energy                                                                 | Stable Salt Reactor–Wasteburner (SSR-W)           | salt in pins |
| Natura Resources LLC                                                          | Molten Salt Research Reactor (MSRR)               | salt-fueled  |
| NA Dollezhal Research and Development Institute of Power Engineering (NIKIET) | RI IZhSR                                          | salt-fueled  |

Table 1.1: *(Continued)*

| Design Organization                           | Design Name                                                                               | MSR Type    |
|-----------------------------------------------|-------------------------------------------------------------------------------------------|-------------|
| Oak Ridge National Laboratory                 | Small fluoride salt-cooled High Temperature Reactor (SmAHTR) <sup>1</sup>                 | salt-fueled |
| Seaborg Technology                            | compact molten salt reactor (CMSR)                                                        | salt-fueled |
| Shanghai Institute of Applied Physics (SINAP) | Thorium Molten Salt Reactor–Liquid Fuel (TMSR-LF1)                                        | salt-fueled |
| SINAP                                         | Thorium Molten Salt Reactor–Solid Fuel (TMSR-SF1)                                         | salt-cooled |
| TerraPower                                    | Molten Chloride Fast Reactor (MCFR)                                                       | salt-fueled |
| Terrestrial Energy                            | integral molten salt reactor (IMSR)                                                       | salt-fueled |
| ThorCon International                         | ThorCon                                                                                   | salt-fueled |
| University of California, Berkeley            | Mark 1 Pebble-Bed Fluoride-Salt-Cooled High Temperature Reactor (Mk1 PB-FHR) <sup>1</sup> | salt-cooled |
| Wielenga Innovation Foundation                | Wielenga Innovative Static Salt Reactor (WISSR)                                           | salt-fueled |

<sup>1</sup>It is unclear whether this design is currently being actively worked on.

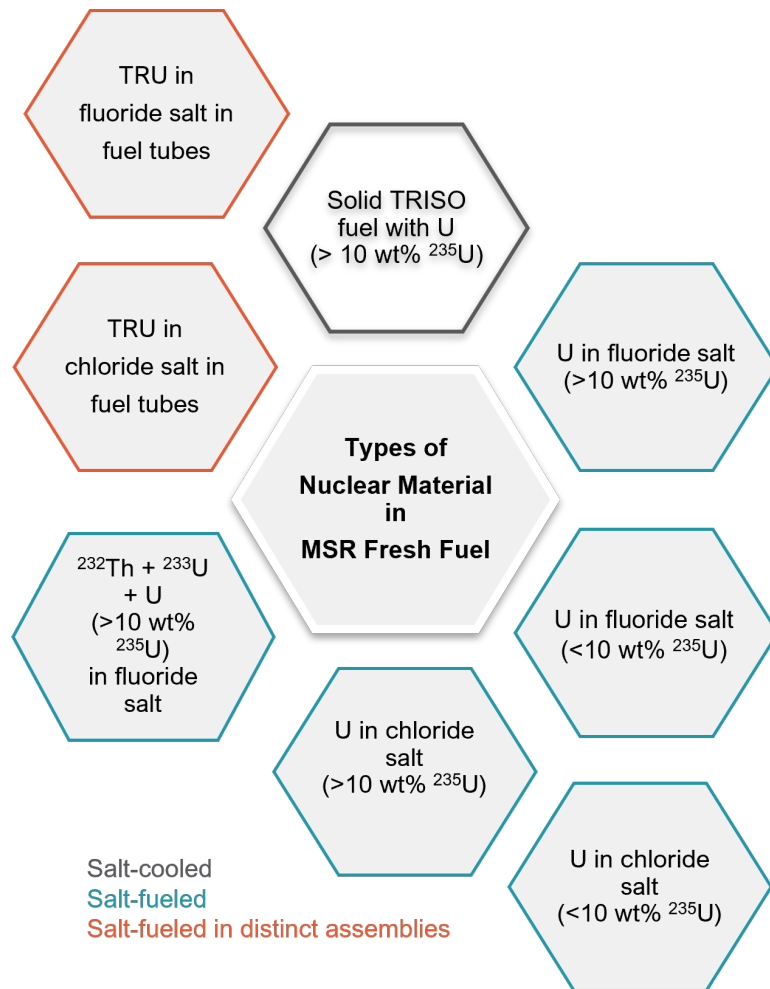


Figure 1.2: Nuclear material types seen in MSR design concepts (Adapted from [6])

## 1.2 Safeguards and Security

International nuclear safeguards applied by the IAEA are technical measures in place to ensure that a Member State is using nuclear material and facilities only for peaceful purposes in accordance with its legally binding agreement. For States with comprehensive safeguards agreements, the IAEA's generic safeguards objectives are (i) to detect any diversion of declared nuclear material at declared facilities or locations outside facilities where nuclear material is customarily used; (ii) to detect any undeclared production or processing of nuclear material at declared facilities or locations outside facilities; and (iii) to detect any undeclared nuclear material or activities in the State as a whole [29]. Nuclear security typically refers to the goal of preventing or detecting theft of fissile or source material or sabotage of a nuclear facility. In the US, nuclear security is referred to as domestic safeguards. The overall goal of US domestic safeguards is to ensure that special nuclear material (SNM) is not stolen or otherwise diverted from a facility [30]. Domestic safeguards incorporate both physical protection and material control and accountancy (MC&A) to achieve this goal [31]. Nuclear material accountancy, containment (e.g., tamper-indicating devices), and surveillance (e.g., cameras) are commonly used as safeguards and security measures to meet both domestic and international objectives in nuclear facilities.

For domestic safeguards purposes in the US, any operator licensed by the Nuclear Regulatory Commission (NRC) must maintain a MC&A program that tracks and verifies the SNM (i.e., Pu,  $^{233}\text{U}$ , or uranium enriched in the isotope  $^{235}\text{U}$ ) at the facility n[32]. Although currently licensed reactors in the US (i.e., LWRs) are not required to submit a Fundamental Nuclear Material Control plan with their licensing application, it is very likely that the NRC will require MSR facilities to do so because they are bulk processing facilities [33]. Outside the US, most countries have regulations that require states to generate a control and accountancy plan for materials defined by the IAEA as nuclear material (i.e., most Pu, U, and Th).

Additionally, States with legally binding comprehensive safeguards agreements with the IAEA must declare quantities and locations of nuclear material to the IAEA. The IAEA then independently verifies those quantities at the material locations. Several of the organizations listed in Table 1.1 have plans to construct and operate demonstration MSR facilities within this decade. Therefore, facility operators and some regulatory agencies (e.g., European Atomic Energy Community (Euratom) and the IAEA) will need methods and technologies to quantify fissile and fertile isotopes within MSR fuel. The fissile materials (and fertile materials in the case of the IAEA) will need to be quantified in both solid-fueled, salt-cooled, and liquid salt-fueled MSRs. All current solid-fueled, salt-cooled MSR concepts use TRISO fuel in pebble form. The quantities of nuclear material within the nonfissile salt coolant in salt-cooled reactors will be negligible. Therefore, the material accountancy challenges for salt-cooled MSR designs will be similar to the challenges for other (e.g., gas-cooled) pebble-bed reactor designs.

Although the IAEA has been implementing safeguards in facilities for decades, it has never implemented safeguards in liquid-fueled MSRs. The IAEA has identified the need for additional R&D in this area, as stated in its 2022–2023 Development and Implementation Support Programme for Nuclear Verification. This Programme is the IAEA’s biennial publication that outlines the IAEA Department of Safeguards’ near-term efforts that require extrabudgetary support to implement safeguards. The 2022–2023 publication includes Outcome 3, under Task SGCP-003: Safeguards Approaches: “Enhanced ability to safeguard new types of facilities through development of safeguards concepts and approaches for pyroprocessing plants and small modular and/or Gen IV reactors” [34]. Additionally, the IAEA’s Resource Mobilization Priorities describes the Department of Safeguards priorities for enhancing its capabilities. Focus Area V: Core Activities includes V.6.C4: “Ability to perform process monitoring and associated data analysis for safeguarding facilities, particularly advanced reactors with liquid or pebble fuel” and requests support from partners through financial resources, collaboration, expertise, and R&D [35].

Additionally, US DOE leadership has also stated the need to consider safeguards and security during the design phases of reactor and fuel cycle facility development. This effort is called safeguards and security by design. At a presentation to the joint 2023 International Nuclear Materials Management (INMM) and European Safeguards Research and Development Association (ESARDA) annual meeting, the National Nuclear Security Administration (NNSA) Administrator, Jill Hruby, said “new reactor types, especially small modular reactors, or SMRs, and advanced reactors, or ARs, will also require new safeguards and security approaches. With advanced reactors in the early stages of development, there is an opportunity for governments, regulators, and the nuclear industry to work together not only to strengthen safety features, but also security and safeguards features of nuclear reactors and their associated fuel cycle facilities” [36].

Lastly, a 2023 US National Academies published report from the “Laying the Foundation for New and Advanced Nuclear Reactors in the United States” committee states “The International Atomic Energy Agency (IAEA) and the Department of Energy (DOE) should identify the funding, personnel, regulatory analyses, and key technology gaps for pilot programs in international safeguards for advanced reactors” [37].

Despite MSR concepts being distinct from LWRs, effective safeguards approaches can be developed. This work uses MSRs as a case study of reactors with continuously circulating fuel to (i) identify a specific, high-priority safeguards technical objective that is common across MSR vendor designs, (ii) propose a potential technical solution, and (iii) evaluate several technology options to meet the objective.

### **1.3 Original Contributions**

Following a thorough literature review of all current liquid-fueled MSR design concepts, the author performed a critical analysis to define safeguards-relevant design features for liquid-fueled MSRs. Next, the author identified MC&A objectives across

a nominal liquid-fueled MSR and proposed MC&A elements based on current MC&A technologies. Through a gap analysis, the author down-selected the highest-priority R&D facility-level MC&A objective for technology development. This research is novel in that there was no defined methodology for performing these critical analyses and it has not been done before. This work should guide stakeholders in identifying clear MC&A objectives that need further R&D.

The MC&A objective down-selected through the gap analysis for R&D was quantifying, or monitoring fresh fuel entering difficult to access areas within the MSR system. The author developed a computational tool kit that combines SCALE 6.3.1/ORIGEN input and output files, Python scripts, and MCNP 6.2 input and output files to calculate and evaluate gamma and neutron emissions from the salt, transport these emissions through piping and insulation, and generate detector responses in modeled detectors external to the pipes. The author identified MSR salt chemistry and pipe parameters that represent a broad range of current liquid-fueled MSR designs and performed a parametric study analyzing how different impacts affect the feasibility of using penetrating radiation measurements techniques for safeguards and security. This work identifies promising methods for monitoring unirradiated fuel salt in liquid-fueled MSRs for both safeguards and security purposes.

## Chapter 2

# MSR Facility-Level Safeguards

## Objectives

The NRC has decades of experience supporting facility operators in developing and implementing MC&A plans for bulk processing facilities that have operated in the US: fuel fabrication facilities and enrichment plants. The IAEA has decades of experience implementing safeguards in these facility types. The IAEA also has experience developing and implementing safeguards approaches in PUREX reprocessing facilities, conversion facilities, and R&D facilities that use nuclear material in bulk form. However, MSR facilities would be distinct from these existing facilities. MSR facilities are distinct from reprocessing facilities (PUREX, electrochemical, etc.) in the following ways:

1. Most MSR design concepts have no chemical separation of nuclear material within the facility; only a few proposed designs include chemical separation of fissile material
2. MSR facilities will likely be lower throughput facilities than reprocessing facilities (there will likely only be small amounts of material arriving at and leaving the facility and these transfers will likely be low frequency)

3. The material at MSR facilities would require high temperatures (e.g., above 500 °C) to remain as a liquid; the salt eutectics solidify at room temperature

MSR facilities are distinct from fuel fabrication facilities and enrichment facilities in the following ways:

1. There are significant production and removal terms to material balances in an MSR because of transmutation and depletion from irradiation; accountable material is not just moving within the facility or process streams, it is being created and destroyed with inventories changing rapidly
2. The material is practically inaccessible when behind the biological shielding (e.g., confinement) because of the lack of physical access points and high radiation environments when the reactor is operating and after shutdown (due to decay of fission products)
3. MSRs will likely be lower throughput facilities than enrichment and fuel fabrication facilities

Typically, facilities are divided into material balance areas (MBAs), and nuclear material quantities must be reported to a regulatory body when accountable materials move from one MBA to another. For example, facility operators are required, for security purposes, to account for and control nuclear material under license agreements. Material accountancy regulations in the US will require declaration of  $^{235}\text{U}$  to the whole gram and natural uranium (NU) (or depleted uranium (DU)) to the whole kilogram [38] any time the quantities change within an MBA. In countries with comprehensive safeguards agreements in force with the IAEA, facility operators must report the quantities of  $^{235}\text{U}$ ,  $^{233}\text{U}$ , total uranium, thorium, and plutonium to the IAEA. Facility operators report changes in nuclear material quantities in each defined MBA. In the US, these changes are reported in the Nuclear Material Management and Safeguards System. In States with comprehensive safeguards agreements in force, these changes are captured in inventory change reports and are reported to the IAEA via the State or regional authority responsible for safeguards implementation.

There are multiple points in the nuclear material flows of future MSR facilities where measurements might be taken for safeguards or security purposes. The first goal of this research was to identify the facility-level safeguards technical objectives in a liquid-fueled MSR. Considering the broad range of liquid-fueled MSR concepts currently under design, a list of facility-level technical safeguards objectives was defined. This list enables consideration of the goals against (i) existing and feasible technology systems that might be used to meet these goals and (ii) the breadth of the conceptual designs of MSRs. Considering these aspects allowed the author to select a high-priority technical safeguards objective that (i) could be reasonably achieved within the next  $\approx 5$  years—consistent with the timeline that vendors plan for liquid-fueled MSR demonstration reactors to become operational, (ii) could not be easily met with existing monitoring technologies already used for safeguards purposes, and (iii) could be met for a variety of conceptual reactor designs. Selecting a high-priority objective ensures that near-term research involving modeling, monitoring technology selection, and design development for a solution will be relevant, intentional toward meeting safeguards goals, and useful to stakeholders (e.g., facility operators, State or regional authorities responsible for safeguards, the IAEA). At this time, it is not clear which of the many advanced reactor designs will overcome obstacles to licensing and construction and become operational. With the breadth of the MSR design concepts, it is important to consider monitoring technology solutions that might apply to as many of the design concepts as reasonable.

This research focuses on liquid-fueled MSR design concepts because (i) more vendor designs are liquid-fueled (i.e., salt-fueled vs. salt-cooled) MSRs, and (ii) when considering material accountancy, liquid-fueled reactors are the most distinct from traditional LWRs.

## 2.1 Safeguards-Relevant Design Features of MSRs

Specific MSR designs vary significantly within the class of MSR reactor concepts [6]. While some work has been done to classify or categorize MSR designs ([7, 39]), these categories were either limited to specific fuel cycles (e.g., [7] focused on thorium fuel cycle MSRs only) or are focused on, for example, neutronics considerations and not specifically on design features that are relevant to safeguards or security. Table 2.1 shows the comprehensive list of safeguards-relevant design features of liquid-fueled MSRs developed by the author. This table was developed following a literature review of past and current MSR vendor designs (Section 1.1) and includes notes on the relevance of each feature to both domestic and international nuclear safeguards.

## 2.2 Safeguards Objectives and Prospective MC&A Plan

Because of the breadth of liquid-fueled MSR designs and the distinct objectives of domestic and international nuclear safeguards, a single, generic safeguards approach cannot be developed for MSRs. However, by assessing the common features of most designs, the author identified the domestic safeguards objectives (which are likely similar to technical international safeguards objectives) and prospective elements that will likely comprise an MC&A plan to meet domestic safeguards requirements (i.e., in the US's case, the NRC) to ensure nuclear material is not stolen or otherwise removed from a future facility. Figure 2.1 displays the three likely distinct control areas (facilities or the IAEA may designate these as an MBAs or sub-MBAs) for a liquid-fueled MSR facility. Table 2.2\* includes facility-level MC&A objectives for a nominal low-enriched uranium (LEU)-fueled MSR. Note that the table includes

---

\*Reference [40] somewhat similarly developed a prospective safeguards approach focusing on international safeguards and a specific MSR design—ThorCon's thermal, graphite-moderated, modular system. There are many similar elements of the two independently developed approaches, even with these distinctions.

the US terminology “SNM” and “MC&A” because it is targeted for US domestic safeguards approaches. However, the content is relevant for and translatable to nuclear material accountancy and control for international safeguards approaches.

A notable contribution of this research is a novel methodology that will assist in the selection and development of technology systems to meet safeguards objectives for continuously circulating reactor systems. Although the proposed research identifies a solution for a specific safeguards facility-level technical objective defined in Chapter 3, perhaps more importantly, it introduces a framework that can be applied to other safeguards challenges in advanced reactor concepts with continuously circulating fuel (e.g., liquid-fueled MSR and pebble-bed reactors).

Table 2.1: Safeguards-relevant design features of MSRs

| Design Feature Category     | Design Feature                                                                                   | Description                                                                                                                                                                                                                                                                            | Relevance to Domestic Safeguards                                                                                                                   | Relevance to International Safeguards                                                                                                                                                                                                                             |
|-----------------------------|--------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Fuel                        | Fissile material content (type, physical/chemical form, quantity, enrichment) in fresh fuel salt | Designs include different plans for fissile material in the fresh fuel, including <5 wt % $^{235}\text{U}$ , 10–20 wt % $^{235}\text{U}$ , Th, Pu, and other actinides.                                                                                                                | Fissile material is accountable material; LEU > 10% enriched has different regulations than LEU < 5% in US domestic safeguards.                    | Fissile material types, quantities, enrichments, and locations must be declared to, and independently verified by, the IAEA.                                                                                                                                      |
|                             | Fertile material content (type, quantity) in fresh fuel salt                                     | In salt-fueled breeder MSRs, fertile material will be added to salt in either the primary salt loop or a separate blanket salt loop. Burner reactor designs will also contain fertile material (e.g., $^{238}\text{U}$ ).                                                              | Fertile material is not accountable material but is helpful in estimating/bounding the quantities of SNM that will be created through irradiation. | Fertile material types, quantities, and locations must be declared to, and can be independently verified by, the IAEA.                                                                                                                                            |
| Operational plans/practices | Inventory of fresh fuel salt held at the facility                                                | Facility operators will likely plan to store different amounts of fresh fuel onsite.                                                                                                                                                                                                   | Inventories and containment and surveillance of this material will require MC&A resources.                                                         | Inventories and containment and surveillance of this material will require IAEA and facility operator resources.                                                                                                                                                  |
|                             | Method and frequency in which makeup salt is added                                               | Designs will likely incorporate different plans for adding makeup salt.                                                                                                                                                                                                                | MC&A plans will require either surveillance or measurements to verify that no SNM is diverted during refueling.                                    | Material balance evaluations require quantification of the nuclear material added to the system during refueling. The method and frequency with which salt is added will determine what technologies are practical for the IAEA to use to monitor fuel additions. |
|                             | Frequency in which reactor components are replaced                                               | Components within MSR designs will have to be replaced periodically (e.g., in thermal spectrum MSRs, the graphite moderator blocks will have to be replaced every few years). When reactor components are replaced, fuel salt will be drained from the system and stored in a tank(s). | MC&A plans should incorporate considerations to ensure no diversion of SNM during replacement of reactor components.                               | Accounting for nuclear material during maintenance will likely require significant IAEA resources. These could be opportunities for the IAEA to perform inventories of irradiated salt in tank(s).                                                                |

Table 2.1: *(Continued)*

| Design Feature Category | Design Feature                                                                     | Description                                                                                                                                                                                                                                                 | Relevance to Domestic Safeguards                                                                                                                                        | Relevance to International Safeguards                                                                                                                                                                                                                                           |
|-------------------------|------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Physics of the reactor  | Power of the reactor                                                               | Some salt-fueled MSR designs are planned as significantly smaller (e.g., <300 MWth) than large, commercial LWRs (~1,000 MWth); other designs exist for larger (~1,000 MWth) salt-fueled MSRs.                                                               | The power of the reactor impacts the throughput of fissile material, which impacts the total quantity of fissile material on the site.                                  | The power of the reactor impacts the throughput of fissile and fertile material and bounds the amount of undeclared fissile material that could be produced at the facility; this impacts IAEA resources to ensure timely detection of diversion/misuse of the facility.        |
|                         | Breeding ratio (connected with neutron energy spectrum)                            | The breeding ratio determines how effectively the reactor is transmuting fertile material into fissile material.                                                                                                                                            | The breeding ratio will impact the total quantity of fissile material within the reactor over the core lifetime.                                                        | The breeding ratio will impact the total quantities of fissile and fertile material within the reactor over the core lifetime; both must be declared to the IAEA.                                                                                                               |
|                         | Neutron energy spectrum (connected with breeding ratio)                            | The neutron energy spectrum in which the reactor primarily operates impacts how much Pu is produced in U/Pu fuel-cycle reactors.                                                                                                                            | Thermal energy spectrum MSRs will typically have lower amounts of Pu in their waste; Pu waste characterization will likely be a part of a facility's MC&A plan.         | Thermal energy spectrum MSRs will typically have lower amounts of Pu in their waste; IAEA frequency and intensity of inspections will be impacted by the quantities of nuclear material.                                                                                        |
| Other systems           | Chemical processing of salt to separate fissile material and re-use as makeup fuel | Some salt-fueled MSR designs (especially those with high breeding ratios) plan to separate fissile material from the salt and use it as makeup feed to refuel the reactor during operation. This approach is used in some U/Pu and Th/U fuel-cycle designs. | Nuclear material in this recycle stream, where fissile material is separated, stored, and returned to the reactor as makeup fuel, may need to be accurately quantified. | IAEA might have to independently verify quantities of nuclear material within decay tanks or streams of fissile material being separated. In Th fuel cycles, this may include verification of <sup>233</sup> Pa, which some MSR concepts plan to separate.                      |
|                         | Off-gas system                                                                     | Some designs include an off-gas system to actively filter out gaseous fission and activation products. These systems sometimes include decay tanks.                                                                                                         | Online measurements could occur in the off-gas system to provide data relevant to MC&A.                                                                                 | NDA measurements of fission products and/or ratios of certain actinides are sometimes used to determine burnup or other factors that are relevant in quantifying nuclear material in irradiated fuel. Cover gas and off-gas systems will impact the quantities of these gasses. |

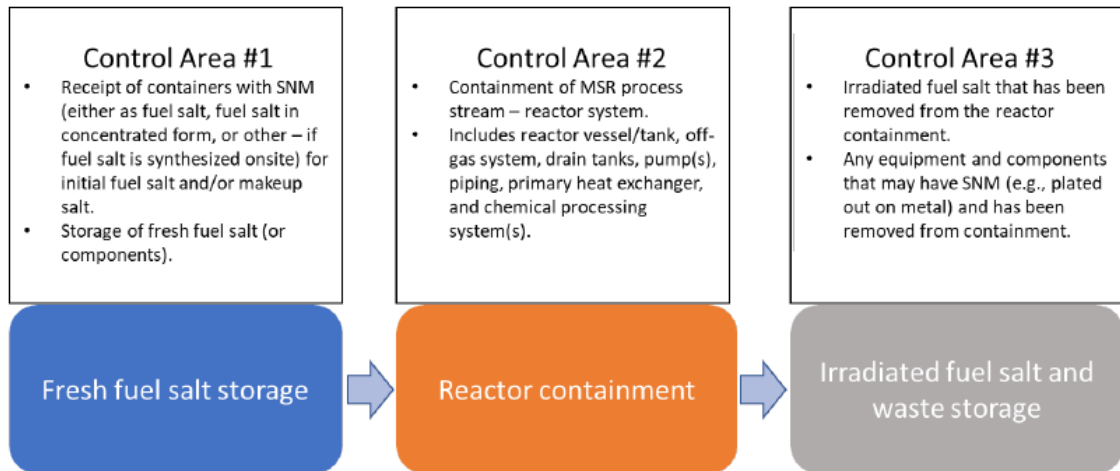


Figure 2.1: Prospective control areas in a liquid-fueled MSR facility [41]

Table 2.2: MC&A technical objectives and elements for a liquid-fueled MSR facility [41]

|                                          | MC&A Objectives                                         | SNM in Area                                                                                         | Physical Form of SNM                                                        | Measurement Environment                                                                                                  | MC&A Elements                                                                                                                                  |
|------------------------------------------|---------------------------------------------------------|-----------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------|
| Control Area #1: Fresh fuel salt storage | Quantify SNM upon receipt of material.                  | LEU (≤5 wt%)                                                                                        | SNM is within salt in a solid form.                                         | Fuel salt contained within metal (or metal alloy) containers.                                                            | Unique identifiers (serialized tags) on each container for tracking.                                                                           |
|                                          |                                                         |                                                                                                     |                                                                             |                                                                                                                          | Assay material being added to containment from containers; NDA and DA: gamma spectroscopy, mass spectrometry, hybrid k-edge densitometer, etc. |
|                                          | Ensure no SNM is removed from containers stored onsite. |                                                                                                     | Fuel salt will be heated to a liquid prior to entering reactor containment. | LEU has weak radioactive emissions.                                                                                      | Confirm integrity of TIDs on containers upon receipt and during inventories at facility.                                                       |
|                                          |                                                         |                                                                                                     |                                                                             |                                                                                                                          | Camera surveillance used to identify serial number of container(s) added to reactor containment.                                               |
|                                          | Quantify SNM entering reactor containment.              |                                                                                                     |                                                                             | Weigh fuel salt containers upon receipt, during inventories, and after salt is removed and added to reactor containment. |                                                                                                                                                |
|                                          |                                                         |                                                                                                     |                                                                             | TIDs on any valves that could be manually opened to allow access to the system.                                          |                                                                                                                                                |
| Control Area #2: Reactor containment     | Ensure no SNM is removed from reactor containment       | LEU (≤5 wt%)                                                                                        | SNM is liquid in salt matrix.                                               | Extremely high radiation due to fission products.                                                                        | TIDs on all pipes/hatches that could be used to access the containment.                                                                        |
|                                          |                                                         |                                                                                                     |                                                                             |                                                                                                                          | Measure actinide concentrations in fuel salt using NDA and/or DA, and total salt volume.                                                       |
|                                          |                                                         | Dynamic concentrations as <sup>235</sup> U is depleted and Pu is produced from neutron irradiation. | Molten salt is not physically accessible.                                   | Elevated ambient temperature.                                                                                            | Camera surveillance on all access points to detect removal of SNM.                                                                             |
|                                          |                                                         |                                                                                                     |                                                                             |                                                                                                                          | In-situ NDA techniques (gamma spectroscopy), processing monitoring for indications of diversion.                                               |

Table 2.2: (Continued)

|                                                                                                                                                | MC&A Objectives                                                       | SNM in Area                                                                 | Physical Form of SNM                                       | Measurement Environment                                               | MC&A Elements                                                                                                                                    |
|------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------|-----------------------------------------------------------------------------|------------------------------------------------------------|-----------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------|
| Control Area #3:<br>Irradiated fuel salt and waste storage                                                                                     | Quantify SNM in salt leaving reactor containment.                     | LEU ( $\leq 5$ wt%)                                                         | Assumed SNM is in salt matrix either as a liquid or solid. | Irradiated fuel salt is in shielded containers.                       | TIDs placed on doors of storage area.                                                                                                            |
|                                                                                                                                                | Ensure no SNM is removed from containers or equipment stored onsite.  |                                                                             |                                                            |                                                                       | TIDs placed on storage containers once filled with irradiated fuel salt or reactor components.                                                   |
|                                                                                                                                                |                                                                       |                                                                             |                                                            |                                                                       | Assay material being removed from containment to containers; NDA and DA: gamma spectroscopy, mass spectrometry, hybrid k-edge densitometer, etc. |
|                                                                                                                                                |                                                                       | TIDs placed on equipment entry points (e.g., waste reactor vessel closure). |                                                            |                                                                       |                                                                                                                                                  |
|                                                                                                                                                | Estimate holdup of SNM in equipment removed from reactor containment. | Plutonium and other actinides in irradiated fuel salt.                      |                                                            | Irradiated fuel salt is very radioactive because of fission products. | NDA holdup measurements and imaging techniques to locate material.                                                                               |
|                                                                                                                                                |                                                                       |                                                                             |                                                            |                                                                       | Camera surveillance on entry/exit points to storage area(s).                                                                                     |
|                                                                                                                                                |                                                                       |                                                                             |                                                            |                                                                       | Weigh empty and filled containers during inventories or upon changes to content.                                                                 |
| In-situ gamma-ray spectroscopy, process monitoring on piping and/or containers as SNM is moved out of reactor containment and into containers. |                                                                       |                                                                             |                                                            |                                                                       |                                                                                                                                                  |

<sup>†</sup>There will be minimal changes in radionuclide inventories over time due to radioactive decay (e.g.,  $^{241}\text{Pu} \rightarrow ^{241}\text{Am}$ ).

## Chapter 3

# Liquid-Fueled MSR Fresh Fuel Monitoring

Chapter 2 described the safeguards-relevant design features of MSRs and a critical analysis of the MC&A objectives across a future MSR facility. This chapter describes the MC&A objective selected for further research and described in the remaining chapters.

Safeguards approaches in fuel cycle facilities like enrichment, conversion, fuel fabrication, and reprocessing plants can rely heavily on nuclear material accountancy using mass balance evaluations, where material is quantified as it enters and exits an MBA. However, mass balances in existing LWR and PHWR reactor facilities require quantification of nuclear material into and out of the MBA as well as comparison against computationally predicted quantities in irradiated fuel because of fissile material consumption and production during irradiation. These computationally predicted quantities are generated using reactor physics codes based on the operational history of the reactor. Predicted quantities for irradiated MSR—as well as other advanced reactors—fuel may have significant uncertainties due to lack of opportunity for verification and validation of the reactor physics codes, as well as uncertainties in the nuclear data used to generate predicted quantities. Additionally,

the measurement environments of difficult to access areas within liquid-fueled MSR facilities (i.e., within the biological boundary that surrounds the reactor system) will be challenging. The high-temperature and high-radiation environments of MSR containment make theft of material challenging, but the concept of nuclear material being self protecting is only applicable to domestic safeguards. Facility operators will still need to account for nuclear material, and the IAEA will still need to verify that no declared nuclear material was diverted and that the facility was not misused for non-peaceful purposes. This means that initial safeguards approaches will likely rely heavily on containment and surveillance around the reactor's difficult to access areas to ensure that irradiated nuclear material is not removed. However, accounting for nuclear material in any fuel salt going into, or coming out of, the reactor containment will be critical.

Accurate material accountancy of both fissile and fertile materials within the fresh fuel will be essential to any safeguards approach. Accounting for nuclear material in fresh fuel feed is important not only to ensure there is no diversion of declared nuclear material during online addition of makeup salt but also to accurately bound the values of fissile material that can be produced from fertile materials undergoing irradiation. Fresh fuel measurements are the last chance of accurately accounting for nuclear material before it enters the part of the process flow that is inaccessible due to physical boundaries, limited process entry/exit points, high radiation fields, and high temperatures. If the declared quantities of fertile nuclear material that enter the reactor are inaccurate (either intentionally, to conceal diversion, or by accident), no measurements down the line will be able to accurately identify diversion of nuclear material produced from the irradiation of that fertile material.

There are no defined current approaches for monitoring feed for either domestic or international safeguards purposes in a liquid-fueled MSR. Based only on currently available monitoring technologies, a utility seeking a construction and/or operating license for a liquid-fueled MSR in the US may rely on containment, surveillance, and administrative controls. Containment elements might include tamper-indicating

devices on containers with fresh fuel salt, doors to fresh fuel storage areas, and valves that allow flow of salt to/from the reactor. Surveillance elements would likely include cameras on access points to the reactor. Administrative controls would likely include two-person verification and documentation of any actions related to fresh fuel additions. If relying on currently available monitoring technologies, the IAEA may plan to verify nuclear material quantities in any fresh fuel salt (or fuel salt components like  $\text{UCl}_4$  or  $\text{UCl}_3$ , depending on how material arrives at an MSR facility). Then, it might rely on containment and surveillance (e.g., cameras) to ensure that the quantities of nuclear material in initial and makeup salt added to the reactor were as declared. A nondestructive, in situ monitoring system would significantly improve on this system by not requiring review of camera feed and being more objective, data-driven, and reliable than administrative controls and procedural compliance. Overall, this system would significantly increase confidence that nuclear material was not diverted and the facility was not being misused.

Although ongoing research is currently assessing monitoring technologies that could be used for online measurements of fuel during irradiation, there is little to no research on developing a system of monitoring technologies to quantify nuclear material in fresh fuel salt being added to the reactor. There will be data relevant to quantifying nuclear material in fresh fuel salt for facility operations purposes (e.g., as specifications from the fuel fabrication facility, potentially from sampling or irradiated salt chemistry for corrosion monitoring). However, it is important for domestic safeguards purposes for the facility operator to be able to quantify material not just in containers received from off-site, but as the fuel enters the reactor. Likewise, it will be important for international safeguards purposes for the IAEA to be able to independently verify quantities entering containment.

Reference [42] provides a review of candidate techniques for material accountancy in irradiated uranium- and plutonium-bearing molten salts. Many of these techniques could have applications for quantifying actinides in unirradiated fresh fuel salts in MSR environments as well. Current research is investigating techniques for material

accountancy in irradiated fuel salts (e.g., optical spectroscopy [43], electrochemical sensors [44], and hybrid K-edge densitometry (HKED) [45, 46]). However, most of these technologies either require sensors that physically contact the salt or optical windows installed in piping. These requirements may require safety assessments or affect the willingness of facility operators to install the equipment within their system. In situ radiation detection systems that can be placed on the outside of piping has not been thoroughly explored for fresh, unirradiated fuel prior to this research. This research explores the feasibility of using in situ gamma or neutron detection external to a pipe for monitoring fresh fuel feed, where fresh fuel salt is being transferred into difficult to access areas.

Although it remains important to consider the widest breadth of future MSR concepts as reasonable to ensure this research is applicable to future operational designs, the wide breadth of conceptual designs makes it difficult to assess all design parameters. Table 3.1 includes planned fresh fuel salts for selected liquid-fueled design concepts that have been published. The fresh fuel salts listed in the table represent plans for initial fuel salt (i.e., the salt that will be the initial loading of the MSR when first starting operations). Some liquid-fueled MSR designers have plans for initial fuel salt that has distinct isotopic composition from makeup fuel salt (i.e., the salt that will be added over the reactor lifetime). The makeup salt in some designs has significantly lower  $^{235}\text{U}$  enrichments (and could be natural or DU or thorium). Very few current near-term designs include plans to add fissile material (e.g.,  $^{233}\text{U}$  bred during reactor operations in a separate blanket loop or within the main fuel salt loop but then chemically separated) back into the fuel salt loop.

Most near-term designs propose using an initial fuel salt feed that includes  $^{235}\text{U}$  in either a fluoride-based (for most thermal neutron energy spectrum designs) or chloride-based (for fast neutron energy spectrum designs) salt eutectic. This research explores the feasibility of monitoring uranium-bearing unirradiated fresh fuel with no other nuclear materials (e.g., plutonium, thorium) present within chloride and fluoride fuel salts. Future work could apply the methodology described in this research to

Table 3.1: Fuel salts proposed in select current liquid-fueled MSR design concepts [7, 9, 14, 17, 18, 21, 25, 47–51]

| Design Organization       | Design Name   | Fuel Salt                                                                                        | U Enrichment (wt% $^{235}\text{U}$ ) |
|---------------------------|---------------|--------------------------------------------------------------------------------------------------|--------------------------------------|
| Alpha Tech Research Corp. | ARC Generator | LEU in fluoride-based salt                                                                       | LEU                                  |
| Copenhagen Atomics        | Waste Burner  | $\text{LiF} - \text{ThF}_4 - (\text{U or Pu})\text{F}_4$                                         | unpublished                          |
| CNRS                      | MSFR          | $\text{LiF} - (\text{U}, \text{Pu})\text{F}_3 - \text{ThF}_4$                                    | unpublished                          |
| Flibe Energy              | LFTR          | Fuel salt: $\text{LiF} - \text{BeF}_2 - \text{UF}_4$ [+ $^{233}\text{U}$ bred in a blanket loop] | unpublished                          |
| Moltex Energy             | SSR-W         | spent Canadian deuterium uranium (CANDU) fuel in chloride-based salt                             | <NU                                  |
| Natura Resources          | MSRR          | $\text{LiF} - \text{BeF}_2 - \text{UF}_4$                                                        | $\approx 19.5$                       |
| Seaborg                   | CMSR          | fluoride-based salt                                                                              | unpublished                          |
| SINAP                     | TMSR-LF1      | $\text{LiF} - \text{BeF}_2 - \text{UF}_4$ [+ $\text{ThF}_4$ ]                                    | 19.75                                |
| TerraPower                | MCFR          | $\text{NaCl} - \text{UCl}_3$                                                                     | HALEU (12 to <20)                    |
| Terrestrial Energy        | IMSR          | $\text{UF}_4$ in fluoride-based carrier salt                                                     | LEU (<5)                             |
| ThorCon International     | ThorCon       | $\text{NaF} - \text{BeF}_2 - \text{UF}_4$                                                        | LEU                                  |

fuel salts with different chemical compositions to determine the feasibility of radiation monitoring external to piping for different fresh fuel salt options. All proposed uranium enrichments are less than 20.0 wt%  $^{235}\text{U}$ , but they range from LEU values traditionally used in LWRs to higher assay uranium enrichments termed high-assay low-enriched uranium (HALEU). HALEU enrichments are below 20.0 wt%  $^{235}\text{U}$  and typically above 10.0 wt%  $^{235}\text{U}$ . This work analyzes different enrichments ranging from NU to 19.75 wt%  $^{235}\text{U}$ .

The literature contains few descriptions of the practical aspects of how liquid-fueled MSR developers might plan to load initial and makeup salt into the reactor. This is likely because most liquid-fueled MSR designs are still in the conceptual design phase and may not have determined the details of fresh fuel loading at this time. Although the lack of specific information is challenging because it introduces more unknowns and requires assumptions, it creates opportunities to practice safeguards and security by design and allows the safeguards and security communities to make recommendations to industry about what design decisions might best facilitate safeguards and security implementation.

A key assumption of this research is that the fuel salt being added to the system will be loaded in liquid form. Because of the high melting point of proposed salt eutectics used as fuel in MSRs, the salt eutectics (and salt components) will be in solid form when stored in containers at a facility at room temperature and pressure. Facilities may choose to load fuel salt as a combined salt eutectic (e.g.,  $\text{NaCl-UCl}_3$  or  $\text{LiF-BeF}_2\text{-UF}_4\text{-ZrF}_4$ ) or as salt components (e.g.,  $\text{NaCl}$  and  $\text{UCl}_3$ ; FLiBe components and  $\text{UF}_4$ ). Additionally, fresh fuel salt could be added as a liquid or a solid. Facility operators could heat the salt (e.g., in salt batches within a furnace) and add initial and makeup salt to the system in liquid form, or they could add the salt in solid form. If the salt is added in solid form, the fuel salt would likely be in item form: either irregular from, for example, mechanically breaking apart the crystallized solid salt eutectic or regular if distinct solid units (e.g., spheres or compacts) are formed in the salt synthesis process. To move forward with this research, the author evaluated a homogeneous salt eutectic in liquid form within the piping. In situ monitoring using gamma and neutron signatures would be most effective if fuel salt is in a homogeneous, liquid form within the piping. This likely represents many liquid-fueled MSR designer plans. It is possible that liquid-fueled MSR developers could alter their method

of adding unirradiated fuel salt to incorporate heating the salt to liquid before feeding the salt if it is demonstrated that this has significant benefits to material accountancy for the purposes of meeting international safeguards obligations and nuclear security best practices. Most designs are early enough in the conceptual design phase that they could modify the practical aspects of how they plan to add initial or makeup fuel salt. Therefore, this research is likely relevant to the broadest range of liquid-fueled MSR vendors as is reasonable.

The measurement techniques explored in this research require the assumption that either molten salt could be held stationary within a pipe or that the moving salt would provide a constant signature over a period of time during which the salt could be measured. This may not be realistic or feasible in a real facility environment. Holding molten salt stationary in a pipe will be challenging because valves in high-temperature, corrosive MSR environments are difficult to work with. Pneumatic movement of salt from, for example, a furnace or fresh fuel tank to another tank within the difficult to access area could potentially be modified such that salt remains within a portion of piping for a period of time, but further work needs to be done to determine whether that period of time would be sufficient for measurements to be made external to the pipe. Additionally, further work should be done to assess realistic concealment techniques that may challenge the assumption that the salt moving through a pipe would provide a constant signature over a period of time for which the salt could be measured.

# Chapter 4

## Simulated Radiation Signatures from Fresh Fuel Salt

This research analyzes the feasibility of using in situ radiation detection to quantify, or monitor\*, additions of fresh fuel salt being fed through piping into a reactor system. Uranium-bearing, unirradiated fuel salt proposed for use in liquid-fueled MSRs will emit radiation in several forms, including gamma rays, x-rays, alpha particles, and neutrons. Any of these signatures might be a useful signature for quantifying or monitoring fresh fuel salt. This chapter describes the source term of the salt based on spontaneous decay and induced nuclear reactions, both qualitatively and quantitatively, through an analysis using SCALE 6.3/Oak Ridge Isotope Generation code (ORIGEN) [52].

Chloride-based salts typically have a larger uranium mass fraction than fluoride-based salts compared with molten fuel salts that use  $^{235}\text{U}$  as the fissile material. Table 4.1 includes chemical compositions of fuel salts for a selection of historical and current chloride-based and fluoride-based fuel salts.

The two salts analyzed in this research were a chloride-based salt:  $\text{NaCl-UCl}_3$  (66–34 mol%) (similar to TerraPower’s MCFR [50], which is 67–34 mol%) and a fluoride-based salt:  $\text{LiF-BeF}_2\text{-UF}_4\text{-ZrF}_4$  (65–29.1–0.9–5 mol%) (used in the ORNL MSRE) [53]. The mass fractions for total uranium, each isotope of uranium, and other salt components are listed

---

\*If uncertainties in nuclear material quantities are high, monitoring to confirm consistency with declared operations may still be useful.

Table 4.1: Salt compositions in select fresh MSR fuel salts

| MSR Design Name                          | Fuel Salt                                                                     | Reference |
|------------------------------------------|-------------------------------------------------------------------------------|-----------|
| Chloride salts                           |                                                                               |           |
| Aircraft Nuclear Propulsion Program      | NaF–ZrF <sub>4</sub> –UF <sub>4</sub> (53.09–40.73–6.18 mol%)                 | [54]      |
| Two-Fluid Molten Salt Breeder Reactor    | LiF–BeF <sub>2</sub> –UF <sub>4</sub> (68.5–31.3–0.2, mol%)                   | [54]      |
| Single-Fluid Molten Salt Breeder Reactor | LiF–BeF <sub>2</sub> –ThF <sub>4</sub> –UF <sub>4</sub> (71.5–16–12–0.5 mol%) | [54]      |
| Molten Salt Reactor Experiment           | LiF–BeF <sub>2</sub> –UF <sub>4</sub> –ZrF <sub>4</sub> (65–29.1–0.9–5 mol%)  | [53]      |
| Fluoride salts                           |                                                                               |           |
| TerraPower Molten Chloride Fast Reactor  | NaCl–UCl <sub>3</sub> (67–33 mol%)                                            | [50]      |
| Molten Salt Fast Reactor                 | LiF–(U,Pu)F <sub>3</sub> (77.5–22.5 mol%)                                     | [14]      |

in Table 4.2. The mass fractions in the table assume lithium is enriched to 99.99 atom%  $^7\text{Li}$ . Natural lithium is 92.41 atom%  $^7\text{Li}$ . Lithium-6 has a high cross section for neutron absorption and produces tritium upon neutron absorption [55]. Therefore, MSR developers propose using lithium enriched in the isotope  $^7\text{Li}$ . The MSRE used lithium enriched in the isotope  $^7\text{Li}$  to greater than 99.99% [56]. This also assumes that chlorine is enriched to 75 atom% in the isotope  $^{37}\text{Cl}$ . The abundance of  $^{37}\text{Cl}$  in natural chlorine is 24.24 atom%. Although natural chlorine could be used in fuel salt,  $^{35}\text{Cl}$  introduces a number of challenges [56]. Chlorine-35 has a higher neutron absorption cross section than chlorine-37 at neutron energies up to approximately 1.75 MeV, including the resonance region, which leads to a higher rate of undesirable parasitic neutron absorption [55]. Additionally,  $^{35}\text{Cl}$  produces  $^{36}\text{Cl}$  upon neutron absorption, which has a long half-life ( $t_{1/2}$ ) of  $3.01 \times 10^5$  years so presents a problem as a water-soluble long-lived radionuclide [57]. Lastly, the  $^{35}\text{Cl}$  cross section for the  $(n,p)$  reaction is not insignificant. That reaction yields  $^{35}\text{S}$ , which is both highly corrosive and introduces the possibility for formation of uranium sulfide, which is a solid [57]. The author selected four distinct uranium enrichments to analyze: NU, 2 wt%  $^{235}\text{U}$ , 5 wt%  $^{235}\text{U}$ , and 19.75 wt%  $^{235}\text{U}$ . These selections were driven by stated intentions of developers currently pursuing liquid-fueled MSR designs, as described in Chapter 3. All other elements (i.e., F, Be, Zr, Na) are assumed to have natural isotopic compositions. For the two salts analyzed in this research, the uranium mass fractions in the chloride salt are approximately an order of magnitude higher than in the fluoride salt.

## 4.1 Modes of Decay

Passive radiation emissions from the salt include:

1.  $\alpha$  particles from the  $\alpha$  decay of  $^{234}\text{U}$  ( $t_{1/2}=2.46 \times 10^5$  years),  $^{235}\text{U}$  ( $t_{1/2}=7.04 \times 10^8$  years), and  $^{238}\text{U}$  ( $t_{1/2}=4.468 \times 10^9$  years)
2. neutrons from

- (a)  $(\alpha, n)$  reactions due to the absorption of  $\alpha$  particles—from the decay of uranium isotopes—by light elements, including beryllium<sup>†</sup>, F, Li, and Cl
- (b) spontaneous fission of  $^{234}\text{U}$  ( $t_{1/2\text{sf}}=1.42 \times 10^{16}$  years [59]),  $^{235}\text{U}$  ( $t_{1/2\text{sf}}=9.8 \times 10^{18}$  years [59]), and  $^{238}\text{U}$ <sup>‡</sup>.
- (c) self-induced fission of  $^{235}\text{U}$  or  $^{238}\text{U}$
- (d)  $(\gamma, n)$  reactions in e.g.,  $^9\text{Be}$

### 3. gammas and x-rays from

- (a)  $\alpha$  decay of  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$
- (b) spontaneous fission of  $^{238}\text{U}$ ,  $^{234}\text{U}$ , and  $^{235}\text{U}$
- (c) self-induced fission of  $^{235}\text{U}$  and  $^{238}\text{U}$

The neutron source term from spontaneous fission in uranium-based salts will be small because of the long uranium isotope half-lives for spontaneous fission. In fluoride- and chloride-based salts with uranium, some passive neutrons emitted will be from  $(\alpha, n)$  reactions. Figures 4.1 and 4.2 depict the  $(\alpha, n)$  cross sections for the low-Z elements present in the fluoride- and chloride-based fuel salts analyzed in this research. Note that neutrons emitted from  $(\alpha, n)$  reactions in the salt will mostly be less than 10 MeV. The  $(\alpha, n)$  cross sections for the low-Z isotopes in the fluoride-based salt are higher than the cross sections for the low-Z isotopes in the chloride-based salt, leading to higher rates of  $(\alpha, n)$  neutrons being produced in fluoride salts if the uranium mass fractions were the same for the two salts. If  $^{233}\text{U}$  were present in the fresh fuel salt, the  $(\alpha, n)$  neutron source term would be larger, as the  $t_{1/2}$  of  $^{233}\text{U}$  is  $1.592 \times 10^5$  years, which is shorter than  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ .

If the salt is irradiated before entering the reactor system with a neutron source (e.g., AmLi,  $^{252}\text{Cf}$ , AmBe), neutrons, gammas, and x-rays will be emitted from induced fission of  $^{235}\text{U}$  and  $^{238}\text{U}$ .

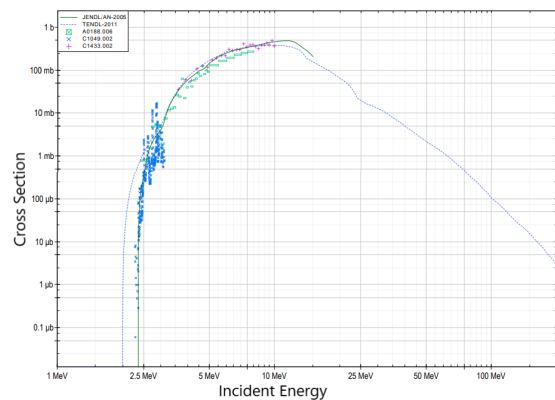
---

<sup>†</sup>Beryllium also undergoes an  $(\alpha, 2n)$  reaction at a threshold energy of 1.851 MeV [58]

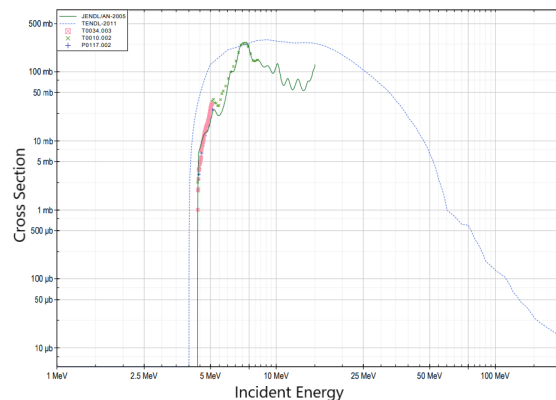
<sup>‡</sup>No spontaneous fission-specific  $t_{1/2}$  was found in the literature for  $^{238}\text{U}$ , but the spontaneous fission yield for  $^{238}\text{U}$  is higher than for  $^{235}\text{U}$  [58], so the  $^{238}\text{U}$  spontaneous fission  $t_{1/2}$  is shorter than that of  $^{235}\text{U}$

Table 4.2: Mass fractions of fresh fuel salts

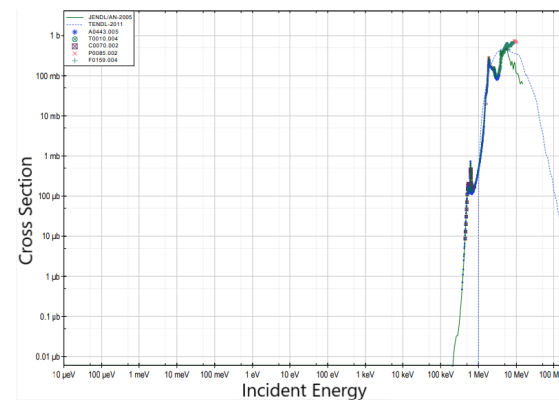
| Element/ Isotope                                                                              | Mass Fraction           |                         |                         |                        |  |
|-----------------------------------------------------------------------------------------------|-------------------------|-------------------------|-------------------------|------------------------|--|
| <i>Fluoride salt: LiF–BeF<sub>2</sub>–UF<sub>4</sub>–ZrF<sub>4</sub> (65–29.1–0.9–5 mol%)</i> |                         |                         |                         |                        |  |
| Total Li                                                                                      | 0.10916                 |                         |                         |                        |  |
| <sup>6</sup> Li                                                                               | 1.092E-5                |                         |                         |                        |  |
| <sup>7</sup> Li                                                                               | 0.10915                 |                         |                         |                        |  |
| Total Be                                                                                      | 0.06278                 |                         |                         |                        |  |
| Total Zr                                                                                      | 0.10918                 |                         |                         |                        |  |
| Total F                                                                                       | 0.66760                 |                         |                         |                        |  |
| Total U                                                                                       | 0.05128                 |                         |                         |                        |  |
| <i>Uranium enrichment</i>                                                                     |                         |                         |                         |                        |  |
|                                                                                               | NU                      | 2 wt%                   | 5 wt%                   | 19.75 wt%              |  |
| <sup>234</sup> U                                                                              | 2.748×10 <sup>−6</sup>  | 9.242×10 <sup>−6</sup>  | 2.310×10 <sup>−5</sup>  | 9.106×10 <sup>−5</sup> |  |
| <sup>235</sup> U                                                                              | 3.70 × 10 <sup>−4</sup> | 1.04 × 10 <sup>−3</sup> | 2.60 × 10 <sup>−3</sup> | 0.01023                |  |
| <sup>238</sup> U                                                                              | 0.05091                 | 0.05023                 | 0.04866                 | 0.04096                |  |
| <i>Chloride salt: NaCl–UCl<sub>3</sub> (66–34 mol%)</i>                                       |                         |                         |                         |                        |  |
| Total Na                                                                                      | 0.09642                 |                         |                         |                        |  |
| Total Cl                                                                                      | 0.38930                 |                         |                         |                        |  |
| <sup>35</sup> Cl                                                                              | 0.09732                 |                         |                         |                        |  |
| <sup>37</sup> Cl                                                                              | 0.29197                 |                         |                         |                        |  |
| Total U                                                                                       | 0.51428                 |                         |                         |                        |  |
| <i>Uranium enrichment</i>                                                                     |                         |                         |                         |                        |  |
|                                                                                               | NU                      | 2 wt%                   | 5 wt%                   | 19.75 wt%              |  |
| <sup>234</sup> U                                                                              | 2.756×10 <sup>−5</sup>  | 9.269×10 <sup>−5</sup>  | 2.316×10 <sup>−4</sup>  | 9.133×10 <sup>−4</sup> |  |
| <sup>235</sup> U                                                                              | 3.70 × 10 <sup>−3</sup> | 0.01041                 | 0.02603                 | 0.10261                |  |
| <sup>238</sup> U                                                                              | 0.51055                 | 0.50378                 | 0.48802                 | 0.41076                |  |



(a)  $^{19}\text{F}$

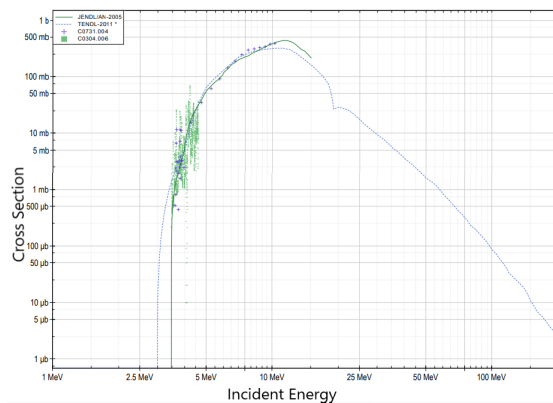


(b)  $^7\text{Li}$

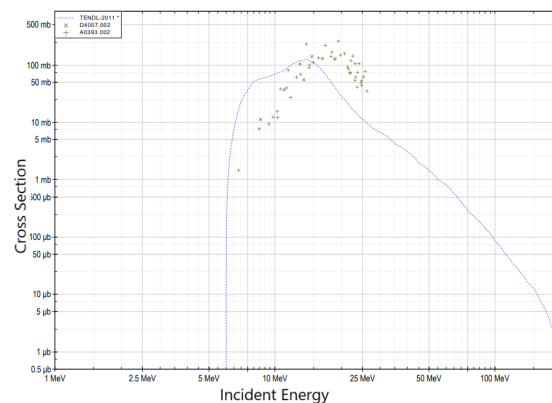


(c)  $^9\text{Be}$

Figure 4.1:  $(\alpha, n)$  cross sections for low-Z elements in FLiBe-based salt [60]



(a)  $^{23}\text{Na}$



(b)  $^{35}\text{Cl}$

Figure 4.2:  $(\alpha, n)$  cross sections for low-Z elements in chloride-based salt [60]

## 4.2 Modeling Passive Radiation Emissions from Fresh Fuel Salt

Several options are available to simulate passive radiation emissions from fresh fuel salt. An appropriate computational tool must account for both decay (i.e., alpha decay, spontaneous fission) and probabilities of  $(\alpha, n)$  reactions. Explicit alpha particle transport is generally computationally expensive, which is limiting when relying on software tools that employ Monte Carlo methods, such as Monte Carlo N-Particle (MCNP). SRIM/TRIM, a charged particle transport code package, is not appropriate for analyzing  $(\alpha, n)$  emissions from salt within a pipe because it can only be used with planar geometries [61]. Although MCNP6 developers have incorporated some improved capabilities related to alpha transport and reliance on  $(\alpha, n)$  cross section libraries, this work used the ORIGEN module within SCALE 6.3.1 and compared the results with SOURCES 4C to generate the simulated passive gamma-ray and neutron emissions from unirradiated molten salt. Both SCALE/ORIGEN and SOURCES 4C have fast computational times and have been well benchmarked (note, to the author’s knowledge, neither of the codes has been benchmarked experimentally against unirradiated molten fluoride or chloride uranium-bearing salt) [52, 62–64]. The computational methodology in SCALE/ORIGEN to generate  $(\alpha, n)$  source terms has been adapted from SOURCES 4B, a previous version of SOURCES 4C [52]. Both codes use a thick-target approximation, which is reasonable in this research application because the radius of the material is much greater than the  $\alpha$  track lengths, and almost all neutrons generated through  $(\alpha, n)$  reactions are from the salt material. Both codes also compute a delayed neutron source term, but this term is negligible in passive emissions from fresh fuel salt because of such low rates of self-induced fission. SCALE/ORIGEN was selected as the primary source of computationally predicted passive emissions from fresh fuel salt because of the convenience of aging the salt to evaluate the buildup of daughters of uranium decay. Because fuel salt components may be combined weeks, months, or even years before being loaded into a reactor as initial or makeup salt, this research explored different times since salt components were integrated. If, for example, a fuel salt synthesis facility were built separately from reactors, similar to how LWRs have separate fuel fabrication facilities,

fuel salt components may be combined and stored in sealed containers before shipment and storage at facilities. Thus, for this work, five distinct times since salt components were combined (i.e., time since salt synthesis) were selected:

1. 1 day
2. 1 week
3. 1 month
4. 1 year
5. 10 years

This allowed for the analysis of build up of uranium daughters to assess whether the time since synthesis would affect the feasibility of different measurement methods. Chapter 5 describes how the passive radiation emissions computed in this work using SCALE/ORIGEN were used as a source term in additional transport simulations to generate expected responses within defined gamma and neutron detectors placed on the outside of salt piping. Research described in Chapter 5 and 6 accounts for attenuation of the radiation within the salt, piping, and, to some extent, surrounding materials. This chapter describes the simulated passive emissions of the zero-dimensional salt to analyze fundamental signature differences between chloride- and fluoride-based salts with different uranium enrichments and times since synthesis. Appendix A includes the SCALE/ORIGEN input files used for these calculations.

#### 4.2.1 SCALE/ORIGEN Passive Neutron Emission Results

Figure 4.3a depicts the calculated total passive neutron emissions from the fluoride-based salt, per gram of salt for different uranium enrichments. Total neutrons include neutrons emitted from  $(\alpha, n)$  reactions and spontaneous fission. There are higher total neutron emissions for higher enrichments because higher enrichments have higher percentages not only of  $^{235}\text{U}$  but also  $^{234}\text{U}$ , which is due to enrichment methods that preferentially select lighter (or smaller, depending on the enrichment method) isotopes. Therefore, when enriching uranium in the isotope 235, the isotopic ratio of 234 is also increased. Uranium-234 has a  $t_{1/2}$  that is more than 3 orders of magnitude shorter than uranium-235. Therefore,  $\alpha$  particles are being emitted from salts with higher enrichments at higher rates, increasing the rate of  $(\alpha, n)$  reactions and producing neutrons at a higher rate from the salt. Figure 4.3b

shows the total passive neutron emissions from the chloride-based salt for different uranium enrichments. These values were qualitatively compared to the results using SOURCES 4C and the shapes of the curves and energy peaks matched for both the fluoride and chloride salts.

There is no significant change in the neutron emissions over time since salt synthesis because none of the uranium daughters are significant contributors to the total neutron population. Figure 4.4 shows total neutron emissions for the fluoride and chloride salts, including both 19.75 wt%  $^{235}\text{U}$  and NU enrichments, for all five times since synthesis.

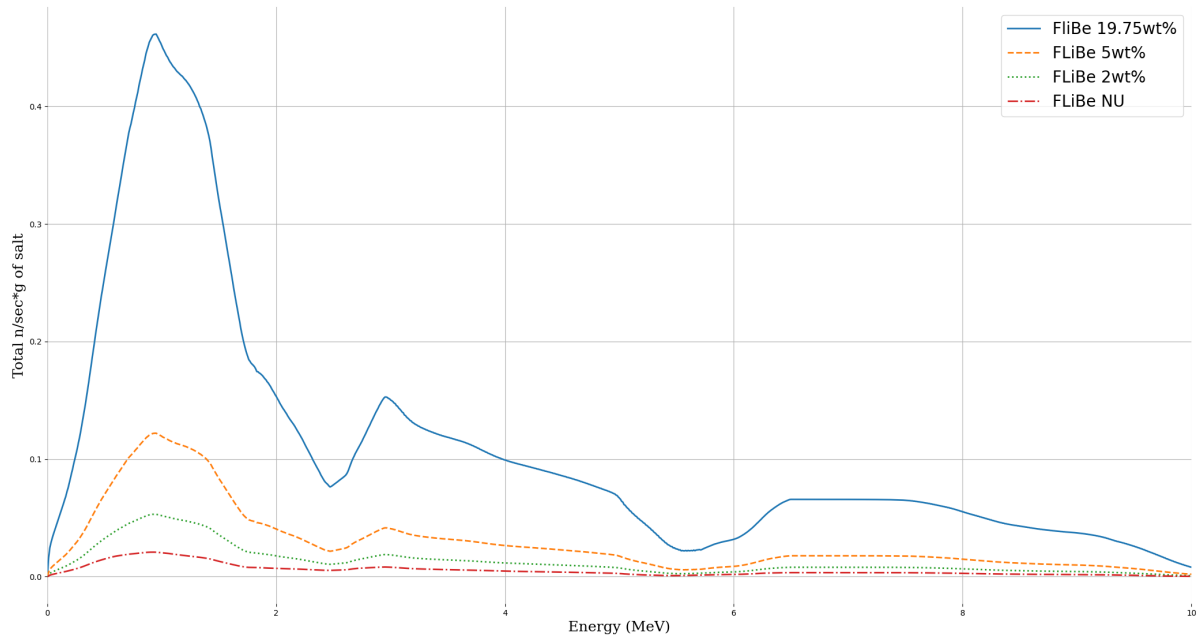
Figure 4.5 shows the difference between the  $(\alpha, n)$  source term versus the total neutrons for each enrichment for both the fluoride- and chloride-based salts. At high enrichments, the majority of total neutrons from both salt types are from  $(\alpha, n)$  reactions. At lower enrichments, a larger fraction of passive neutrons born in fluoride-based salts are from  $(\alpha, n)$  reactions than in chloride-based salts. In chloride salt with uranium of NU, most passive neutron emissions are from spontaneous fission (primarily of  $^{238}\text{U}$ ).

## 4.2.2 SCALE/ORIGEN Gamma-Ray Emission Results

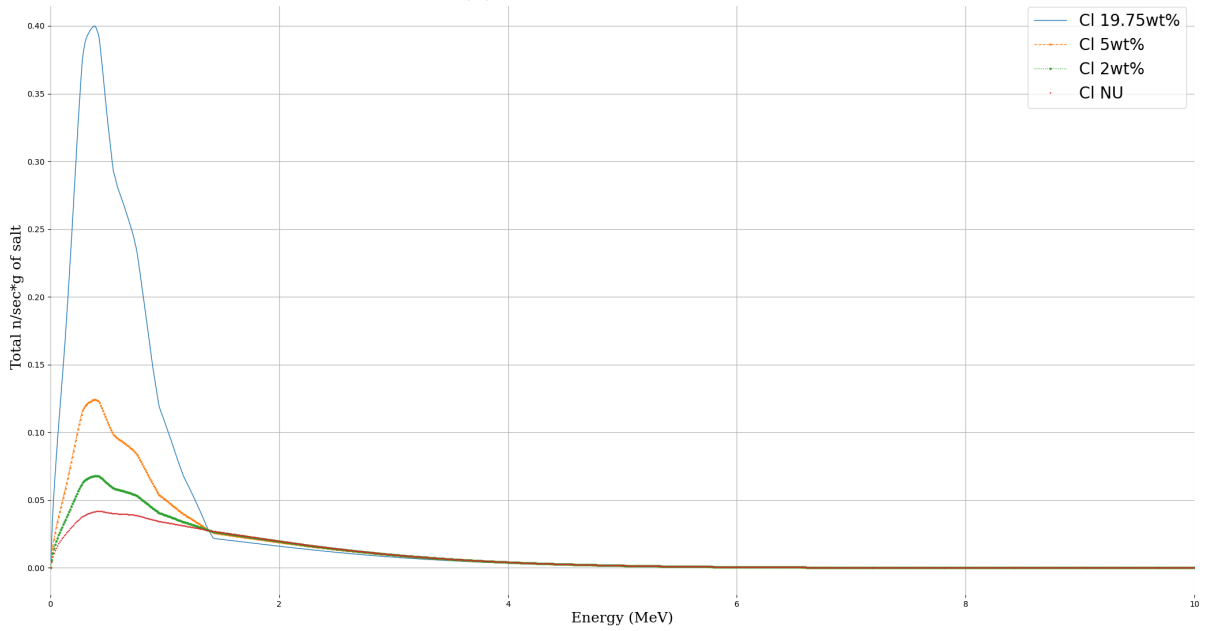
Figure 4.6 depicts the calculated energy-dependent gamma-ray emissions from the fluoride- and chloride-based salts, per gram of salt for different uranium enrichments. There are higher rates of gamma-ray emissions for the chloride salts because of the order of magnitude higher uranium loading in the chloride salt as compared to the fluoride salt. Additionally, higher enrichments will have higher gamma-ray emission rates because of more  $^{235}\text{U}$  and  $^{234}\text{U}$ , which have shorter half-lives than  $^{238}\text{U}$ , and therefore higher  $\alpha$  decay rates with accompanying  $\gamma$  emissions. Different peaks in this figure correspond to the different gamma-ray energies emitted during uranium decay. Chapter 6 includes the analysis of different photopeaks based on simulated results of these gamma-rays from the salt transported through piping and insulation.

Figures 4.7 and 4.8 show the energy-dependent gamma-ray emissions over different times since synthesis for each uranium enrichment. After several months, the uranium daughters reach equilibrium. The emission rates are approximately equal after that time period, so the 1 year and 10 year time since synthesis emission rates are essentially the same. If

salt is synthesized before the time of equilibrium, that will influence the gamma-dependent measurement results unless these differences are accounted for.

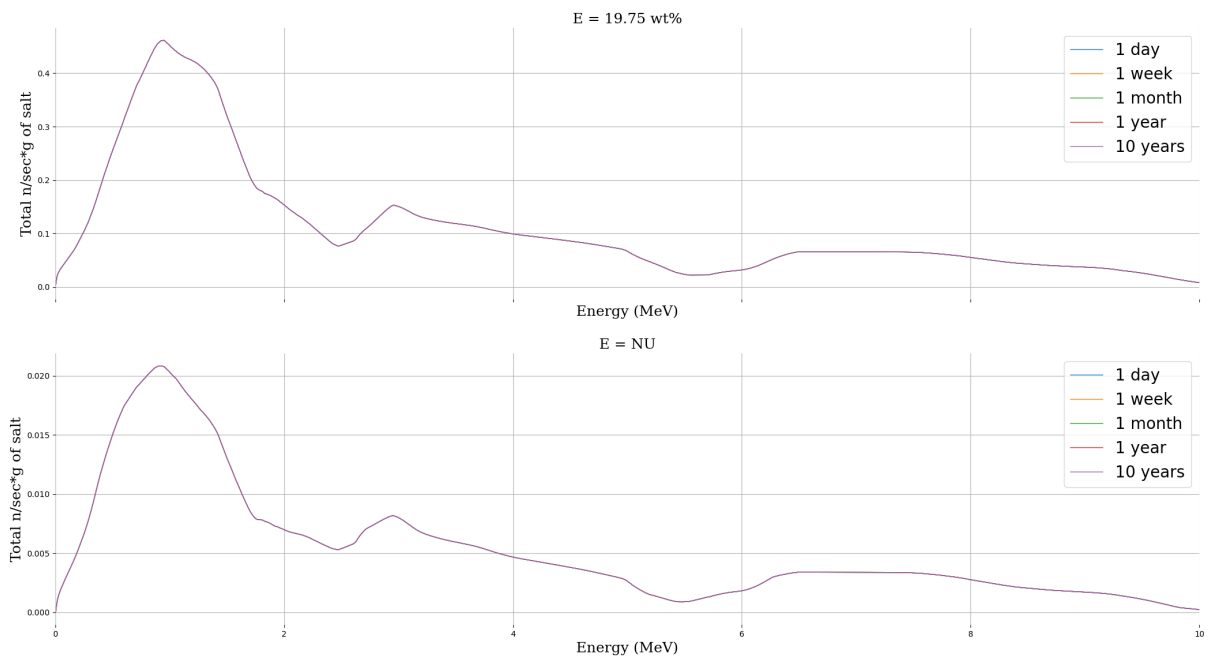


(a) Fluoride-based salt

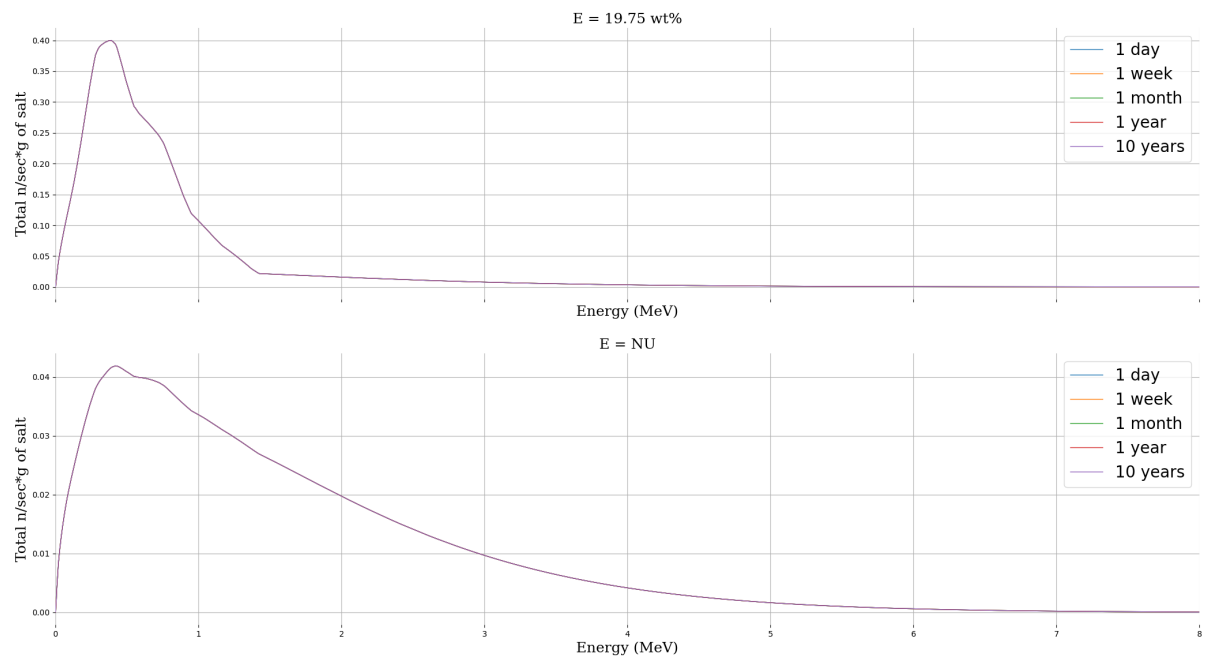


(b) Chloride-based salt

Figure 4.3: Calculated passive total neutron emissions from fluoride- and chloride-based salts over different enrichments using SCALE/ORIGEN



(a) Fluoride-based salt



(b) Chloride-based salt

Figure 4.4: Calculated passive total neutron emissions from fluoride- and chloride-based salts over different times since synthesis using SCALE/ORIGEN

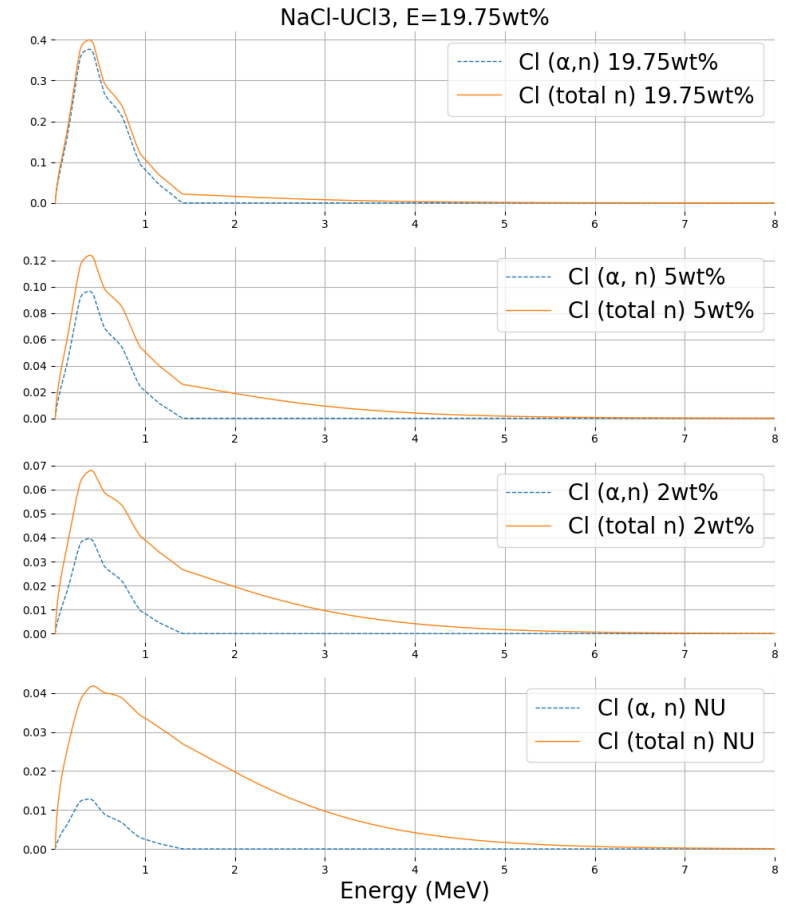
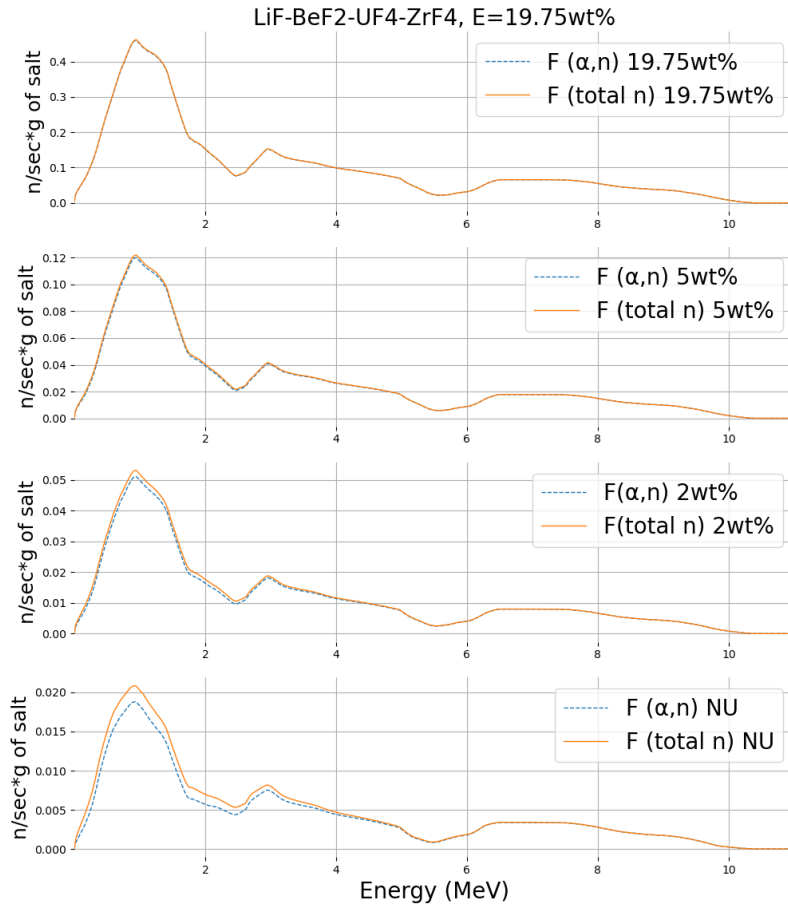


Figure 4.5: Comparison of calculated passive total neutron emissions and ( $\alpha, n$ ) emissions from fluoride- and chloride-based salts with different enrichments using SCALE/ORIGEN

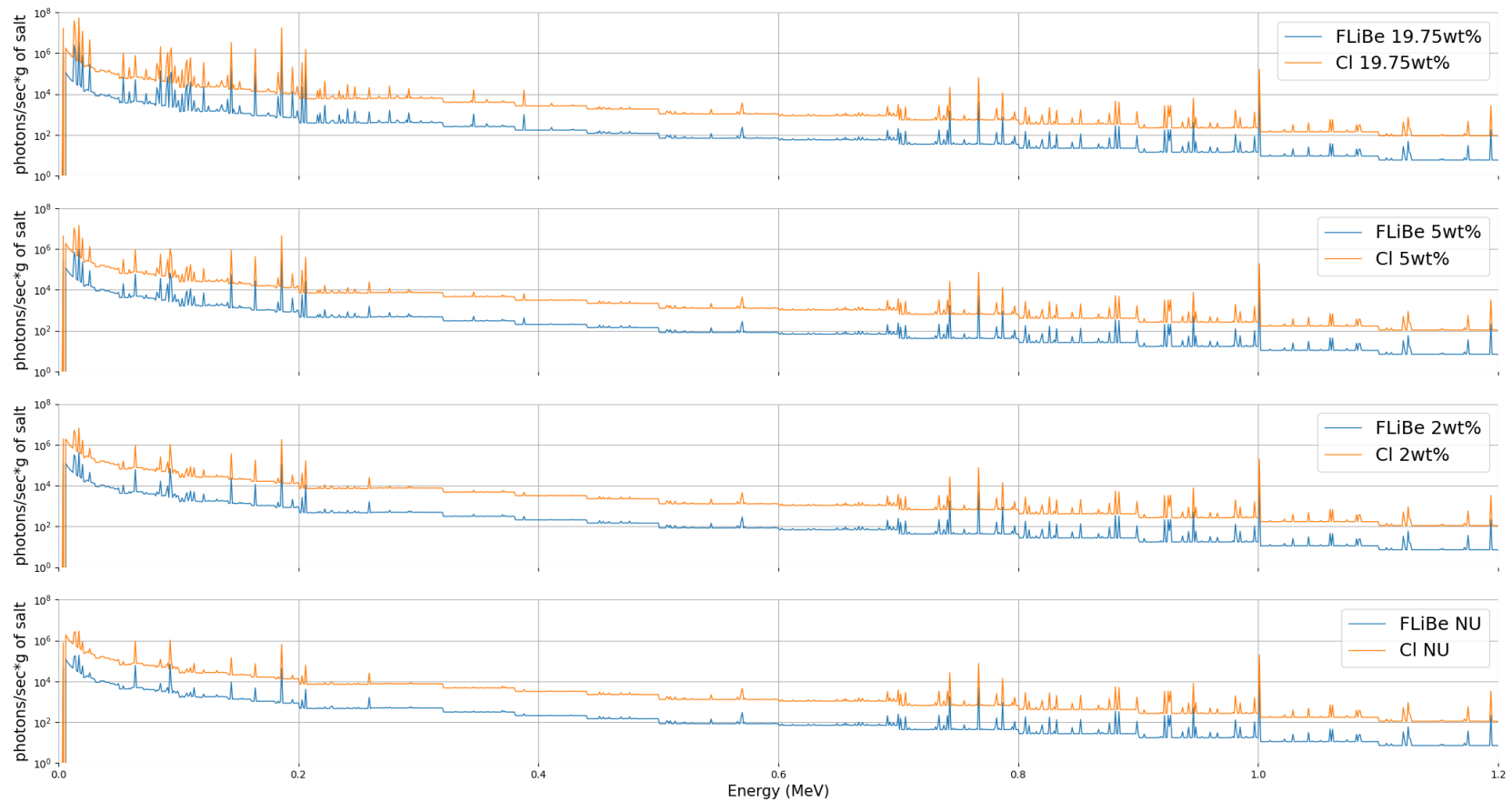


Figure 4.6: Calculated gamma-ray emissions from fluoride- and chloride-based salt over different enrichments from SCALE/ORIGEN, for a 1 year time since synthesis

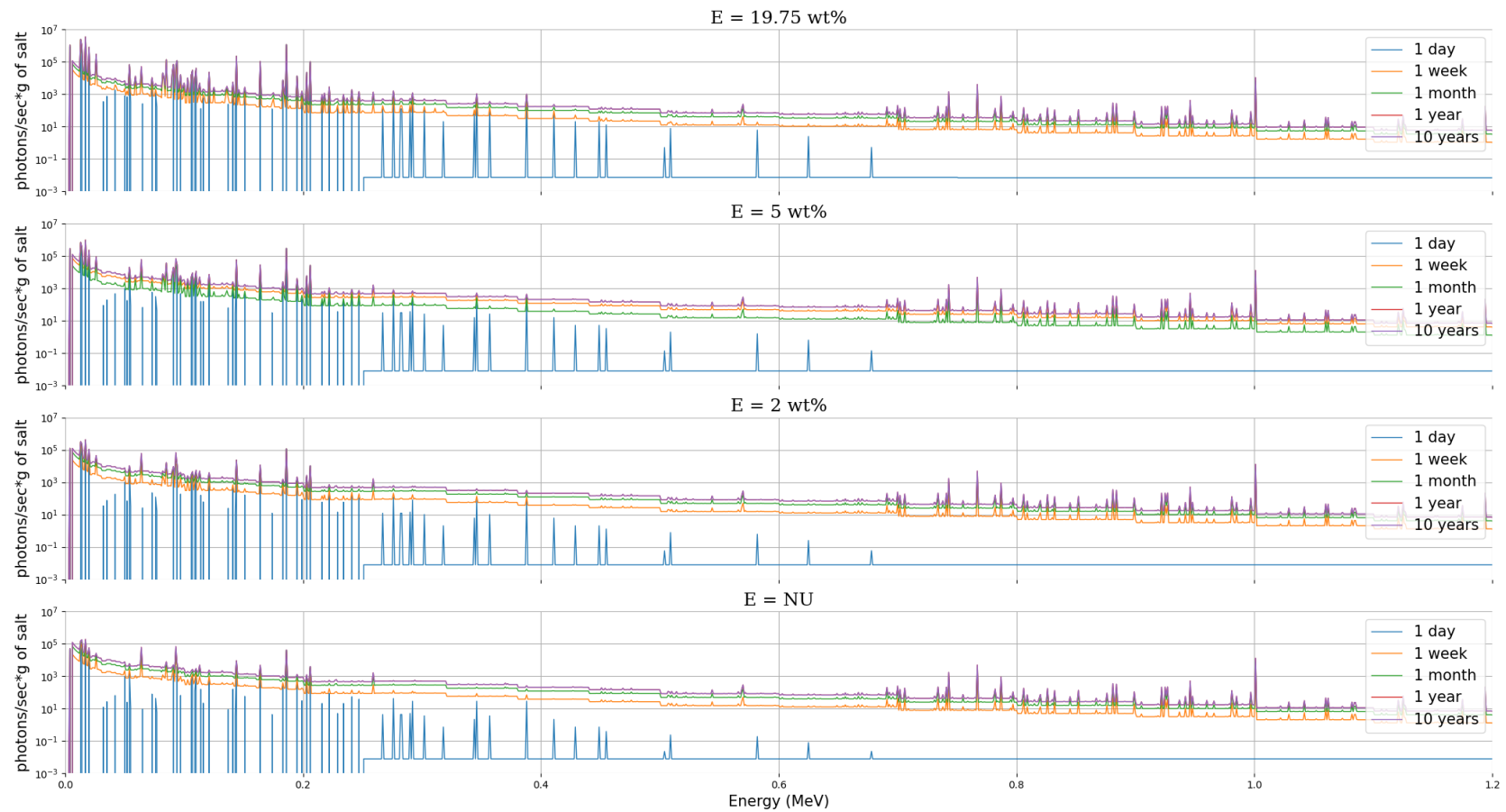


Figure 4.7: Calculated gamma-ray emissions from fluoride-based salt over different enrichments and times since salt synthesis from SCALE/ORIGEN

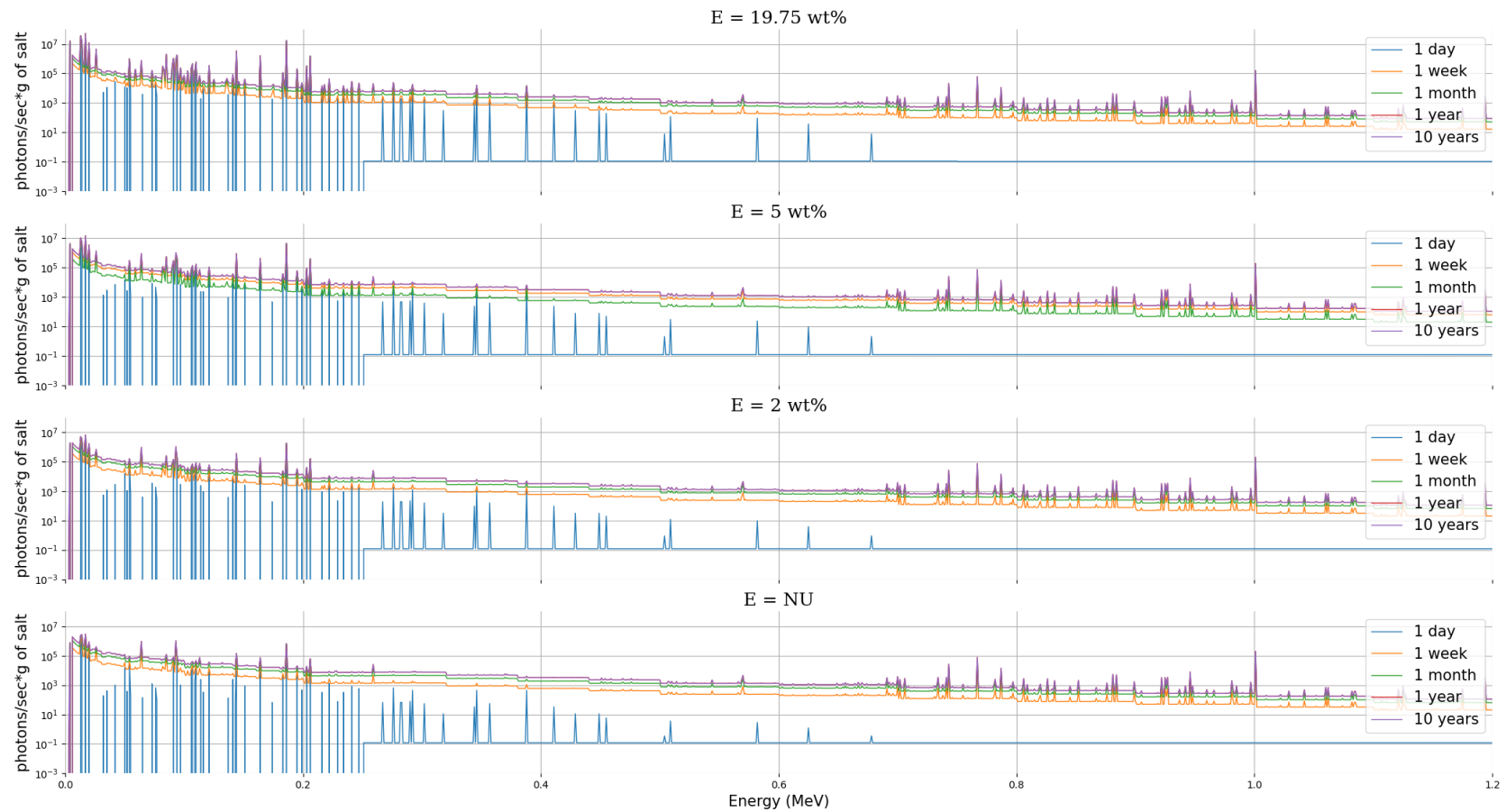


Figure 4.8: Calculated gamma-ray emissions from chloride-based salt over different enrichments and times since salt synthesis from SCALE/ORIGEN

# Chapter 5

## Radiation Signature

## Modeling-Methodology

The photon and neutron emissions from unirradiated fuel salt described in Chapter 4 can be measured and used to determine, or verify, parameters about the fuel salt that are relevant for material accountancy and international safeguards implementation. Limited opportunities currently exist to experimentally measure radiation signatures of fresh fuel salt on the outside of pipes. In the US, some national laboratories and advanced reactor developers do have uranium-bearing salt. However, currently, these are typically small quantities of total salt with only DU or NU and not especially conducive to measurement campaigns with instrumentation on the outside of salt-containing piping. In the US, measurements may be possible on uranium-bearing fluoride-based fuel salt at Abilene Christian University's Next Lab, in preparation for its MSRR, in the next few years [65]. TerraPower LLC will also likely have uranium-bearing chloride-based fuel salt available in flow loops for measurements in the next 1–2 years. Owing to the limited availability of uranium-bearing salt loops, and to best prepare for any future experimental measurement campaigns, this research focuses on a feasibility study to identify promising in situ gamma and neutron detection methods for monitoring unirradiated fuel salt in liquid-fueled MSRs. Of these methods,  $\gamma$ -ray spectrometry with sodium iodide (NaI) and high-purity germanium (HPGe), total neutron counting, passive neutron coincidence counting, and active neutron coincident counting were explored.

Passive gamma spectrometry could potentially be useful in determining parameters related to the  $^{235}\text{U}$  and total uranium quantities in fuel salt added to a liquid-fueled MSR system. First, gamma-ray measurements could be used to determine the uranium enrichment within the fuel salt. Uranium isotopes have distinct gamma-ray energies that can be counted to determine information relevant to material accountancy and safeguards and security applications. The ratio of photopeaks at these specific energies (e.g., 185.72 keV from the alpha decay of  $^{235}\text{U}$  and 1,001 keV from the  $\beta^-$  decay of  $^{234\text{m}}\text{Pa}$ , a  $^{238}\text{U}$  daughter) can be compared to determine the enrichment of the sample being measured without the use of enrichment standards or determination of geometry-dependent calibration constants [58]. Gamma-ray detectors have historically and frequently been used by the IAEA to determine the uranium isotopic assay for verification purposes. Previously, the cascade header enrichment monitor (CHEM) and, more recently, the On-line Enrichment Monitor (OLEM) have been used by the IAEA to verify uranium enrichment of  $\text{UF}_6$  gas in header pipes at gas centrifuge enrichment plants [66]. The NaI-based HM-5 hand held assay probe is used to verify uranium enrichment in fresh fuel assemblies. In the near future, the IAEA is planning to use the high-resolution M400 CdZnTe-based HM-6 for the same purposes. The electrically cooled high-purity germanium system (ECGS) and general enrichment meter (GEM) code are also used by IAEA safeguards inspectors to verify uranium enrichment [66, 67]. If gamma spectrometry was used to determine uranium enrichment in fuel salt, additional measurements would likely be required by the IAEA to fully and independently verify the nuclear material masses in fresh fuel salt. Such additional measurements may include techniques to quantify the uranium concentration within the fuel salt (e.g., HKED, x-ray fluorescence) and total volume (or mass and density) of the fuel salt [68, 69].

Secondly, gamma-ray spectrometry could potentially be used to determine the  $^{235}\text{U}$ ,  $^{238}\text{U}$ , or total uranium mass in the fuel salt. This could be done by measuring the area under the photopeak at gamma-ray energies relevant to the specific isotope in question and knowing the energy-dependent detection efficiency. Determining uranium isotopic masses using gamma spectrometry usually requires correction factors for electronic losses, self-attenuation within the salt, and attenuation within any materials between the salt and the detector (in this research application: pipe wall, insulation, insulation jacket, inert gas) [58]. The IAEA

uses the In Situ Object Counting System (ISOCS) code to calculate the full energy peak (photopeak) efficiencies. Uranium-235 mass can be determined by combining the photopeak efficiencies with the net peak count rate, the gamma branching ratio, and the specific activity of  $^{235}\text{U}$  [70, 71].

Gamma-ray measurements could also potentially be used to determine the relative concentrations of different elements within the salt. Relative concentrations of salt components (e.g., Be, F) could potentially be determined through count rate peaks from the associated  $\gamma$  in  $(\alpha, \gamma n)$  reactions. Although relative concentrations of different elements in fuel salt might be a useful tool to verify the overall salt chemical composition, it may not be as direct of a method to determine the uranium and  $^{235}\text{U}$  mass, which is the ultimate technical goal for MC&A and safeguards at this point within the process stream.

Total neutron counting through passive neutron measurements is useful when one type of neutron source (i.e.,  $(\alpha, n)$ , spontaneous fission, or induced fission) dominates the others [58]. In unirradiated fluoride fuel salts and chloride fuel salts with enriched uranium, the  $(\alpha, n)$  neutron source term dominates the spontaneous fission and induced fission neutron source term, as described in Chapter 4. Therefore, total neutron counting could potentially be useful in determining the  $(\alpha, n)$  source term; which will correlate to the  $^{234}\text{U}$  mass because it is the dominant alpha emitter in all fuel salts with enrichments greater than natural. If the  $^{234}\text{U}$  and  $^{235}\text{U}$  isotopic enrichments can be assumed to be proportional, then the  $^{235}\text{U}$  mass can be indirectly determined through total neutron counting [72]. In some cases, the  $^{234}\text{U}$  and  $^{235}\text{U}$  isotopic enrichments may not be proportional (e.g., if tails from uranium enrichment plants are re-enriched, as indicated as an option in [73]). If these ratios cannot be assumed to be proportional, total uranium mass and  $^{235}\text{U}$  mass can also be determined if enrichment can be verified. Coincidence counting takes advantage of how induced and spontaneous fissions emit multiple neutrons nearly simultaneously (i.e., time correlated), whereas typically one neutron is emitted from  $(\alpha, n)$  reactions (i.e., random in time). Thus, coincidence counting can help determine the mass of the material undergoing fission. In unirradiated, uranium-bearing fuel salt with low spontaneous fission rates, the majority of coincident neutrons are coming from the fission of  $^{235}\text{U}$ . Coincidence counting of passive neutron emissions may be useful in determining fissile (i.e.,  $^{235}\text{U}$ ) mass, if the rate of self-induced fission is high enough

within the salt and the required count times are practical. Total neutron counting is less sensitive to changes in the detector efficiency. However, total counting is more sensitive to background neutrons [58]. At this point in liquid-fuel MSR designs, most developers may not yet have determined all of the practical aspects of how they will feed their initial and makeup fuel salt into their reactor systems. Therefore, it is unclear how much background neutron radiation may be present from other neutron sources around the pipe where fresh feed enters the reactor system.

Active neutron interrogation and coincidence counting may be useful in determining the total fissile material (i.e.,  $^{235}\text{U}$ ) mass in the fuel salt. The IAEA uses active coincidence counting to verify uranium mass in several unirradiated nuclear material applications, including the active well coincidence counter (AWCC) (used by the IAEA to assay uranium in metals, oxides, and fuel pebbles), the uranium neutron coincidence collar, and the fast neutron coincidence collar (both used to determine the linear mass density of uranium in fresh fuel assemblies) [66, 74]. The feasibility of total neutron counting, passive coincidence counting, and active interrogation coincidence counting are all explored in this research.

To determine the feasibility of passive gamma-ray, passive neutron, and active neutron measurements for nuclear safeguards or security applications in monitoring fresh fuel salt in future liquid-fueled MSRs, detectors were modeled on the outside of pipes containing unirradiated fuel salt. A pipe filled with salt was modeled in MCNP 6.2 [75]. Detector responses were captured for multiple potential measurement scenarios. To streamline the process and allow for a parametric study of different salt compositions, detector types and geometries, and pipe geometries and materials, a pipeline was developed to generate and analyze data. Based on user-defined inputs described in Section 5.2, this pipeline is a series of Python scripts that generates source definitions from SCALE/ORIGEN OPUS files for passive gamma and neutron simulations, creates MCNP input decks, parses MCNP output files, and analyzes output data across a variety of input parameters [76]. Figure 5.1 depicts the software tools used and the pipeline flow.

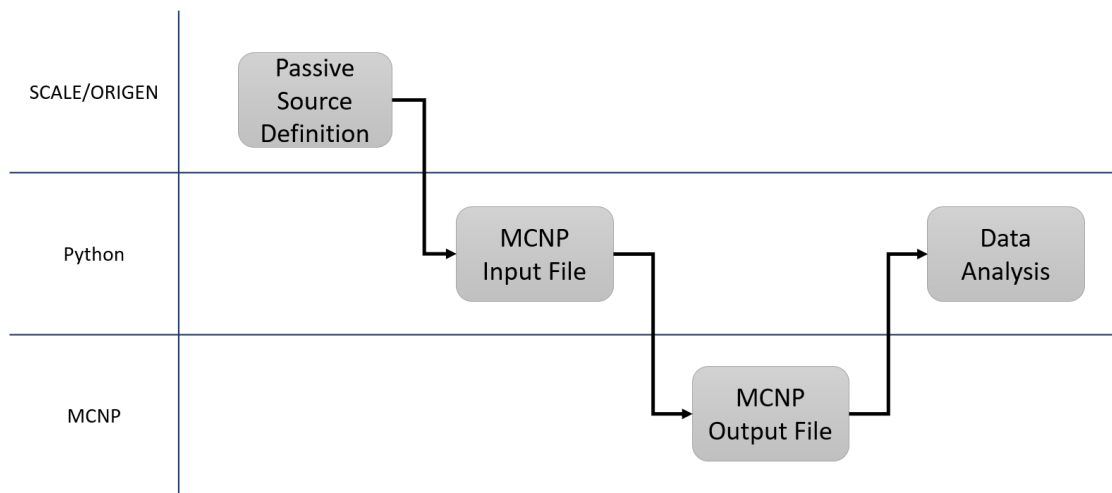


Figure 5.1: Flow between different software packages to generate and analyze data for this research

## 5.1 Modeling Assumptions

This section addresses the assumptions made when developing the model and generating the data through simulations. The models include the assumption that both the fluoride- and chloride-based fuel salts are homogeneous, and that the radiation sources are equally distributed throughout the salt volume within the pipe. There is little in the literature to corroborate or refute this assumption. An assumption of homogeneity is made in other studies incorporating computational modeling on irradiated in-process salt [41, 77]. Experimental validation of this assumption should be done, and verified for different salt synthesis methods and fuel salt chemical compositions. The models also assume there is no difference in the radiation source term at the wall of the pipe vs. at the center of the pipe. That is, there is no distinct layer of uranium-containing material that has plated out on the wall of the pipe. Piping materials are selected because of their resistance to corrosion, and feed piping will likely have significantly less—and potentially only periodic—exposure to the salt (i.e., if makeup salt is added in batches instead of continuously). For these reasons, there may not be as much residual material buildup or corrosion damage to the pipe wall as in piping within the reactor system under irradiation. However, any buildup on the wall would influence the results generated from these simulations. There is little in the literature to determine whether there will be any notable buildup at the pipe wall. Interactions between salts and piping and equipment is an area of active research. However, it is unclear whether any of that analysis will be relevant to piping containing unirradiated fuel salt with only periodic exposure to the salt (e.g., daily, weekly, or even less frequent batches of makeup salt added through piping).

## 5.2 Model Parameters Evaluated

This section describes the parameter options used in simulations. Each of the parameter options were selected to be representative of the liquid-fueled MSR designs that are expected to be deployed in the nearest term, prioritizing designs developed in the US.

### 5.2.1 Salt Composition

Two salt eutectics were selected to represent two salt types in liquid-fueled MSR designs using LEU or HALEU fuel salt: a fluoride-based salt and a chloride-based salt. These two salt options are described in Chapter 4, and the isotopic composition is given in Table 4.1. These salt compositions were selected because the fluoride-based salt was used in the MSRE, and several developers have proposed using a uranium-bearing fluoride (specifically a FLiBe-based salt eutectic). The chloride-based salt is representative of TerraPower’s MCFR and MCRE designs.

Four uranium enrichments were analyzed: NU, 2 wt%  $^{235}\text{U}$ , 5 wt%  $^{235}\text{U}$ , and 19.75 wt%  $^{235}\text{U}$ . Some developers, such as TerraPower, have previously suggested they may use NU as makeup fuel salt in their MCFR. Using NU requires no enrichment, which significantly reduces both costs and proliferation risks related to the unirradiated fuel. The 2 wt%  $^{235}\text{U}$  and 5 wt%  $^{235}\text{U}$  enrichments are representative of enrichment levels developers planning to use LEU are proposing. Multiple liquid-fueled MSR developers have proposed using LEU for reasons including current limited access to HALEU. The 19.75 wt%  $^{235}\text{U}$  was selected to represent HALEU, which is proposed by multiple developers, including fast reactors as initial fuel salt. In general, reactor size will be significantly smaller for the same power output if 19.75 wt%  $^{235}\text{U}$  were used over lower enrichments. Fast reactors benefit from this especially because of the lack of a neutron moderator. However, sources of HALEU are currently extremely limited and HALEU introduces high proliferation risks as the frequency of safeguards inspections may increase in facilities with HALEU to meet IAEA timeliness goals associated with detecting diversion.

The salt densities used in the model were  $2.14647\text{ g/cm}^3$  for the  $\text{LiF–BeF}_2\text{–UF}_4\text{–ZrF}_4$ , based on the ORNL technical manual describing the salt used in the MSRE at 648 C, which is above the melting temperature for the salt [53]. The density used in models for the  $\text{NaCl–UCl}_3$  was  $3.3\text{ g/cm}^3$  [78]. Other publications use higher values for  $\text{NaCl–UCl}_3$  density at the composition (66–34 mol%) and temperature (600 °C) used in this model [79, 80]. Density measurements of many proposed salts, including  $\text{NaCl–UCl}_3$ , are an active area of research.

### 5.2.2 Pipe Material

Two piping material options were selected for this model, depending on the type of salt used. Both of the piping materials used in this model are based on practical selections that advanced reactor developers and the commercial nuclear utilities may plan to use in the future: Hastelloy N alloy (UNS N10003) is modeled with the fluoride-based salt and Inconel 625, a nickel–chromium alloy (UNS N06625/W.Nr. 2.4856), is modeled with the chloride-based salt. Hastelloy N has good oxidation resistance to hot fluoride salts in the temperature range that fresh fuel salt would be heated to. (The melting point of the  $\text{LiF–BeF}_2\text{–UF}_4\text{–ZrF}_4$  salt used in the MSRE is  $449^\circ\text{C}$  and fuel salt entered the MSRE vessel at  $635^\circ\text{C}$  [81, 82].) Hastelloy N was used in the MSRE during operation. Inconel 625 was selected because of its corrosion resistance and range of service temperature (up to  $982^\circ\text{C}$ ) [83]. However, liquid-fueled MSR developers or operators might use Inconel 600 (used in ORNL’s Liquid Salt Test Loop), Hastelloy C-276 (used in ORNL’s Facility to Alleviate Salt Technology Risks (FASTR), or stainless steel alloy 316H [84]. Stainless steel alloy 316H is already currently code qualified under the American Society of Mechanical Engineers (ASME), which would significantly reduce costs and risk; however, it is not ideal for halide services [85]. Table 5.1 includes the chemical compositions defined in specification sheets, as well as used in each of the pipe materials used in models for this research.

Pipe densities were only available at room temperature from material data sheets. The following densities were used in the model: Hastelloy N:  $8.86\text{ g/cm}^3$  and Inconel 625:  $8.44\text{ g/cm}^3$ . The densities of these metal alloys may vary at higher temperatures.

### 5.2.3 Pipe Dimensions

A pipe segment of 100 cm length was selected for this research with varying outer diameter and thickness. The pipe was enclosed in a cylinder of inert gas (argon). No other external equipment or components (e.g., floor, walls, tanks, shielding) were included in the model at this time because this is a focused feasibility study, so ideal conditions with no environmental background were assumed. Additionally, realistic surroundings are unknown at this time because it is too early in the design process to estimate the physical layout and practical

Table 5.1: Chemical composition of Hastelloy N and Inconel 625 pipe materials

| Element    | Z  | Hastelloy N           |                              | Inconel 625           |                              |
|------------|----|-----------------------|------------------------------|-----------------------|------------------------------|
|            |    | Weight % <sup>1</sup> | Weight% Modeled <sup>2</sup> | Weight % <sup>1</sup> | Weight% Modeled <sup>2</sup> |
| Nickel     | 28 | 71.00<br>balance      | 68.77                        | 58.00 min             | 57.38                        |
| Chromium   | 14 | 7.00                  | 7.00                         | 20.00–23.00           | 23.00                        |
| Molybdenum | 42 | 16.00                 | 16.00                        | 8.00–10.00            | 9.00                         |
| Iron       | 26 | 5.00 max              | 4.95                         | 5.00 max              | 4.00                         |
| Silicon    | 14 | 1.00 max              | 1.00                         | 0.50 max              | 0.25                         |
| Manganese  | 25 | 0.80 max              | 0.80                         | 0.50 max              | 0.50                         |
| Carbon     | 6  | 0.08 max              | 0.08                         | 0.10 max              | 0.10                         |
| Cobalt     | 27 | 0.20 max              | 0.20                         | 1.0 max               | 1.0                          |
| Aluminum   | 13 | (0.35 max)            | 0.15                         | 0.40 max              | 0.30                         |
| Titanium   | 22 |                       | 0.20                         | 0.40 max              | 1.0                          |
| Niobium    | 41 |                       |                              | (3.15–4.15)           | 2.03                         |
| Tantalum   | 73 |                       |                              |                       | 2.12                         |
| Copper     | 29 | 0.35 max              | 0.35                         |                       |                              |
| Tungsten   | 74 | 0.50 max              | 0.50                         |                       |                              |
| Sulfur     | 16 |                       | 0.015 max                    | 0.0003                |                              |
| Phosphorus | 15 |                       | 0.015 max                    | 0.015                 |                              |

<sup>1</sup>As defined by the material data sheet [83, 86]

<sup>2</sup>Used in MCNP materials definition

<sup>3</sup>Maximum value for Al and Ti combined

<sup>4</sup>Range for Nb and Ta combined

scenarios related to feeding material in future demonstration or commercial liquid-fueled MSR facilities.

Multiple pipe outer dimensions were modeled, including 2.54, 5.08, 10.16, 20.32, and 30.48 cm. Pipe thicknesses of 0.254, 0.625, and 2.54 cm were modeled to explore the sensitivity of different radiation signatures from various pipe dimensions. In reality, pipe dimensions in molten salt loop test facilities are based on American National Standards Institute (ANSI) pipe schedules. The modeled combinations of pipe outer diameter and pipe thickness are not explicitly based on pipe schedules, but were selected to represent the range of reasonable outer dimensions and thicknesses, as well as explore bounding cases to determine their effects on the ability to use measurements techniques to monitor nuclear material within fresh fuel salt.

#### **5.2.4 Insulation**

Because the melting temperatures of molten fuel salts are high, insulation material and aluminum pipe insulation jacketing sheets are used in molten salt test loops. The insulation and aluminum jacketing used in these models is based on that used in ORNL's FASTR chloride molten salt loop. It includes 5.08 cm of silica blanket type SB-2000 insulation and 0.04064 cm of aluminum jacketing [85, 87, 88]. Table 5.2 includes the chemical composition of the insulation material.

#### **5.2.5 Detectors**

The wide variation in proposed fuel salt chemical compositions,  $^{235}\text{U}$  enrichments, pipe dimensions, practical aspects of adding unirradiated fuel salt, and pipe materials will affect the radiation emitted and optimal detector type and geometry for detecting that radiation. Although future detectors could be optimized for specific facilities, it would be resource intensive to optimize detectors for each proposed liquid-fueled MSR design at each projected measurement point. At this point in time, it is unknown which of the many MSR (or advanced reactor, in general) designs might actually make it to operation and require MC&A. The goal of this work is to determine promising measurement technologies that could

Table 5.2: Chemical composition of insulation

| <b>Component</b>               | <b>Wt %<sup>1</sup></b> | <b>Wt %<br/>Modeled<sup>2</sup></b> |
|--------------------------------|-------------------------|-------------------------------------|
| SiO <sub>2</sub>               | 95–97                   | 97                                  |
| Al <sub>2</sub> O <sub>3</sub> | 2–4                     | 3                                   |
| Other Oxides                   | <1                      | 0.0                                 |

<sup>1</sup>As defined by ZRCI Refractory Composites [87]

<sup>2</sup>Used in MCNP materials definition

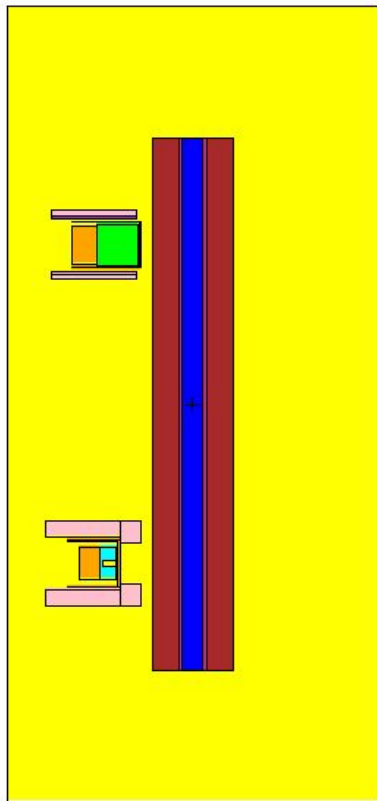
potentially be used for multiple liquid-fueled MSR designs such that any recommended design changes by liquid-fueled MSR developers can be implemented early in the design process (i.e., safeguards and security by design). Additionally, design of technologies for material accountancy can progress without being too far behind to be useful if and when future liquid-fueled MSR facilities become operational and require MC&A implementation. For these reasons, detector designs were selected for modeling based on existing designs that have already been approved for use by the IAEA Department of Safeguards and used in operational facilities. These detectors and the parameters used in modeling are described in Sections 5.2.5 and 5.2.5.

### **Gamma Detectors**

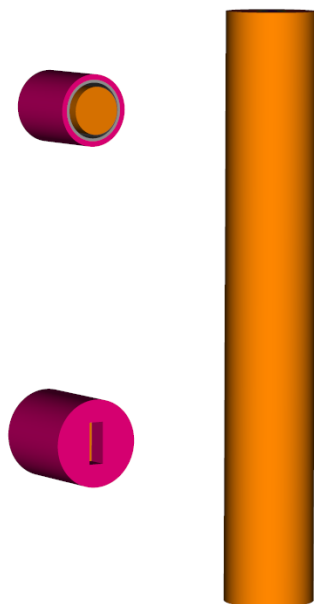
Two gamma detectors were modeled on the outside of the pipe to determine the feasibility of using either a NaI scintillation detector or HPGe detector. These two selections allow comparison of two detector types used by the IAEA and industry for material accountancy with notably different energy resolutions. Figure 5.2 shows the two detectors modeled outside of the pipe.

The NaI spectrometer modeled is a 7.62 cm diameter by 7.62 cm height NaI crystal. Model parameters generally match those described in [89]: 5 mm aluminum can, low-density ( $0.5 \text{ g/cm}^3$ ) aluminum volume behind the crystal that approximates the photomultiplier and tube base, 1.0 cm tungsten collimator lined with 0.5 cm of iron. This detector is based on the continuous enrichment monitor (CEMO), which was previously used to monitor  $\text{UF}_6$  in enrichment plants [90]. Figure 5.3 shows the MCNP model of the NaI detector.

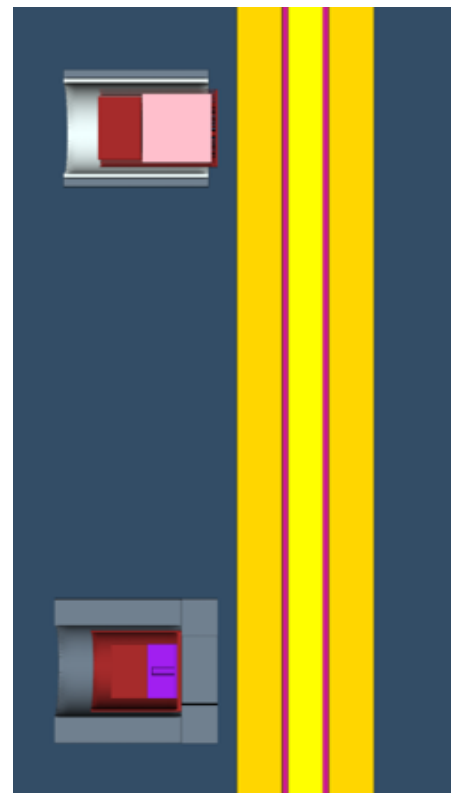
The HPGe detector model is based on the IAEA's CHEM, which was also used in enrichment plants to monitor  $\text{UF}_6$  enrichment. Model parameters generally follow those described in [89]: a small coaxial HPGe crystal with a diameter of 6.0 cm and a thickness of 3.0 cm, 2.5 mm dead layer, 9 mm diameter inner bore, 5 mm aluminum can, a low-density ( $0.5 \text{ g/cm}^3$ ) aluminum volume behind the crystal that approximates the support structure and preamplifier, and a tungsten collimator with a slit 7.5 cm high, 3 cm wide, and 4 cm deep. Figure 5.4 shows the MCNP model of the HPGe detector.



(a) 2 cm standoff distance



(b) 50 cm standoff distance



(c) Cross section of detectors and piping

Figure 5.2: NaI and HPGe detectors modeled on the outside of a salt-filled pipe surrounded by insulation and aluminum jacketing

Standoff distances of 2 and 50 cm were modeled to determine the effects of a larger distance between the pipe and the detector face. This distance was modeled as the gap between the outermost material (e.g., if insulation and aluminum jacketing were modeled, then the distance between the insulation jacket and the outermost face of the detector casing). Argon was modeled between the pipe (or insulation) and the detector.

Simulations were run in MCNP with  $1 \times 10^8$  particles and F8 tallies, with and without Gaussian energy broadening (GEB) corrections. GEB values were assumed and not experimentally determined. GEB values for the HPGe detector modeled were based on an energy resolution of approximately 0.6 keV at 185 keV, which is the full width at half maximum (FWHM) value for a detector with the same size HPGe crystal. The GEB values used for the NaI detector were based on the similar modeled detector described in [89].

## Neutron Detector

Fresh fuel materials typically do not have a strong enough neutron signature to use passive neutron measurements to quantify  $^{235}\text{U}$ , but liquid-fueled MSR fuel salts are distinct from uranium oxide fuel. The unattended cylinder verification station (UCVS) and passive neutron enrichment monitor (PNEM) both use  $(\alpha, n)$  signatures from the  $^{234}\text{U}$   $\alpha$  decay and low-Z fluorine absorption to infer the  $^{235}\text{U}$  mass in  $\text{UF}_6$  cylinders for IAEA verification purposes and could be built upon to develop systems for monitoring nuclear material in salt [91]. This research explores the feasibility of both passive neutron measurements and active neutron measurements on unirradiated fuel salt in piping.

For this feasibility study, a collar detector was modeled that encircles the fuel salt. For passive neutron simulations, a full collar detector was modeled around the piping. For active neutron simulations, only half of the collar was modeled around the piping, and a neutron source described in Section 5.2.6 was placed opposite the detector, inside the insulation 1 cm outside of the pipe wall. Figure 5.5 depicts the passive neutron measurement model. Figure 5.6 depicts the active neutron measurement model for both a standard and large diameter pipe. A full collar for active interrogation was evaluated in Chapter 7 but was not found to influence the feasibility of the detection technique. The collar detectors modeled

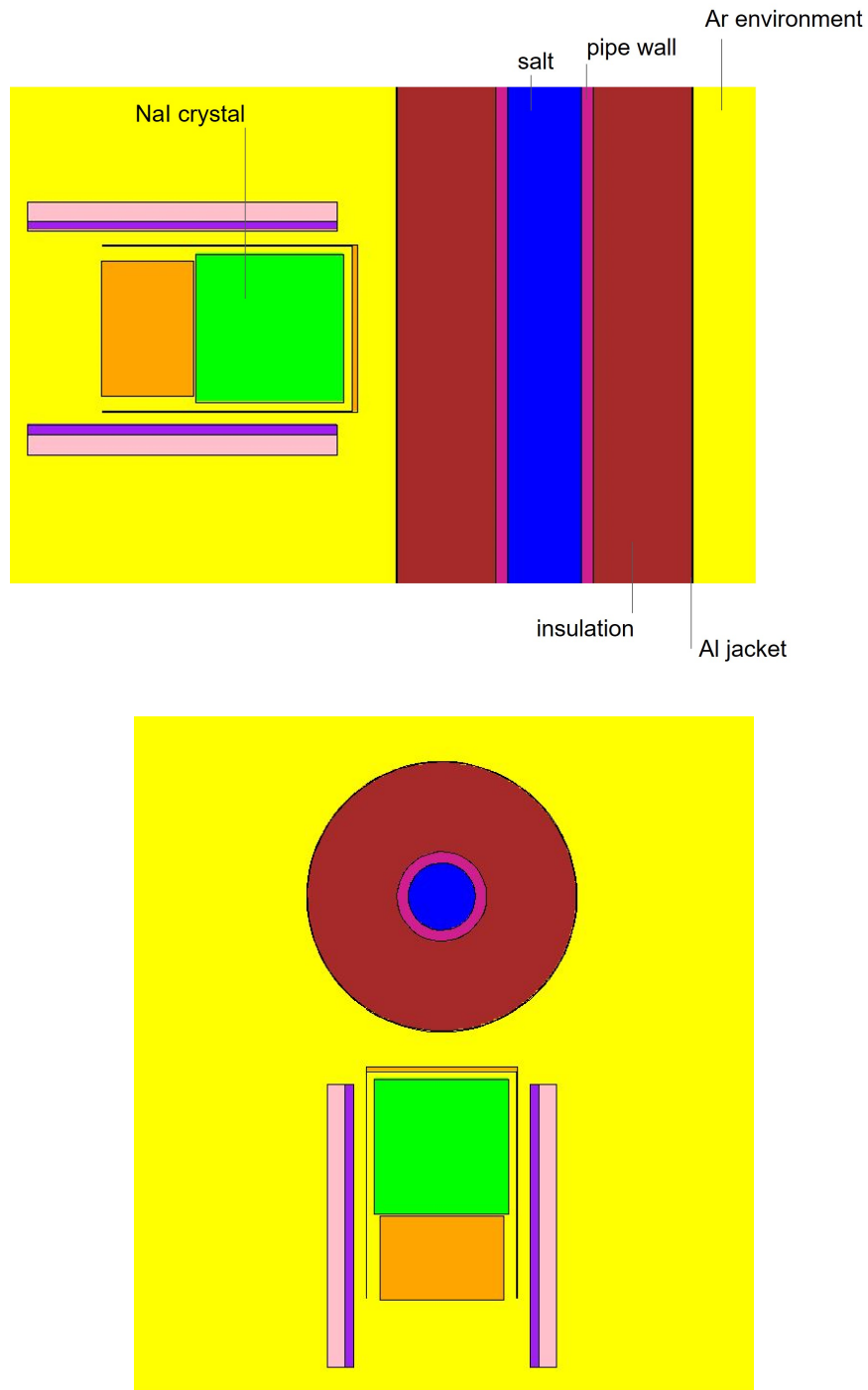


Figure 5.3: NaI detector modeled outside of a salt-filled pipe surrounded by insulation

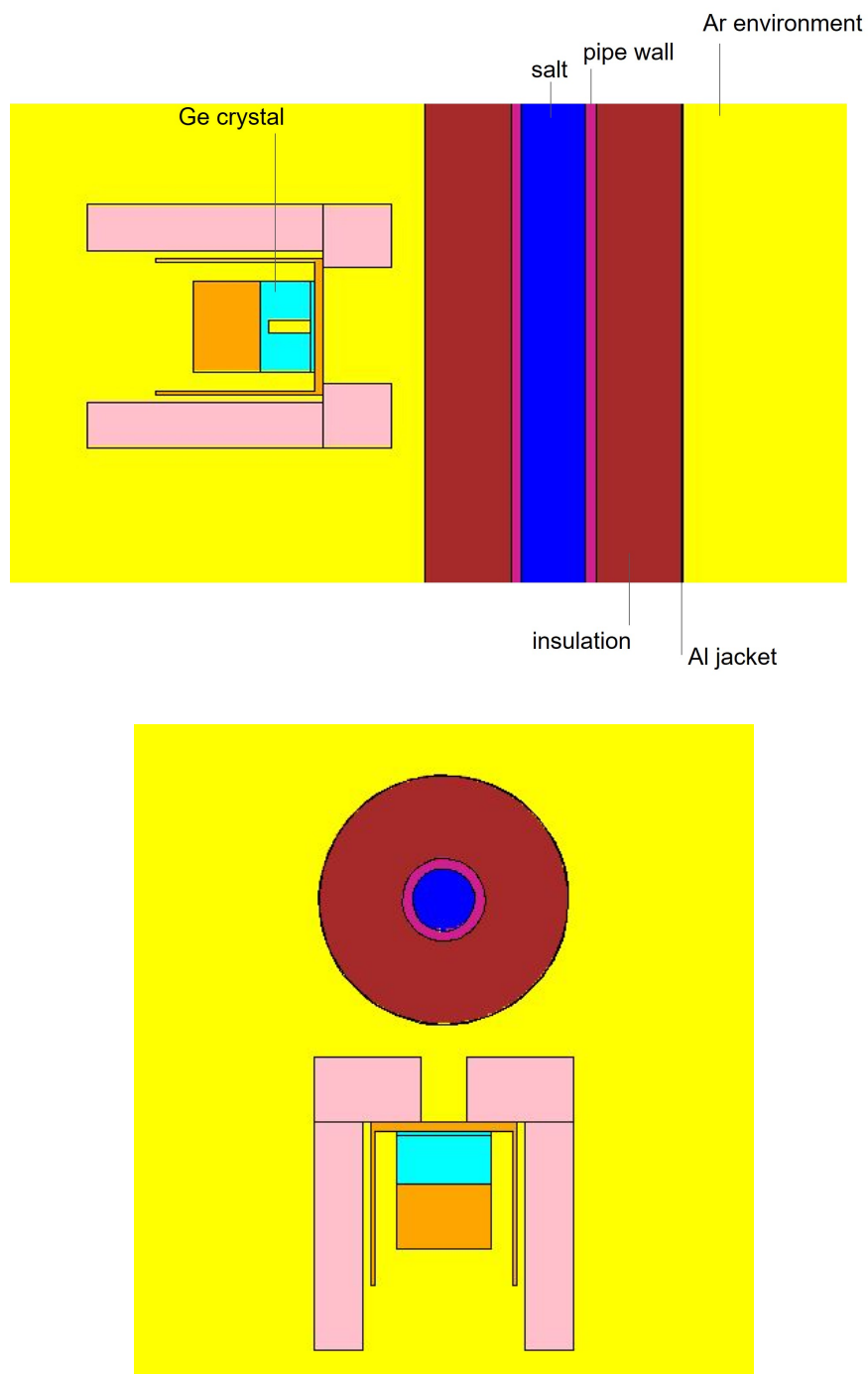


Figure 5.4: HPGe detector modeled outside of a salt-filled pipe surrounded by insulation

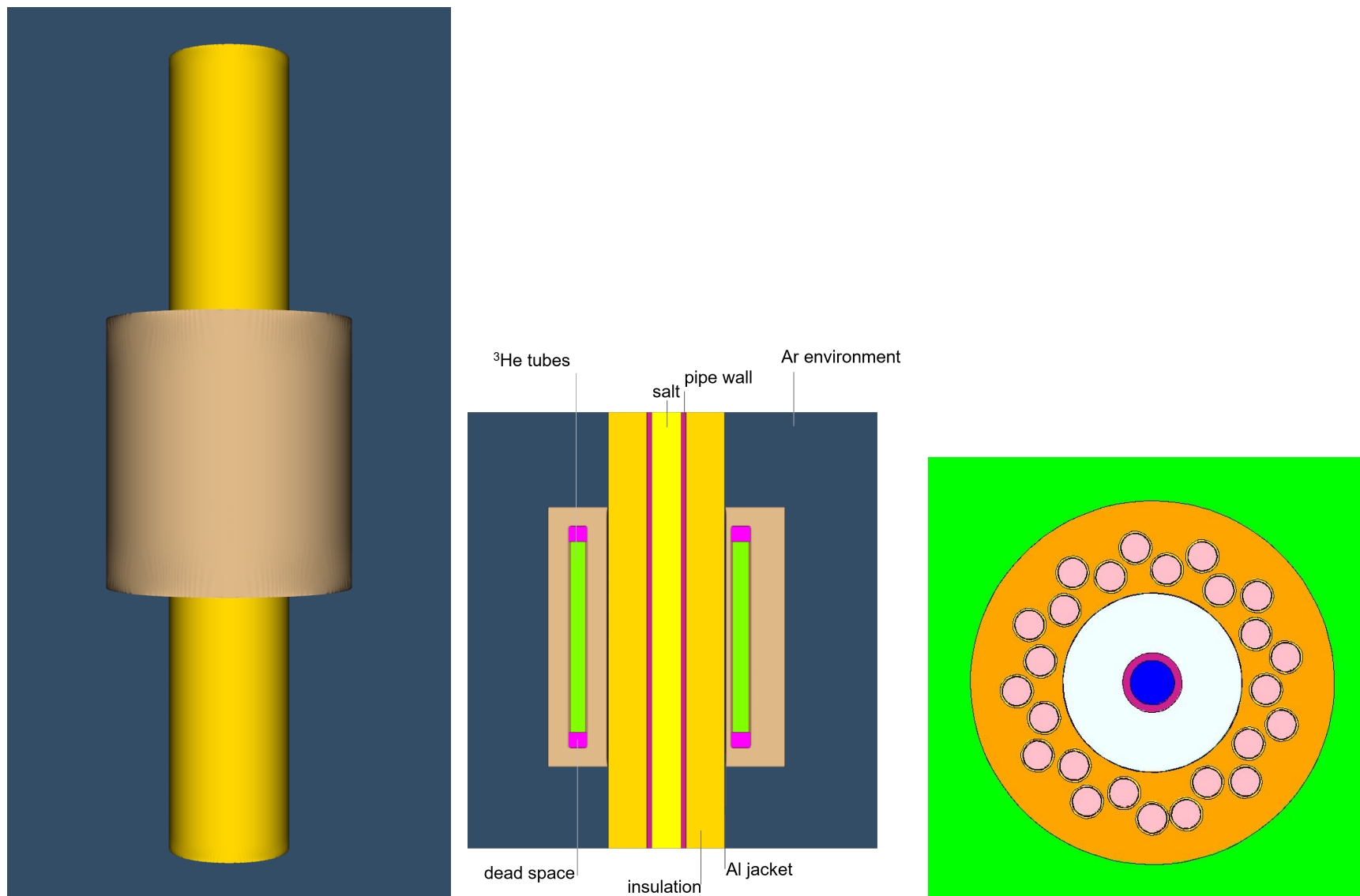
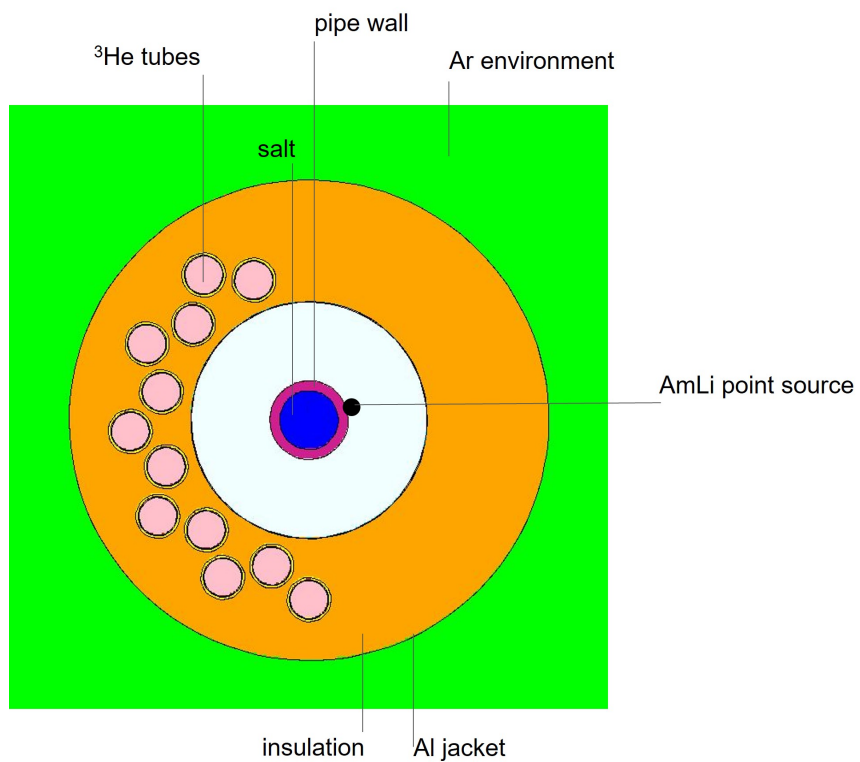
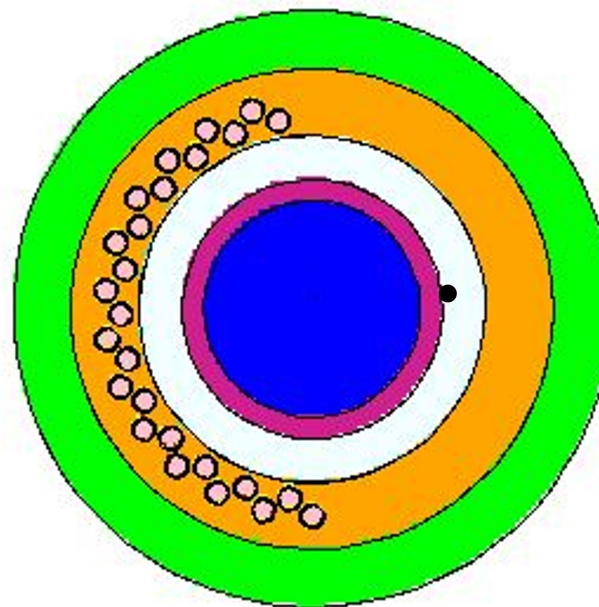


Figure 5.5: Helium-3 collar detector for passive neutron detection



(a) Standard diameter pipe

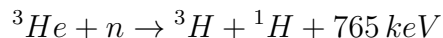


(b) Large diameter pipe

Figure 5.6: Helium-3 half-collar detector for active neutron detection

are based on the AWCC, which is a well detector and an IAEA Authorized Safeguards Equipment. The collar detector modeled surrounds the pipe and has a variable number of  $^3\text{He}$  tubes, depending on the pipe and insulation dimensions such that the number of tubes increases with larger outer diameter pipes and decreases with smaller diameter pipes.

Helium-3 is commonly used in gas-filled thermal-neutron detectors for safeguards applications [66]. These detectors use the following nuclear reaction for detection:



In this model, the  $^3\text{He}$  tubes are oriented in two rings: an inner ring and outer ring, similar to the AWCC. Each  $^3\text{He}$  tube has an inner radius of 1.2192 cm and has a 0.0508 cm thick stainless steel tube wall. A  $1 \times 10^{-8}$  cm thick magnesium coating is modeled on each tube wall, and a dead space is modeled at the top and bottom of each tube. The number of  $^3\text{He}$  tubes was varied based on the circumference of the detector, divided by the diameter of the tubes, alternating the placement of the tubes between an inner and outer ring within the high-density polyethylene (HDPE). A neutron moderator is necessary to reduce the incident neutron energies to increase the probability of  $^3\text{He}$  capture and thus increase the detection efficiency of neutrons from the salt. Thermal neutron detectors, including in the AWCC, often use HDPE. However, HDPE has a melting temperature of approximately 135 °C [92]. Therefore, enough insulation needs to be added between the hot pipe containing greater than  $\approx 500$  °C temperature salts and the HDPE, forced convection for cooling must be employed, or other moderating materials with higher melting temperatures (e.g., graphite) must be employed. This research explored the feasibility of using a HDPE-moderated detector on the outside of insulation and aluminum jacketing. The assumption for this research is that the 5.08 cm of insulation and aluminum jacketing modeled, as described in Section 5.2.4, reduces the temperature to less than the melting temperature of HDPE. This is based on the approximate thickness and type of insulation used in ORNL molten salt test loops, as provided in personal communication by the Molten Salt Test Loop Laboratory Manager, Dr. Kevin Robb. For larger pipe diameters, this amount of insulation may be insufficient to

reduce the temperatures below melting temperatures of HDPE; however, Chapter 6 describes that the insulation blanket material does not notably affect the singles or doubles count rates.

The cross section for the  $^3\text{He}$  neutron capture reaction is strongly dependent on incident neutron energy, thus the efficiency of the neutron detector will vary with the thickness of the moderator (HDPE), as well as the source and detector geometry. Because this research looks at multiple salt compositions and pipe and detector geometries, the HDPE thickness was not optimized for each case. This is because the purpose of this feasibility study was to evaluate monitoring technologies based on existing, IAEA-approved equipment that represents “off-the-shelf” systems. For this feasibility study, 0.635 cm of HDPE (based on AWCC HDPE thickness) was modeled between the inner ring of  $^3\text{He}$  tubes and the aluminum jacket surrounding the insulation, pipe, and fuel salt. If neutron detector designs are pursued in the future for MC&A purposes, detector geometry—including the thickness of any moderator, number, size and placement of  $^3\text{He}$  tubes, should be investigated further.

For passive simulations,  $1 \times 10^6$  particles were run in MCNP6.2 simulations. For active simulations,  $1 \times 10^8$  particles were run in MCNP6.2. F8 capture tallies, summing the counts across all  $^3\text{He}$  tubes, were used to generate the singles rate for total counts and the doubles rate for coincidence counting applications. This methodology is consistent with [93]. An ungated coincidence tally was used and compared to the first moment of a long gate coincidence tally (time delay of 0 and gate width of  $4,000 \mu\text{s}$ ). The first moment of a gated coincidence tally was used to determine the doubles rate (time delay of  $4.5 \mu\text{s}$  and gate width of  $64 \mu\text{s}$ ). This time delay and gate width are standard for the IAEA AWCC.

### 5.2.6 Source Definitions

For passive measurement simulations, the source definitions were based on SCALE/ORIGEN output files, with energy bins for gammas and neutrons defined as shown in Appendix A. A Python script was created to read in ORIGEN opus files, normalize the resulting particles per second, and format into SDEF files for MCNP, with an energy distribution followed by a probability distribution. For active measurement simulations, the source definition is an AmLi point source with an energy and probability distribution based on the source definition used for the AWCC.

# Chapter 6

## Radiation Signatures

### Modeling—Results

This chapter describes the results of the modeling simulations described in Chapter 5. A sensitivity study was performed to determine the impact of MSR design parameters on the feasibility of use of different detector systems. Impacts of the following design parameters (discussed in Section 5.2) were assessed: salt type, uranium enrichment within the salt, pipe material, pipe outer diameter, pipe wall thickness, and presence of insulation material between the detector and the salt-filled pipe. The simulation results are quantitative, but the analysis intentionally encompasses a broad range of scenarios. Therefore, some of the comparisons are qualitative, to make generalizations for future applications. These results assess combinations of realistic design features (i.e., salt chemical composition, uranium enrichment, pipe material, pipe dimensions) and detectors to identify promising (and unfavorable) potential monitoring methods for use in safeguards and security applications, in line with figures of merit discussed in Section 6.1. Future work will further assess specific cases (i.e., optimizing monitoring for a specific salt chemical and isotopic composition).

The criteria used to determine whether measurement techniques were feasible was whether the simulated signal was  $4\sigma$  greater than background. The signal represents different values depending on which measurement technique is explored. For example, when looking at gamma detection of specific energy photopeaks, the signal is the photopeak area. When exploring total neutron counting, the signal is the total neutron (i.e., singles) count rate.

When exploring coincidence counting, the signal is the doubles count rate. Section 6.2 describes how background was estimated for gamma measurements. Both gamma and neutron simulations were run transporting particles generated in the volumetric source term (i.e., the salt) through the salt, piping, insulation, aluminum jacketing, and argon environment. Outside of these interactions, facility walls and concrete flooring were not modeled. This is analogous to assuming that the detector system is well shielded and that there is no environmental background. In reality, a well-shielded detection system is likely for fresh fuel applications, but the environmental background would need to be experimentally determined and optimized. For coincident counting, the background neutron count rate was assumed to be equal to the accidentals count rate, as described in Section 6.3. Overall, the criteria of  $4\sigma$  signal greater than background enables determination of whether a measurement technique is promising (i.e., feasible) and worth investing resources into for future R&D, which is the goal of this research.

All data shown in this section were taken assuming the salt components have been comingled for 1 year (i.e., salt synthesis took place 1 year ago, and all salt components were combined). This allows for the buildup of uranium daughters and optimizes the potential for the 1,001 keV photopeak to be detected. The time since salt combination would impact the detector results described in this chapter in a similar manner as described in Chapter 4. All count rates in this section were determined by multiplying the results of the MCNP tallies,  $T$  by the total number of source particles emitted from the salt. For active measurements, an initial source strength of  $1 \times 10^5$  neutrons per second (nps) was used in Equation (6.1).

$$I_{active} = 1 \times 10^5 \text{ nps} \quad (6.1)$$

This is the approximate source strength of the two AmLi sources in the AWCC. The number of source particles (either photons or neutrons) for passive simulations,  $I_{passive}$ , was calculated using Equation 6.2, where the  $\frac{SourceParticles}{g}$  is the output of ORIGEN (photons or neutrons), as described in Chapter 4, summed for all energies, with units of counts per gram of salt.

$$I_{passive} = \frac{SourceParticles}{g} \rho_{salt} V_{visible} \quad (6.2)$$

To calculate the total salt mass, salt densities,  $\rho_{salt}$ , used in this modeling are described in Section 5.2.1. The volume of salt,  $V_{visible}$ , assuming a portion of the salt in the pipe is visible to the detector, was calculated using Equations 6.3 and 6.4.

$$V_{visible,n} = l_{collar} \pi r_{pipe,inner}^2 \quad (6.3)$$

$$V_{visible,g} = \frac{1}{3} l_{pipe} \pi r_{pipe,inner}^2 \quad (6.4)$$

$l_{pipe}$  is the length of the pipe modeled, 100 cm.  $l_{collar}$  is the length of the neutron coincidence collar. The pipe inner radius,  $r_{pipe,inner}$  is determined based on the defined outer radius and the pipe wall thickness. The quantity  $V_{visible,n}$  was used to calculate the visible volume for neutron detectors, based on the length of the neutron detector. The quantity  $V_{visible,g}$  was used for the visible volume for gamma detectors. This is an estimate and, because it was consistently applied, allows for comparison of results, but may not be accurate in estimating actual count rates in a detector system. In future work, measurement campaigns should be conducted and better visible volumes could be determined for future computational simulations.

## 6.1 Figures of Merit

Each measurement technology or method has more and less desirable associated features (e.g., cost, accuracy, ease of use). Because of the difficulty in objectively assessing and, when necessary, appropriately weighting these features to select the most appropriate technology or method to meet a given technical objective, the author has identified the following figures of merit:

1. Cost of equipment
2. Measurement time to achieve reasonable results
3. Measurement precision
4. Level of intrusiveness
  - Level of intrusiveness to material and process

- Equipment introduction requires changes to the safety basis
  - System physically contacts the salt
  - System introduces a radiation environment where there was not one
  - Level of intrusiveness of data
5. Burden on operator during facility operations
  6. Anticipated maintenance requirements
    - Lifetime of components in the environment
    - Frequency of maintenance (e.g., calibration)
  7. Reliability
    - Frequency of failures in environment
    - Indication of bias, drift, etc., in data
    - Trustworthiness of the data (e.g., considering concealment or spoofing possibilities)

These figures of merit could be applied to individual measurement components or to a combination of measurement components and used to objectively compare different options that will meet a technical objective. In some cases, certain figures of merit will have more value than others and weighting factors could be developed to enable a structured method of comparing different measurement techniques to meet a specific technical objective.

In this research, simulated measurement uncertainties (related to figure of merit number 2) were assessed for gamma- and neutron-based measurement techniques. Future work includes analysis of all figures of merit.

## 6.2 Gamma Detection Results

Chapter 4 discusses and Figure 4.6 shows the photon emissions from the fuel salt. This section describes simulated detector responses to those emissions in the NaI and HPGe detector models described in Chapter 5. The net peak areas for two relevant photopeaks, 185.72 and 1,001 keV, were estimated in the HPGe simulation results using a region of interest (ROI) sum. This method is applicable because the two photopeaks are well-resolved

and there is a high signal-to-background ratio. Equation (6.5) shows the calculation used, where  $A$  is the net peak area of the photopeak,  $P$  is the integral of the ROI around the photopeak and  $B$  is the background [58]. The total  $B$  is calculated using Equation (6.6), using background values from the low energy,  $B_l$ , and high energy,  $B_h$  sides outside the ROI. This calculation is from Table 5-5 in [58] using symmetric placement of background ROIs with the number of channels (i.e., MCNP energy bins) in the low energy background equal to the number of channels in the high energy background, both of which are twice the number of channels in the peak.

$$A = P - B \quad (6.5)$$

$$B = B_l + B_h \quad (6.6)$$

The relative variance of the peak areas is estimated using the following equation:

$$\sigma_A = \frac{\sqrt{P + B_l + B_h}}{A} \quad (6.7)$$

Owing to the many modeling and calculation assumptions, these values may not be representative of actual measurement values for photopeak areas. However, they are valuable in understanding whether the 186 and 1,001 keV photopeaks are distinct and, by comparing the relative net photopeak areas, assessing the potential effects of various parameters. These parameters include MSR design parameters like chemical composition of the salt, uranium enrichment, pipe material, pipe dimensions, and presence of insulation as well as detector parameters like standoff distance between the detector and the pipe.

If the 185.7 keV photopeak is well-resolved and has a reasonable relative variance, it could be useful in determining the  $^{235}\text{U}$  mass in the fuel salt. If the 1,001 keV photopeak is well-resolved and has a reasonable relative variance, it could be useful in determining the  $^{238}\text{U}$  mass in the fuel salt. In some cases, the photopeaks for both 185.7 and 1,001 keV are not detectable. In those cases, calculating enrichment through photopeak ratio methods may not be feasible unless other well-resolved, relevant photopeaks can be identified. The tables in Sections 6.2.1–6.2.6 only include photopeak areas and relative variances if the photopeak

is resolved (i.e., the photopeaks are distinct and at least  $4\sigma$  above background). Otherwise, these values are omitted.

Unless stated otherwise, all data presented are for salt that has had components combined for 1 year, detector standoff distances of 2 cm, and includes the insulation and aluminum jacket, as described in Section 5.2.4, within the model. The results will vary for shorter time periods because the combination of salt components due to the lack of buildup of the  $^{238}\text{U}$  daughters that produce the 1,001 keV gamma.

Figure 6.1 shows a summary of the effects of chemical composition, pipe material, and uranium enrichment (as described in Sections 6.2.1 through 6.2.3) on the 186 keV net photopeak area measured through the simulated HPGe.

### 6.2.1 Impact of Chemical Composition of the Salt

The chemical composition of the salt influences the feasibility of using gamma detection for material accountancy. Gamma detection for material accountancy is likely feasible for both salt types, with certain limitations based on other parameters, as described in Sections 6.2.2–6.2.6. The 185.7 keV count rates will likely be higher for chloride-based salts than fluoride-based salts (at least by a factor of two) because of the significantly higher uranium loading (i.e., mass fraction) in chloride-based salts when all other parameters are equal. Table 6.1 compares different salt types and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count rates. Figure 6.2 shows the 186 keV peak in the modeled NaI detector for 19.75 wt%  $^{235}\text{U}$  uranium in the fluoride salt vs. the chloride salt in Hastelloy N piping of 5.08 cm diameter and 0.625 cm thickness for a 10 min count time.

The mean free path of a material can be used to determine the infinite thickness. If the volume that the detector views (based on the detector geometry, the collimator, and the mean free path of photons within the material) is homogeneous, then more simple methods and tools can be used to determine parameters of the material relevant for MC&A (e.g., the enrichment). Figure 6.3 shows the total mass attenuation coefficients,  $\mu$ , for the two salts

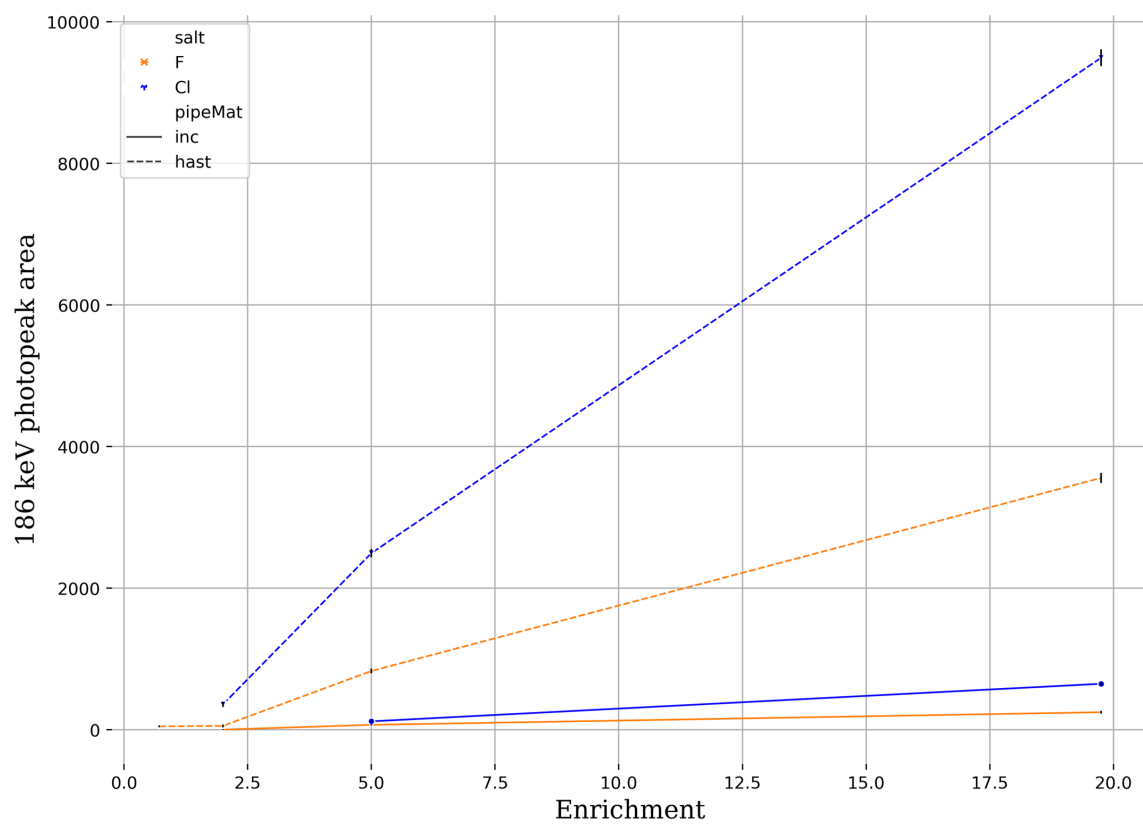


Figure 6.1: Simulated 186 keV net photopeak areas in HPGe for different salt compositions, uranium enrichments, and pipe materials for 10 min count times

Table 6.1: Comparison of HPGe photopeak areas (186 and 1,001 keV) for different chemical compositions of salt with all other factors constant

| Salt Type | $E$<br>(wt% $^{235}\text{U}$ ) | Pipe Material | $d_{\text{pipe,outer}}$<br>(cm) | $t_{\text{pipe}}$<br>(cm) | 186keV |                     | 1,001keV |                     |
|-----------|--------------------------------|---------------|---------------------------------|---------------------------|--------|---------------------|----------|---------------------|
|           |                                |               |                                 |                           | $A^1$  | $\sigma_A^1$<br>(%) | $A^1$    | $\sigma_A^1$<br>(%) |
| F         | 19.75                          | HAST          | 5.08                            | 0.635                     | 3560   | 2.0                 |          |                     |
| Cl        | 19.75                          | HAST          | 5.08                            | 0.635                     | 9490   | 1.3                 |          |                     |
| F         | 5                              | HAST          | 5.08                            | 0.635                     | 831    | 4.2                 |          |                     |
| Cl        | 5                              | HAST          | 5.08                            | 0.635                     | 2496   | 2.4                 |          |                     |
| F         | 2                              | HAST          | 5.08                            | 0.635                     | 56     | 32                  | 17       | 41                  |
| Cl        | 2                              | HAST          | 5.08                            | 0.635                     | 360    | 10.0                |          |                     |
| F         | NU                             | HAST          | 5.08                            | 0.635                     | 49     | 29                  |          |                     |
| Cl        | NU                             | HAST          | 5.08                            | 0.635                     |        |                     | 687      | 3.8                 |

<sup>1</sup>10 min count time

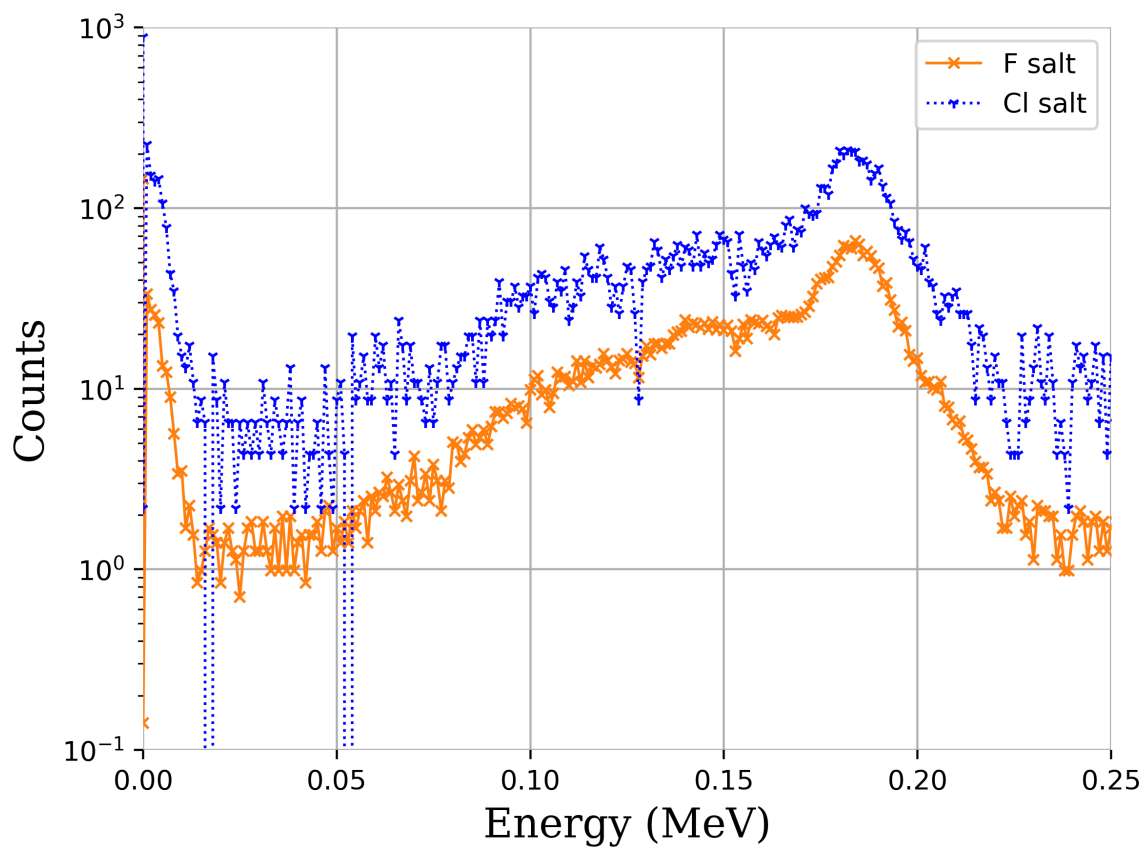


Figure 6.2: Simulated counts in NaI from fluoride and chloride salts with 19.75 wt%  $^{235}\text{U}$ , Hastelloy N piping, and 10 min count times

analyzed in this research, based on the density assumptions described in Chapter 5. Table 6.2 shows the mean free paths and infinite thicknesses calculated for the two salts used in the models. Assuming that any future MSR facilities will use piping similar to what is used in current test and experimental MSR facilities, standard pipe inner diameters will likely be close to or above 2.60 cm (the infinite thickness for the chloride-based salt) and significantly less than 16.51 cm (the infinite thickness for the fluoride-based salt). Therefore, in most practical facility applications, the chloride-based salt could be assumed to be infinitely thick, and the fluoride-based salt cannot be assumed to be infinitely thick. If the sample is infinitely thick, the IAEA can use the GEM code to verify uranium enrichment. In the case that samples are not infinitely thick, the IAEA can use GEM as long as the measured sample geometry is the same as the calibration geometry [67]. In an anticipated scenario where a gamma detector is located on the outside of a pipe taking unattended measurements, it is a reasonable to assume that the measurement and calibration geometries would be identical, so the infinite thicknesses of the material should not impact the feasibility of using gamma detection for material accountancy for either salt type.

### 6.2.2 Impact of Uranium Enrichment

Higher enrichments of uranium within the fuel salt have higher 186 keV photopeak areas and lower relative variances. Table 6.3 compares different enrichments for both the fluoride- and chloride-based salts and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count times. The 186 keV photopeaks are well defined for higher enrichments (i.e., HALEU) and the relative variances are well above what would make them below the minimum detectable activity. However, the simulated count rates from salts with enrichments of 2 wt%  $^{235}\text{U}$  and NU may be too low (or, in the case of fluoride salt, the relative variance too high) to be useful for material accountancy. Figure 6.4 shows the 186 keV peak in the modeled NaI detector for different enrichments in the fluoride and chloride salt, Hastelloy N piping of 5.08 cm diameters and 0.625 cm thickness for a 10 min count time.

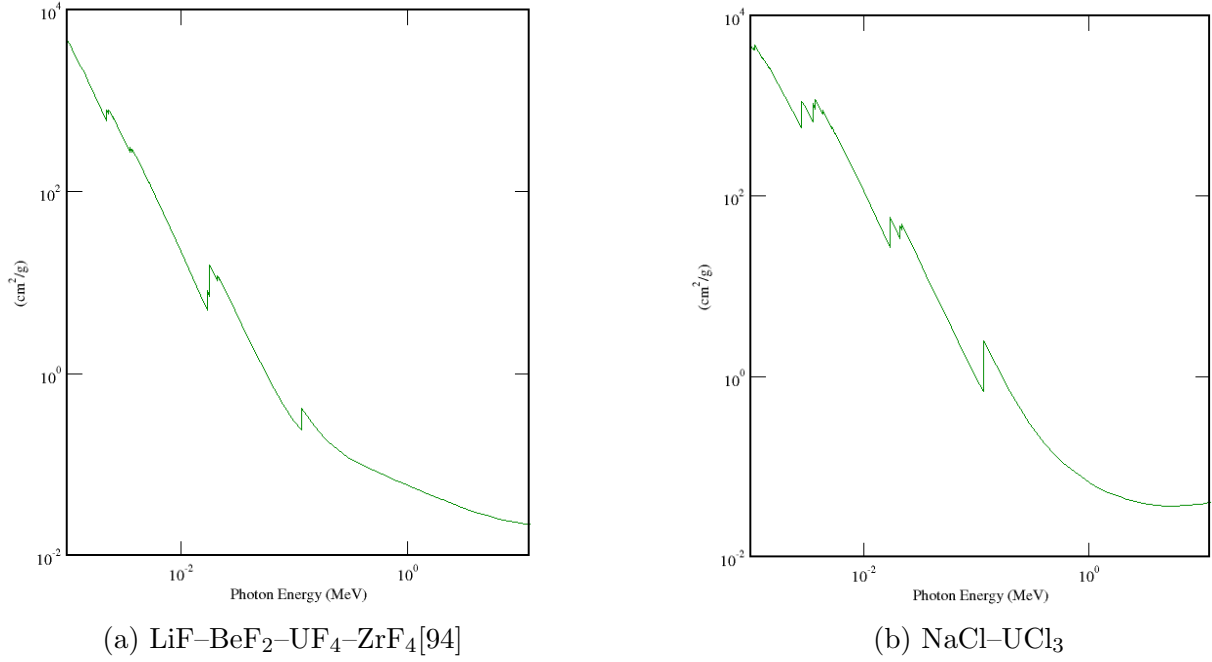


Figure 6.3: Total mass attenuation coefficients for molten salts (without coherent scattering) [94]

Table 6.2: Mean free paths and infinite thicknesses for 186 keV photons in fluoride- and chloride-based unirradiated uranium-bearing fuel salts

| Fuel Salt                                                                       | Density<br>(g/cm <sup>3</sup> ) | Mean<br>Free Path<br>(cm) <sup>1,2</sup> | Infinite<br>Thickness<br>(cm) <sup>3</sup> | Infinite<br>Thickness<br>(in.) <sup>3</sup> |
|---------------------------------------------------------------------------------|---------------------------------|------------------------------------------|--------------------------------------------|---------------------------------------------|
| LiF-BeF <sub>2</sub> -UF <sub>4</sub> -ZrF <sub>4</sub><br>(65-29.1-0.9-5 mol%) | 2.1                             | 2.36                                     | 16.51                                      | 6.50                                        |
| NaCl-UCl <sub>3</sub><br>(66-34 mol%)                                           | 3.3                             | 0.37                                     | 2.60                                       | 1.02                                        |

<sup>1</sup> $1/\mu\rho$  at 186 keV

<sup>2</sup>Calculated using the total mass attenuation coefficient without coherent scattering [94]

<sup>3</sup>Defined as 7 mean free paths, the distance for which the error in assuming infinite sample size is less than 0.1%

Table 6.3: Comparison of HPGe photopeak areas (186 and 1,001 keV) for different uranium enrichments within salt with all other factors constant

| Salt Type | $E$<br>(wt% $^{235}\text{U}$ ) | Pipe Material | $d_{\text{pipe,outer}}$<br>(cm) | $t_{\text{pipe}}$<br>(cm) | 186 keV |                     | 1,001 keV |                     |
|-----------|--------------------------------|---------------|---------------------------------|---------------------------|---------|---------------------|-----------|---------------------|
|           |                                |               |                                 |                           | $A^1$   | $\sigma_A^1$<br>(%) | $A^1$     | $\sigma_A^1$<br>(%) |
| F         | 19.75                          | HAST          | 5.08                            | 0.635                     | 3558    | 2.0                 |           |                     |
| F         | 5                              | HAST          | 5.08                            | 0.635                     | 831     | 4.2                 |           |                     |
| F         | 2                              | HAST          | 5.08                            | 0.635                     | 56      | 32                  | 17        | 41                  |
| F         | NU                             | HAST          | 5.08                            | 0.635                     | 49      | 29                  |           |                     |
| Cl        | 19.75                          | INC           | 5.08                            | 0.635                     | 650     | 3.9                 | 520       | 5.4                 |
| Cl        | 5                              | INC           | 5.08                            | 0.635                     | 120     | 9.1                 |           |                     |
| Cl        | 2                              | INC           | 5.08                            | 0.635                     |         |                     |           |                     |
| Cl        | NU                             | INC           | 5.08                            | 0.635                     |         |                     | 687       | 3.8                 |

<sup>1</sup>10 min count time

### 6.2.3 Impact of Pipe Material

The material of piping around the salt has an impact on the feasibility of using gamma detection for material accountancy. Table 6.4 compares different pipe materials and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count times. For all enrichments greater than NU, gamma detection for material accountancy using the 185.7 keV photopeak should be feasible for salts in both Hastelloy N and Inconel 625 piping. However, count rates are significantly higher (at least by an order of magnitude) in both salt types in Hastelloy N piping, as compared to Inconel 625 piping.

### 6.2.4 Impact of Pipe Outer Diameter

The outer pipe diameter has an impact on the feasibility of using gamma detection for material accountancy. Table 6.5 compares different pipe outer diameters and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count times. In chloride-based salts inside Inconel 625 piping, gamma detection may only be feasible in small (i.e., approximately 5 cm pipes or smaller) and large (i.e., approximately 30 cm pipes) pipe diameters. In salts in Hastelloy N piping, gamma detection may be useful in pipes of all diameters, but the uncertainties decrease as the pipe diameter increases. Lower uncertainties may be achieved in measurements, or, for the same uncertainties, shorter count times could be implemented or a less costly and lower resolution detector may be used in larger diameter pipes.

### 6.2.5 Impact of Pipe Thickness

The pipe thickness has an impact on the feasibility of using gamma detection for material accountancy. Table 6.6 compares different pipe thicknesses and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count times. Thinner pipe walls clearly attenuate photons less; however, for reasonable pipe wall thicknesses (e.g., 1.27 cm and less), gamma detection for

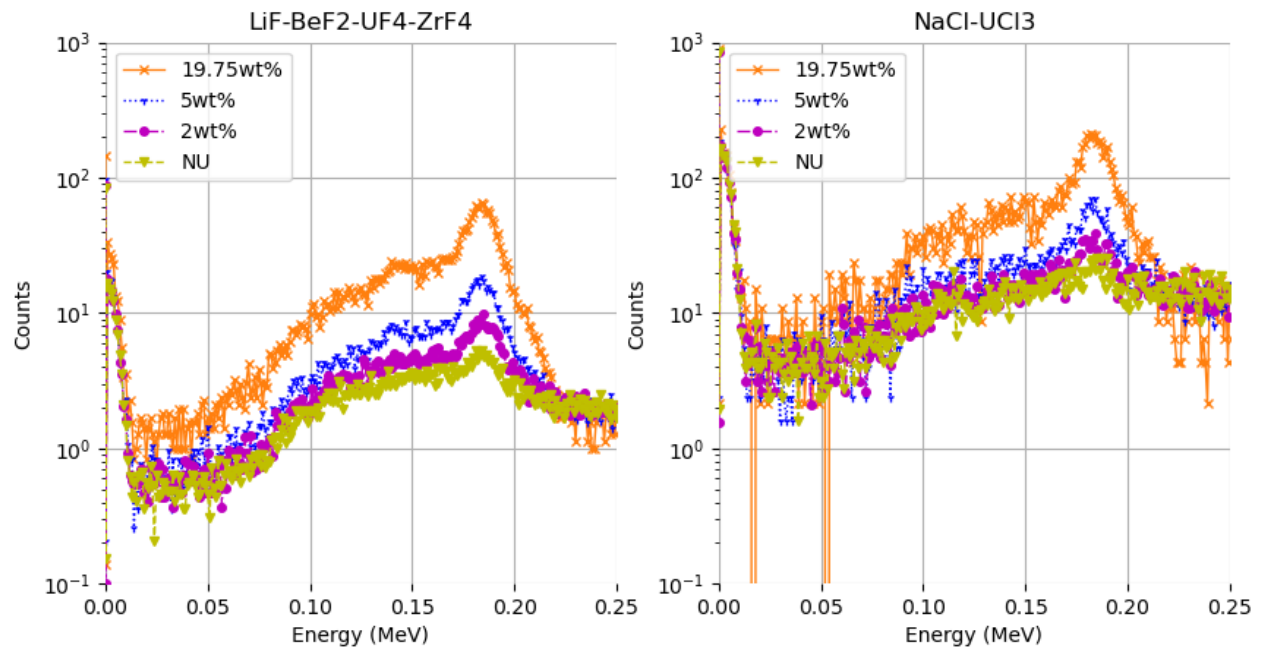


Figure 6.4: Simulated counts in NaI from different uranium enrichments with Hastelloy N piping and 10 min count times

Table 6.4: Comparison of HPGe photopeak areas (186 and 1,001 keV) for different pipe materials with all other factors constant

| Salt Type | $E$<br>(wt% $^{235}\text{U}$ ) | Pipe Material | $d_{\text{pipe,outer}}$<br>(cm) | $t_{\text{pipe}}$<br>(cm) | 186 keV |                     | 1,001 keV |                     |
|-----------|--------------------------------|---------------|---------------------------------|---------------------------|---------|---------------------|-----------|---------------------|
|           |                                |               |                                 |                           | $A^1$   | $\sigma_A^1$<br>(%) | $A^1$     | $\sigma_A^1$<br>(%) |
| F         | 19.75                          | HAST          | 5.08                            | 0.635                     | 3558    | 2.0                 |           |                     |
| F         | 19.75                          | INC           | 5.08                            | 0.635                     | 249     | 7.2                 |           |                     |
| Cl        | 19.75                          | HAST          | 5.08                            | 0.635                     | 9492    | 1.3                 |           |                     |
| Cl        | 19.75                          | INC           | 5.08                            | 0.635                     | 650     | 3.9                 | 520       | 5.4                 |
| F         | 5                              | HAST          | 5.08                            | 0.635                     | 831     | 4.2                 |           |                     |
| F         | 5                              | INC           | 5.08                            | 0.635                     | 70      | 12.3                |           |                     |
| Cl        | 5                              | HAST          | 5.08                            | 0.635                     | 2496    | 2.4                 |           |                     |
| Cl        | 5                              | INC           | 5.08                            | 0.635                     | 120     | 9.1                 |           |                     |
| F         | NU                             | HAST          | 5.08                            | 0.635                     | 49      | 28.8                |           |                     |
| F         | NU                             | INC           | 5.08                            | 0.635                     |         |                     |           |                     |
| Cl        | NU                             | HAST          | 5.08                            | 0.635                     |         |                     | 687       | 3.8                 |
| Cl        | NU                             | INC           | 5.08                            | 0.635                     |         |                     | 687       | 3.8                 |

<sup>1</sup>10 min count time

Table 6.5: Comparison of HPGe photopeak areas (186 and 1,001 keV) for different pipe outer diameters with all other factors constant

| Salt Type | $E$<br>(wt% $^{235}\text{U}$ ) | Pipe Material | $d_{\text{pipe,outer}}$<br>(cm) | $t_{\text{pipe}}$<br>(cm) | 186 keV |                     | 1,001 keV |                     |
|-----------|--------------------------------|---------------|---------------------------------|---------------------------|---------|---------------------|-----------|---------------------|
|           |                                |               |                                 |                           | $A^1$   | $\sigma_A^1$<br>(%) | $A^1$     | $\sigma_A^1$<br>(%) |
| F         | 19.75                          | HAST          | 2.54                            | 0.635                     | 685     | 4.6                 |           |                     |
| F         | 19.75                          | HAST          | 5.08                            | 0.635                     | 3,558   | 2.0                 |           |                     |
| F         | 19.75                          | HAST          | 10.16                           | 0.635                     | 6,656   | 1.5                 |           |                     |
| F         | 19.75                          | HAST          | 20.43                           | 0.635                     | 9,488   | 1.2                 |           |                     |
| F         | 19.75                          | HAST          | 30.48                           | 0.635                     | 11,789  | 0.9                 | 3,930     | 1.6                 |
| Cl        | 19.75                          | INC           | 5.08                            | 0.635                     | 650     | 3.9                 | 520       | 5.4                 |
| Cl        | 19.75                          | INC           | 10.16                           | 0.635                     |         |                     | 1,416     | 2.7                 |
| Cl        | 19.75                          | INC           | 20.32                           | 0.635                     |         |                     | 4,876     | 1.4                 |
| Cl        | 19.75                          | INC           | 30.48                           | 0.635                     |         |                     | 5,794     | 1.3                 |
| Cl        | 19.75                          | HAST          | 2.54                            | 0.635                     | 3,937   | 1.9                 |           |                     |
| Cl        | 19.75                          | HAST          | 5.08                            | 0.635                     | 9,492   | 1.3                 |           |                     |
| Cl        | 19.75                          | HAST          | 10.16                           | 0.635                     | 31,149  | 0.62                |           |                     |
| Cl        | 19.75                          | HAST          | 20.43                           | 0.635                     | 27,951  | 0.66                |           |                     |
| Cl        | 19.75                          | HAST          | 30.48                           | 0.635                     | 17,680  | 0.75                | 17,680    | 0.75                |

<sup>1</sup>10 min count time

material accountancy should be feasible. For all but very large outer diameter Inconel 625 pipes with chloride salt, the photopeak area of the 186 keV may be useful with reasonable uncertainties for use in material accountancy. Smaller diameter pipes have lower uncertainties.

## 6.2.6 Impact of Insulation

The presence of pipe insulation does not influence the feasibility of using gamma detection to quantify  $^{235}\text{U}$  using the 186 keV photopeak. This is notable because it means that traditional gamma detectors and supporting electronics could likely be used to monitor fresh fuel salt additions from the outside of piping insulation at temperatures near room temperature, as opposed to temperatures above or near the melting temperatures of the salts. However, the presence of the aluminum jacket may negatively affect the ability to quantify the 1,001 keV photopeak if the salt is in hastelloy piping. With the insulation jacket off in both fluoride and chloride 19.75 wt%  $^{235}\text{U}$  salts, the 1,001 keV photopeak area can be quantified, and therefore the enrichment can be determined. Table 6.7 compares results from models with and without insulation and aluminum jacketing surrounding the pipe and includes photopeak areas,  $A$ , and their relative variances,  $\sigma_A$ , for the 186 and 1,001 keV photopeaks in the modeled HPGe detector for 10 min count times. When the insulation and aluminum jacket were not modeled, the distance between the pipe and the detectors did not change. The gap was filled with argon inert gas. Figure 6.5 shows the 186 keV peak in the NaI detector modeled with and without insulation and the aluminum jacket around the fluoride salt with 19.75 wt%  $^{235}\text{U}$  and Hastelloy N piping of 5.08 cm diameters and 0.625 cm thickness for a 10 min count time. Practically speaking, it may be possible to remove the aluminum jacket for measurements or, if an in situ monitor is stationary around the piping, to have extra insulation of other material that does not attenuate photons between the pipe and the detector to reduce the temperatures enough to optimize detector and supporting electronic performance.

Table 6.6: Comparison of HPGe photopeak areas (186 and 1,001 keV) for different pipe thicknesses with all other factors constant

| Salt Type | $E$<br>(wt% $^{235}\text{U}$ ) | Pipe Material | $d_{\text{pipe,outer}}$<br>(cm) | $t_{\text{pipe}}$<br>(cm) | $A^1$ 186 keV | $\sigma_A^1$<br>(%) | $A^1$ 1,001 keV | $\sigma_A^1$<br>(%) |
|-----------|--------------------------------|---------------|---------------------------------|---------------------------|---------------|---------------------|-----------------|---------------------|
| F         | 19.75                          | HAST          | 2.54                            | 0.635                     | 685           | 4.6                 |                 |                     |
| F         | 19.75                          | HAST          | 2.54                            | 0.254                     | 3,001         | 2.1                 |                 |                     |
| F         | 19.75                          | HAST          | 5.08                            | 1.27                      | 734           | 4.3                 |                 |                     |
| F         | 19.75                          | HAST          | 5.08                            | 0.635                     | 3,558         | 2.0                 |                 |                     |
| F         | 19.75                          | HAST          | 5.08                            | 0.254                     | 9,464         | 1.2                 |                 |                     |
| F         | 19.75                          | HAST          | 30.48                           | 2.54                      | 749           | 3.7                 |                 |                     |
| F         | 19.75                          | HAST          | 30.48                           | 0.635                     | 11,789        | 0.92                | 3,930           | 1.6                 |
| Cl        | 19.75                          | INC           | 5.08                            | 1.27                      | 58            | 13                  | 87              | 21                  |
| Cl        | 19.75                          | INC           | 5.08                            | 0.635                     | 650           | 3.9                 | 520             | 5.4                 |
| Cl        | 19.75                          | INC           | 5.08                            | 0.254                     | 2,528         | 2.9                 | 281             | 11.4                |
| Cl        | 19.75                          | HAST          | 5.08                            | 1.27                      | 2,485         | 2.5                 |                 |                     |
| Cl        | 19.75                          | HAST          | 5.08                            | 0.635                     | 9,492         | 1.3                 |                 |                     |
| Cl        | 19.75                          | HAST          | 5.08                            | 0.254                     | 30,708        | 0.64                |                 |                     |
| Cl        | 19.75                          | INC           | 30.48                           | 2.54                      |               |                     |                 |                     |
| Cl        | 19.75                          | INC           | 30.48                           | 0.635                     |               |                     | 5,794           | 1.3                 |
| Cl        | 19.75                          | HAST          | 30.48                           | 2.54                      |               |                     |                 |                     |
| Cl        | 19.75                          | HAST          | 30.48                           | 0.635                     | 17,680        | 0.8                 | 17,680          | 0.8                 |

<sup>1</sup>10 min count time

Table 6.7: Comparison of HPGe photopeak areas (186 and 1,001 keV) with and without insulation with all other factors constant, pipe outer diameter = 5.08 cm and thickness = 0.635 cm

| Salt Type | Pipe Material | Insulation On | Al Jacket On | 186 keV |                     | 1,001 keV |                     |
|-----------|---------------|---------------|--------------|---------|---------------------|-----------|---------------------|
|           |               |               |              | $A^1$   | $\sigma_A^1$<br>(%) | $A^1$     | $\sigma_A^1$<br>(%) |
| F         | HAST          | True          | True         | 3,558   | 2.0                 |           |                     |
| F         | HAST          | True          | False        | 1,623   | 3.4                 | 30.0      | 23                  |
| F         | HAST          | False         | False        | 1,623   | 3.4                 | 30.0      | 23                  |
| Cl        | HAST          | True          | True         | 9,492   | 1.3                 |           |                     |
| Cl        | HAST          | True          | False        | 5,396   | 1.8                 | 455       | 5.9                 |
| Cl        | HAST          | False         | False        | 5,396   | 1.8                 | 455       | 5.9                 |
| Cl        | INC           | True          | True         | 650     | 3.9                 | 520       | 5.4                 |
| Cl        | INC           | True          | False        | 650     | 3.9                 | 520       | 5.4                 |
| Cl        | INC           | False         | False        | 650     | 3.9                 | 520       | 5.4                 |

<sup>1</sup>10 min count time

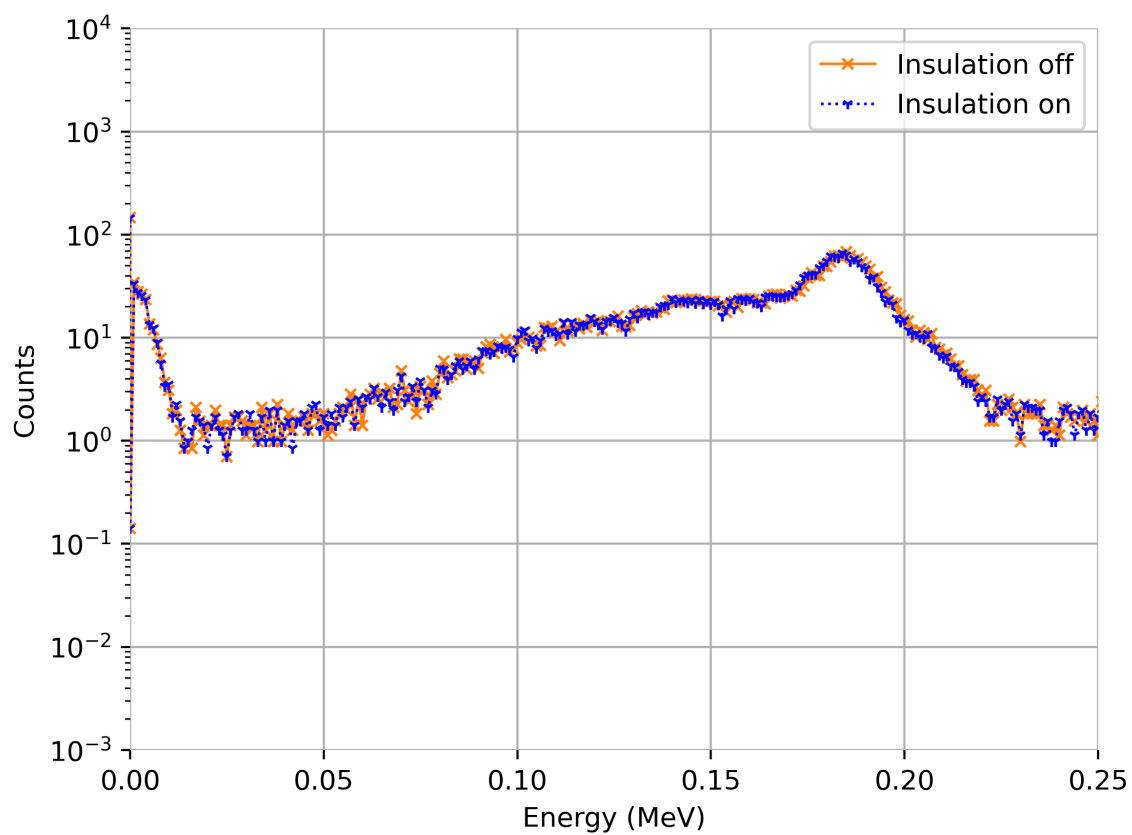


Figure 6.5: Simulated counts in NaI from fluoride salt with uranium of 19.75 wt%  $^{235}\text{U}$  with and without insulation around Hastelloy N piping with 10 min count times

## 6.3 Neutron Detection

Figures 4.3a and 4.3b show the simulated passive neutron emissions from the fuel salts. This section includes the simulated detector responses from measuring neutrons (primarily  $(\alpha, n)$  in the  $^3\text{He}$  collar detector surrounding the pipe and insulation. It also includes the simulated detector response for a half collar with  $^3\text{He}$  tube detectors from neutrons from induced fissions through active interrogation of the salts within the pipe.

To calculate the total neutron counts within the detectors, the tally results from F8 capture tallies in MCNP 6.2 were used, considering all  $^3\text{He}$  tubes as the detection volume. The first moment of an ungated tally is proportional to the singles count rate. The first moment of the gated tally is proportional to the doubles count rate, or the coincidence counting rate. The methodology is based on that described in Example 3.2 in [93]. The singles count rate,  $S$ , is calculated as the number of neutrons counted in the detector (i.e., an output of the ungated F8 tally),  $C_{\text{singles}}$ , divided by the number of source particles started in MCNP (e.g., 1E6 for passive simulations and  $1 \times 10^8$  for active simulations),  $nps$ , multiplied by the initial neutron source strength  $I$ , as defined in Equations (6.2) and (6.1). See Equation (6.8).

$$S = \frac{C_{\text{singles}}}{nps} I \quad (6.8)$$

The doubles count rate,  $D$ , is calculated as the number of doubles counted in the detector (i.e., the first moment of the gated F8 tally),  $C_{\text{doubles}}$ , divided by the number of source particles started in MCNP,  $nps$ , multiplied by the initial neutron source strength,  $I$ . See Equation (6.9).

$$D = \frac{C_{\text{doubles}}}{nps} I \quad (6.9)$$

For coincidence counting to be feasible, the doubles rate must be high enough so that coincident neutrons can be distinguished from background neutrons and count times are reasonable in length for use in an operational facility. Because liquid-fueled MSR developers do not necessarily have all practical aspects of how they will feed initial or makeup salt into a reactor system, it is difficult to state the limit for a reasonable count time. Long

count times may not be unreasonable if the facility can plan for these time delays early in the design process. However, long count times plus undesirable statistics giving moderate-to-high uncertainties may not be desirable for either facility operators or the IAEA. With detector design and detector and source geometry optimization, these values could potentially be higher.

Singles relative uncertainty,  $\sigma_S$  can be estimated as described in (6.10), where  $S$  is defined in Equation (6.8). The count time,  $t$ , is assumed to be 60 min, which is a long count time for material moving in operations but perhaps not unreasonable if planned for early in the design phases of these facilities. Additionally, this is a conservative time limit that allows determination of whether measurement techniques are feasible and worth optimizing and benchmarking with experimental measurements.

$$\sigma_S = \frac{\sqrt{(St)}}{St} \quad (6.10)$$

Doubles relative uncertainty,  $\sigma_D$ , can be estimated as described in (6.11), where  $D$  is defined in Equation (6.9) and the accidentals rate,  $A$ , is defined in (6.12) [58].  $G$  is the gate width (i.e., 64  $\mu s$ ). In MCNP,  $A$  could also be equivalent as  $S$  in the equivalent (i.e., same salt composition, detector, and pipe geometry) passive neutron gated tally, as run in a separate simulation, but (6.12) was used in this feasibility study.

$$\sigma_D = \frac{\sqrt{(2A + D)}}{D\sqrt{t}} \quad (6.11)$$

$$A = S^2 G \quad (6.12)$$

Tables 6.8 and 6.9 include the singles and doubles count rates and relative uncertainties for simulated passive neutron measurements for the fluoride- and chloride-based salts, respectively. Tables 6.10 and 6.11 include the singles and doubles count rates and relative uncertainties for simulated active neutron measurements for the fluoride- and chloride-based salts, respectively. Relative uncertainties are only included if the count rate is at least  $4\sigma$  greater than the background. Sections 6.3.1–6.3.2 provide additional analysis of whether different neutron measurements may be feasible.

Table 6.8: Singles and doubles rates (and relative uncertainties) for simulated passive neutron counts in LiF–BeF<sub>2</sub>–UF<sub>4</sub>–Zr<sub>4</sub>

| $E$<br>(wt%) | Pipe<br>Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | Insulation<br>Modeled | Aluminum<br>Jacket<br>Modeled | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> |
|--------------|------------------|--------------------------|--------------------|-----------------------|-------------------------------|--------------|--------------------------------|----------------------|----------------------------------|
| 19.75        | HAST             | 30.48                    | 0.635              | True                  | True                          | 470          | $7.7 \times 10^{-2}$           | 4.0                  | 17                               |
| 19.75        | HAST             | 20.32                    | 0.635              | True                  | True                          | 181          | 0.12                           | 1.2                  | 23                               |
| 19.75        | HAST             | 10.16                    | 0.635              | True                  | True                          | 34           | 0.29                           | $9.6 \times 10^{-2}$ | <i>53</i>                        |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | True                          | 5.7          | 0.70                           | $8.8 \times 10^{-3}$ | <i>98</i>                        |
| 19.75        | HAST             | 5.08                     | 0.254              | True                  | True                          | 8.2          | 0.58                           | $1.1 \times 10^{-2}$ | <i>108</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | False                         | 6.0          | 0.68                           | $3.1 \times 10^{-3}$ | <i>291</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | False                 | False                         | 6.0          | 0.68                           | $3.1 \times 10^{-3}$ | <i>291</i>                       |
| 19.75        | INC              | 5.08                     | 0.635              | True                  | True                          | 5.5          | 0.71                           | $7.5 \times 10^{-3}$ | <i>112</i>                       |
| 5            | HAST             | 30.48                    | 0.254              | True                  | True                          | 133          | 0.14                           | 0.73                 | <i>27</i>                        |
| 5            | HAST             | 5.08                     | 0.635              | True                  | True                          | 1.5          | 1.4                            | $1.6 \times 10^{-3}$ | <i>146</i>                       |
| 5            | HAST             | 5.08                     | 0.254              | True                  | True                          | 2.2          | 1.1                            | $2.7 \times 10^{-3}$ | <i>125</i>                       |
| 2            | HAST             | 5.08                     | 0.635              | True                  | True                          | 0.67         | 2.0                            | $6.9 \times 10^{-4}$ | <i>159</i>                       |
| NU           | HAST             | 5.08                     | 0.635              | True                  | True                          | 0.28         | 3.2                            | $2.4 \times 10^{-4}$ | <i>202</i>                       |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.9: Singles and doubles rates (and relative uncertainties) for simulated passive neutron counts in NaCl-UCl<sub>3</sub>

| $E$<br>(wt%) | Pipe<br>Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | Insulation<br>Modeled | Aluminum<br>Jacket<br>Modeled | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> |
|--------------|------------------|--------------------------|--------------------|-----------------------|-------------------------------|--------------|--------------------------------|----------------------|----------------------------------|
| 19.75        | INC              | 30.48                    | 0.635              | True                  | True                          | 127          | 0.15                           | 1.2                  | 15                               |
| 19.75        | INC              | 30.48                    | 0.254              | True                  | True                          | 137          | 0.14                           | 1.6                  | 13                               |
| 19.75        | INC              | 20.32                    | 0.635              | True                  | True                          | 54.7         | 0.23                           | 0.37                 | 22                               |
| 19.75        | INC              | 10.16                    | 0.635              | True                  | True                          | 12.3         | 0.48                           | $4.7 \times 10^{-2}$ | <i>40</i>                        |
| 19.75        | INC              | 5.08                     | 0.635              | True                  | True                          | 2.35         | 1.1                            | $4.0 \times 10^{-3}$ | <i>91</i>                        |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | True                          | 2.41         | 1.1                            | $4.2 \times 10^{-3}$ | <i>90</i>                        |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | False                         | 2.46         | 1.1                            | $5.3 \times 10^{-3}$ | <i>73</i>                        |
| 19.75        | HAST             | 5.08                     | 0.635              | False                 | False                         | 2.46         | 1.1                            | $5.3 \times 10^{-3}$ | <i>73</i>                        |
| 5            | INC              | 5.08                     | 0.635              | True                  | True                          | 0.94         | 1.7                            | $8.0 \times 10^{-4}$ | <i>184</i>                       |
| 2            | INC              | 5.08                     | 0.635              | True                  | True                          | 0.66         | 2.1                            | $4.3 \times 10^{-4}$ | <i>242</i>                       |
| NU           | INC              | 5.08                     | 0.635              | True                  | True                          | 0.53         | 2.3                            | $3.4 \times 10^{-4}$ | <i>250</i>                       |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.10: Singles and doubles rates (and relative uncertainties) for simulated active neutron counts in LiF–BeF<sub>2</sub>–UF<sub>4</sub>–Zr<sub>4</sub>

| $E$<br>(wt%) | Pipe<br>Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | Insulation<br>Modeled | Aluminum<br>Jacket<br>Modeled | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> |
|--------------|------------------|--------------------------|--------------------|-----------------------|-------------------------------|--------------|--------------------------------|--------------|----------------------------------|
| 19.75        | HAST             | 30.48                    | 0.635              | True                  | True                          | 14,102       | $1.4 \times 10^{-2}$           | 33.3         | <i>63</i>                        |
| 19.75        | HAST             | 30.48                    | 0.254              | True                  | True                          | 14,413       | $1.4 \times 10^{-2}$           | 48.8         | <i>44</i>                        |
| 19.75        | HAST             | 20.32                    | 0.635              | True                  | True                          | 13,474       | $1.4 \times 10^{-2}$           | 20.0         | <i>100</i>                       |
| 19.75        | HAST             | 10.16                    | 0.635              | True                  | True                          | 11,937       | $1.5 \times 10^{-2}$           | 7.36         | <i>242</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       |
| 19.75        | INC              | 5.08                     | 0.635              | True                  | True                          | 10,462       | $1.6 \times 10^{-2}$           | 2.0          | <i>797</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | False                         | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | False                 | False                         | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       |
| 5            | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,944       | $1.6 \times 10^{-2}$           | 0.69         | <i>2,350</i>                     |
| 2            | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,941       | $1.6 \times 10^{-2}$           | 0.29         | <i>5,550</i>                     |
| NU           | HAST             | 20.32                    | 0.635              | True                  | True                          | 13,309       | $1.4 \times 10^{-2}$           | 1.18         | <i>1,680</i>                     |
| NU           | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,940       | $1.6 \times 10^{-2}$           | 0.12         | <i>13,400</i>                    |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.11: Singles and doubles rates (and relative uncertainties) for simulated active neutron counts in NaCl–UCl<sub>3</sub>

| $E$<br>(wt%) | Pipe<br>Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | Insulation<br>Modeled | Aluminum<br>Jacket<br>Modeled | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> |
|--------------|------------------|--------------------------|--------------------|-----------------------|-------------------------------|--------------|--------------------------------|--------------|----------------------------------|
| 19.75        | INC              | 30.48                    | 0.635              | True                  | True                          | 11,132       | $1.6 \times 10^{-2}$           | 40.0         | <i>41</i>                        |
| 19.75        | INC              | 20.32                    | 0.635              | True                  | True                          | 10,921       | $1.6 \times 10^{-2}$           | 29.3         | <i>56</i>                        |
| 19.75        | INC              | 10.16                    | 0.635              | True                  | True                          | 10,271       | $1.6 \times 10^{-2}$           | 17.4         | <i>88</i>                        |
| 19.75        | INC              | 5.08                     | 0.635              | True                  | True                          | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                       |
| 19.75        | INC              | 5.08                     | 0.254              | True                  | True                          | 10,058       | $1.7 \times 10^{-2}$           | 17.1         | <i>88</i>                        |
| 19.75        | INC              | 2.54                     | 0.254              | True                  | True                          | 10,421       | $1.6 \times 10^{-2}$           | 9.2          | <i>170</i>                       |
| 19.75        | INC              | 5.08                     | 0.635              | True                  | False                         | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                       |
| 19.75        | INC              | 5.08                     | 0.635              | False                 | False                         | 10,043       | $1.6 \times 10^{-2}$           | 10.1         | <i>148</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,541       | $1.6 \times 10^{-2}$           | 10.8         | <i>146</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | True                  | False                         | 10,541       | $1.6 \times 10^{-2}$           | 10.8         | <i>146</i>                       |
| 19.75        | HAST             | 5.08                     | 0.635              | False                 | False                         | 10,541       | $1.6 \times 10^{-2}$           | 10.8         | <i>146</i>                       |
| 5            | INC              | 5.08                     | 0.635              | True                  | True                          | 9,980        | $1.7 \times 10^{-2}$           | 3.7          | <i>401</i>                       |
| 2            | INC              | 5.08                     | 0.635              | True                  | True                          | 9,958        | $1.7 \times 10^{-2}$           | 1.7          | <i>890</i>                       |
| NU           | INC              | 10.16                    | 0.635              | True                  | True                          | 10,063       | $1.7 \times 10^{-2}$           | 1.2          | <i>1,218</i>                     |
| NU           | INC              | 5.08                     | 0.635              | True                  | True                          | 9,946        | $1.7 \times 10^{-2}$           | 0.63         | <i>2,350</i>                     |
| NU           | HAST             | 5.08                     | 0.635              | True                  | True                          | 10,442       | $1.6 \times 10^{-2}$           | 0.69         | <i>13,400</i>                    |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

### 6.3.1 Passive Neutron Detection with $^3\text{He}$ Results

This section describes the modeling results from analyzing simulated passive neutron emissions from salt within pipes and detection in a full collar detector with variable number of  $^3\text{He}$  tubes (based on the pipe diameter).

The changes in  $S$  and  $D$  values for simulations where one parameter was varied, and all others remained constant are expressed in ratios in Tables 6.12–6.17 in the following subsections. All values described in this section, with the exception of those analyzed specifically without insulation, are from models with insulation and an aluminum jacket (described in Section 5.2.4).

#### Impact of Chemical Composition of the Salt

The chemical composition of the salt has a significant impact on the feasibility of passive neutron detection usefulness for material accountancy. Table 6.12 includes  $S$  and  $D$ , their relative uncertainties, and the ratio showing change between the simulated count rates. For total neutron counting methods in passive measurements, the total singles count rates are likely too small for total neutron counting to be a reasonable measurement choice for standard pipe diameters, as included in Table 6.12. The salt type does impact the singles count rate. Single neutrons are detected at about half the rate in the modeled  $^3\text{He}$  detector from chloride-based salts as they are for fluoride-based salts, due to the significantly greater  $(\alpha, n)$  emission rate in fluoride-based salts, despite the order of magnitude greater uranium mass fraction in the chloride-based salt as compared to the fluoride-based salt. Coincidence counting with passive measurements does not give a doubles counts that are  $4\sigma$  above accidental counts for a 60 min count time, so this measurement technique is not feasible for pipe diameters included in Table 6.12.

#### Impact of Uranium Enrichment

The uranium enrichment within the salt has a significant impact on the feasibility of passive neutron detection usefulness for material accountancy. Table 6.13 includes  $S$  and  $D$ , their

Table 6.12: Comparison of simulated passive  $S$  and  $D$  for different chemical compositions of salt with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|----------------------|----------------------------------|---------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 5.7          | 0.70                           | $8.8 \times 10^{-3}$ | <i>98</i>                        | 1       |
| Cl        | 19.75        | HAST          | 5.08                     | 0.635              | 2.4          | 1.1                            | $4.2 \times 10^{-3}$ | <i>90</i>                        | 0.42    |
| F         | 19.75        | INC           | 5.08                     | 0.635              | 5.5          | 0.71                           | $7.5 \times 10^{-3}$ | <i>112</i>                       | 1       |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 2.4          | 1.1                            | $4.0 \times 10^{-3}$ | <i>91</i>                        | 0.44    |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.13: Comparison of simulated passive  $S$  and  $D$  for different uranium enrichments with all other factors constant

| Salt Type | $E$ (wt%) | Pipe Material | $D_{pipe,outer}$ (cm) | $t_{pipe}$ (cm) | $S$ (cps) | $\sigma_S$ (%) <sup>1</sup> | $D$ (cps)            | $\sigma_D$ (%) <sup>1,2</sup> | rel $S$ <sup>3</sup> |
|-----------|-----------|---------------|-----------------------|-----------------|-----------|-----------------------------|----------------------|-------------------------------|----------------------|
| F         | 19.75     | HAST          | 5.08                  | 0.635           | 5.7       | 0.70                        | $8.8 \times 10^{-3}$ | <i>98</i>                     | 1.0                  |
| F         | 5         | HAST          | 5.08                  | 0.635           | 1.5       | 1.4                         | $1.6 \times 10^{-3}$ | <i>146</i>                    | 0.27                 |
| F         | 2         | HAST          | 5.08                  | 0.635           | 0.67      | 2.0                         | $6.9 \times 10^{-4}$ | <i>159</i>                    | 0.12                 |
| F         | NU        | HAST          | 5.08                  | 0.635           | 0.28      | 3.2                         | $2.4 \times 10^{-4}$ | <i>202</i>                    | 0.05                 |
| Cl        | 19.75     | INC           | 5.08                  | 0.635           | 2.4       | 1.1                         | $4.0 \times 10^{-3}$ | <i>91</i>                     | 1.0                  |
| Cl        | 5         | INC           | 5.08                  | 0.635           | 0.94      | 1.7                         | $8.0 \times 10^{-4}$ | <i>184</i>                    | 0.40                 |
| Cl        | 2         | INC           | 5.08                  | 0.635           | 0.66      | 2.1                         | $4.3 \times 10^{-4}$ | <i>242</i>                    | 0.28                 |
| Cl        | NU        | INC           | 5.08                  | 0.635           | 0.53      | 2.3                         | $3.4 \times 10^{-4}$ | <i>250</i>                    | 0.22                 |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio compared to 19.75 wt% <sup>235</sup>U

relative uncertainties, and the ratio showing change between the simulated count rates, as compared to 19.75 wt%  $^{235}\text{U}$  enriched uranium in the same salt type. For both fluoride- and chloride-based salts, with all other factors remaining constant, changing the uranium enrichment within the salt composition changes  $S$  by greater than 60%, and  $D$  by greater than 80% for the four enrichment values explored in this research. Coincidence counting of passive neutrons in the standard diameter pipes evaluated in Table 6.13 is not feasible because the doubles counts are not  $4\sigma$  above accidental counts for a 60 min count time.

### **Impact of Pipe Material**

The pipe material does not have a significant impact on the passive neutron detector total neutron count in the full collar detector. Table 6.14 includes  $S$ ,  $D$ , their relative uncertainties, and the ratio showing change between the simulated count rates. All of these total neutron count rates are likely too low to make total passive neutron counting a feasible accountancy method for standard diameter pipes. Coincidence counting is also not feasible for these standard diameter pipes because the doubles counts are not  $4\sigma$  above accidental counts for a 60 min count time.

### **Impact of Pipe Outer Diameter**

The outer diameter of the pipe, which directly impacts the total salt volume and therefore uranium mass being measured, significantly impacts the feasibility of passive neutron detection for material accountancy. Table 6.15 includes  $S$ ,  $D$ , their relative uncertainties, and the ratio showing change between the simulated count rates, as compared to a 5.08 cm diameter pipe. Total passive neutron counting to determine  $^{234}\text{U}$  mass from  $(\alpha, n)$  neutron emissions may be a feasible measurement method for safeguards or security purposes in large diameter pipes. Coincidence counting of passive neutrons to determine  $^{235}\text{U}$  mass is also potentially feasible in larger diameter pipes. However, the required diameter to make this measurement feasible (approximately 8 in.) is larger than what is used in existing MSR research and test loops (i.e., approximately 1 and 2 in. diameter pipes). Depending on the uranium enrichment, this may be even larger than what would be acceptable for use in a facility because of criticality safety guidelines and requirements.

Table 6.14: Comparison of simulated passive  $S$  and  $D$  for different pipe materials with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|----------------------|----------------------------------|---------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 5.7          | 0.70                           | $8.8 \times 10^{-3}$ | <i>98</i>                        | 1.0     |
| F         | 19.75        | INC           | 5.08                     | 0.635              | 5.5          | 0.71                           | $7.5 \times 10^{-3}$ | <i>112</i>                       | 0.97    |
| Cl        | 19.75        | HAST          | 5.08                     | 0.635              | 2.4          | 1.2                            | $4.2 \times 10^{-3}$ | <i>90</i>                        | 1.0     |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 2.4          | 1.1                            | $4.0 \times 10^{-3}$ | <i>91</i>                        | 1.0     |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* mean the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.15: Comparison of simulated passive  $S$  and  $D$  for different pipe outer diameters with all other factors constant

| Salt Type | $E$ (wt%) | Pipe Material | $D_{pipe,outer}$ (cm) | $t_{pipe}$ (cm) | $S$ (cps) | $\sigma_S$ (%) <sup>1</sup> | $D$ (cps)            | $\sigma_D$ (%) <sup>1,2</sup> | rel $S^3$ | rel $D^3$ |
|-----------|-----------|---------------|-----------------------|-----------------|-----------|-----------------------------|----------------------|-------------------------------|-----------|-----------|
| F         | 19.75     | HAST          | 5.08                  | 0.635           | 5.7       | 0.70                        | $8.8 \times 10^{-3}$ | <i>98</i>                     | 1.0       | 1.0       |
| F         | 19.75     | HAST          | 10.16                 | 0.635           | 34        | 0.29                        | $9.6 \times 10^{-2}$ | <i>53</i>                     | 5.9       | 11        |
| F         | 19.75     | HAST          | 20.32                 | 0.635           | 180       | 1.2                         | 1.2                  | 23                            | 32        | 130       |
| F         | 19.75     | HAST          | 30.48                 | 0.635           | 470       | $7.7 \times 10^{-2}$        | 4.0                  | 17                            | 82        | 450       |
| Cl        | 19.75     | INC           | 5.08                  | 0.635           | 2.4       | 1.1                         | $4.0 \times 10^{-3}$ | <i>91</i>                     | 1.0       | 1.0       |
| Cl        | 19.75     | INC           | 10.16                 | 0.635           | 12        | 4.8                         | $4.7 \times 10^{-2}$ | <i>40</i>                     | 5.2       | 12        |
| Cl        | 19.75     | INC           | 20.32                 | 0.635           | 55        | 2.3                         | 0.37                 | 22                            | 23        | 91        |
| Cl        | 19.75     | INC           | 30.48                 | 0.635           | 127       | 1.5                         | 1.2                  | 15                            | 54        | 310       |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* mean the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio compared to  $d_{outer} = 5.08$  cm

### Impact of Pipe Thickness

The thickness of the pipe being measured likely does not have a significant impact on the feasibility of passive neutron detection use for material accountancy. Table 6.16 includes  $S$ ,  $D$ , their relative uncertainties, and the relative ratio between the simulated count rates for two different pipe thicknesses that are similar to pipe diameters likely to be used in test or operational facilities (e.g., based on Schedule 40 and Schedule 80, as defined by the ASME). In smaller diameter pipes, smaller thickness pipes does reduce some of the neutrons captured in the piping material before they get to the detector and the singles count rate is increased. However, this increased singles rate (or doubles rate) is still not likely high enough to make total passive neutron counting (or passive coincidence counting) feasible. In larger pipe diameters, the change in  $S$  due to the change in pipe wall thickness is likely not enough to significantly impact the feasibility of total passive neutron counting or passive coincidence counting, but it may be a factor that could help optimize counting statistics. Thinner pipe walls, if reasonable for operational considerations, in facilities relying on passive neutron measurements, may be beneficial.

### Impact of Insulation

The presence of insulation or an aluminum jacket around the insulation does not have a significant impact on the feasibility of passive neutron measurement use for safeguards or security. Table 6.17 includes  $S$ ,  $D$ , their relative uncertainties, and the relative ration between the simulated count rates for scenarios with the pipe insulation on and off and the aluminum jacket on and off. The presence of an aluminum jacket makes a small impact on neutron measurements, but the silica insulation does not noticeable attenuate neutrons.

### 6.3.2 Active Neutron Detection with $^3\text{He}$ Results

In general, while coincidence counting in combination with active neutron interrogation is a promising method for material accountancy of  $^{235}\text{U}$  in unirradiated fuel salt, the detector modeled in this research, similar to the AWCC used as a collar around piping, causes relative uncertainties that, in most cases analyzed, greatly exceed those which would be useful for

Table 6.16: Comparison of simulated passive  $S$  and  $D$  for different pipe thicknesses with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ | rel $D$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|----------------------|----------------------------------|---------|---------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 5.7          | 0.70                           | $8.8 \times 10^{-3}$ | <i>98</i>                        | 1.0     | 1.0     |
| F         | 19.75        | HAST          | 5.08                     | 0.254              | 8.2          | 0.58                           | $1.1 \times 10^{-2}$ | <i>108</i>                       | 1.4     | 1.25    |
| F         | 5            | HAST          | 5.08                     | 0.635              | 1.5          | 1.4                            | $1.6 \times 10^{-3}$ | <i>146</i>                       | 1.0     | 1.0     |
| F         | 5            | HAST          | 5.08                     | 0.254              | 2.2          | 1.1                            | $2.7 \times 10^{-3}$ | <i>125</i>                       | 1.45    | 1.7     |
| Cl        | 19.75        | INC           | 30.48                    | 0.635              | 127          | 0.15                           | 1.2                  | 15                               | 1.0     | 1.0     |
| Cl        | 19.75        | INC           | 30.48                    | 0.254              | 137          | 0.14                           | 1.6                  | 13                               | 1.1     | 1.3     |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.17: Comparison of passive  $S$  and  $D$  for pipes with and without insulation; all pipe materials are Hastelloy N and all uranium enrichments are 19.75 wt%  $^{235}\text{U}$

| Salt Type | $E$<br>(wt%) | Pipe Material | Insulation Modeled | Aluminum Jacket Modeled | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps)         | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ <sup>3</sup> |
|-----------|--------------|---------------|--------------------|-------------------------|--------------|--------------------------------|----------------------|----------------------------------|----------------------|
| F         | 5.08         | 0.635         | True               | True                    | 5.7          | 0.70                           | $8.8 \times 10^{-3}$ | <i>98</i>                        | 1.0                  |
| F         | 5.08         | 0.635         | True               | False                   | 6.0          | $6.8 \times 10^{-3}$           | $3.1 \times 10^{-3}$ | <i>291</i>                       | 1.1                  |
| F         | 5.08         | 0.635         | False              | False                   | 6.0          | $6.8 \times 10^{-3}$           | $3.1 \times 10^{-3}$ | <i>291</i>                       | 1.1                  |
| Cl        | 5.08         | 0.635         | True               | True                    | 2.4          | 1.1                            | $4.0 \times 10^{-3}$ | <i>90</i>                        | 1.0                  |
| Cl        | 5.08         | 0.635         | True               | False                   | 2.5          | $1.1 \times 10^{-2}$           | $5.3 \times 10^{-3}$ | <i>73</i>                        | 1.0                  |
| Cl        | 5.08         | 0.635         | False              | False                   | 2.5          | $1.1 \times 10^{-2}$           | $5.3 \times 10^{-3}$ | <i>73</i>                        | 1.0                  |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio compared to insulation and aluminum jacket on

safeguards and security purposes. The high uncertainties are due to the high accidentals rate in the  $^3\text{He}$  tubes from neutrons from the AmLi source. No active neutron coincidence measurements are feasible using the moderated  $^3\text{He}$  tube collar simulated in this research. However, the relative change seen in the doubles count rates is discussed in the following sections to describe the potential impact of the salt chemical composition, uranium enrichment, pipe material, pipe outer diameter, pipe thickness, and presence of insulation in the case that it is helpful for future analysis with different detectors. Chapter 7 describes other detector systems that may be useful.

### **Impact of Chemical Composition of the Salt**

The doubles count rates are higher for chloride-based salts as compared to fluoride-based salts for the salts analyzed for all uranium enrichments, likely due to the order of magnitude higher uranium-loading (i.e., mass fraction of uranium) in chloride fuel salts. Table 6.18 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for different salt chemical compositions with all other parameters held constant.

Although relative uncertainties are high for both chloride- and fluoride-based salts, they are higher in fluoride salts, which is likely due to a higher accidentals rate, potentially due to a lower density salt. The singles rates from passive simulations (see Table 6.12 for the same salts) are several orders of magnitude smaller than the singles rates for active measurements, so the primary source of accidentals is neutrons from the AmLi source and not  $(\alpha, n)$  reactions.

### **Impact of Uranium Enrichment**

The doubles count rates increase as the uranium enrichment increases, which is due to higher mass fractions of  $^{235}\text{U}$  present in the salt to undergo fission caused by the incident AmLi neutrons. Table 6.19 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for different uranium enrichments with all other parameters held constant. Although the doubles count rate may be high enough to make coincidence counting with neutron interrogation feasible for HALEU fluoride salts and LEU and HALEU chloride salts, the relative uncertainties are too high to make this detector setup useful.

Table 6.18: Comparison of simulated active  $S$  and  $D$  for different chemical compositions of salt with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ | rel $D$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|--------------|----------------------------------|---------|---------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 10,955       | $1.6 \times 10^{-2}$           | 2.2          | <i>740</i>                       | 1.0     | 1.0     |
| Cl        | 19.75        | HAST          | 5.08                     | 0.635              | 10,541       | $1.6 \times 10^{-2}$           | 10.8         | <i>150</i>                       | 0.96    | 4.9     |
| F         | 19.75        | INC           | 5.08                     | 0.635              | 10,462       | $1.6 \times 10^{-2}$           | 2.0          | <i>800</i>                       | 1.0     | 1.0     |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>150</i>                       | 0.96    | 5.2     |
| F         | 5            | HAST          | 5.08                     | 0.635              | 10,944       | $1.6 \times 10^{-2}$           | 0.69         | <i>2,350</i>                     | 1.0     | 1.0     |
| Cl        | 5            | HAST          | 5.08                     | 0.635              | 10,478       | $1.6 \times 10^{-2}$           | 4.0          | <i>387</i>                       | 0.96    | 5.8     |
| F         | 2            | HAST          | 5.08                     | 0.635              | 10,941       | $1.6 \times 10^{-2}$           | 0.29         | <i>5,550</i>                     | 1.0     | 1.0     |
| Cl        | 2            | HAST          | 5.08                     | 0.635              | 10,454       | $1.6 \times 10^{-2}$           | 1.7          | <i>929</i>                       | 0.96    | 5.9     |
| F         | NU           | HAST          | 5.08                     | 0.635              | 10,940       | $1.6 \times 10^{-2}$           | 0.12         | <i>13,400</i>                    | 1.0     | 1.0     |
| Cl        | NU           | HAST          | 5.08                     | 0.635              | 10,442       | $1.6 \times 10^{-2}$           | 0.69         | <i>13,400</i>                    | 0.95    | 5.8     |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.19: Comparison of simulated active  $S$  and  $D$  for different uranium enrichments with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S^3$ | rel $D^3$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|--------------|----------------------------------|-----------|-----------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       | 1.0       | 1.0       |
| F         | 5            | HAST          | 5.08                     | 0.635              | 10,944       | $1.6 \times 10^{-2}$           | 0.69         | <i>2,350</i>                     | 1.0       | 0.31      |
| F         | 2            | HAST          | 5.08                     | 0.635              | 10,941       | $1.6 \times 10^{-2}$           | 0.29         | <i>5,550</i>                     | 1.0       | 0.13      |
| F         | NU           | HAST          | 5.08                     | 0.635              | 10,940       | $1.6 \times 10^{-2}$           | 0.12         | <i>13,400</i>                    | 1.0       | 0.05      |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 1,0043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                       | 1.0       | 1.0       |
| Cl        | 5            | INC           | 5.08                     | 0.635              | 9,980        | $1.7 \times 10^{-2}$           | 3.7          | <i>401</i>                       | 0.99      | 0.37      |
| Cl        | 2            | INC           | 5.08                     | 0.635              | 9,958        | $1.7 \times 10^{-2}$           | 1.7          | <i>890</i>                       | 0.99      | 0.16      |
| Cl        | NU           | INC           | 5.08                     | 0.635              | 9,946        | $1.7 \times 10^{-2}$           | 0.63         | <i>2,350</i>                     | 0.99      | 0.06      |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio as compared to 19.75%wt% <sup>235</sup>U

### **Impact of Pipe Material**

Pipe material may not seem to consistently change the doubles count rates for coincidence counting with neutron interrogation. Table 6.20 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for different pipe materials with all other parameters held constant.

### **Impact of Pipe Outer Diameter**

Larger pipe diameters have increased doubles rates and reduced relative uncertainties. The larger doubles rates are due to the increased mass of  $^{235}\text{U}$  in larger volumes within larger diameter pipes. The larger relative doubles rate reduces the relative uncertainty. Table 6.21 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for different pipe outer diameters with all other parameters held constant. Large diameter pipes with salt with HALEU are among the most promising of the measurement scenarios for active neutron interrogation analyzed in this research, but the relative uncertainties are still too high with the modeled detector to be useful for quantifying uranium for safeguards or security purposes.

### **Impact of Pipe Thickness**

Doubles count rates from salt within pipes with thinner walls are higher and have less relative uncertainty than pipes with thicker walls. Table 6.22 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for different pipe thicknesses with all other parameters held constant.

### **Impact of Insulation**

The presence of insulation or aluminum jacketing does not impact the doubles count rates or their relative uncertainties. Table 6.23 includes  $S$  and  $D$ , their relative uncertainties, and the ratios between the simulated count rates for pipes modeled with and without insulation and aluminum jacketing with all other parameters held constant. Practically speaking, if

the detector uses HDPE, insulation will be necessary to reduce the temperatures to below melting temperatures.

Table 6.20: Comparison of simulated active  $S$  and  $D$  for different pipe materials with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S$ | rel $D$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|--------------|----------------------------------|---------|---------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       | 1.0     | 1.0     |
| F         | 19.75        | INC           | 5.08                     | 0.635              | 10,462       | $1.6 \times 10^{-2}$           | 2.0          | <i>797</i>                       | 0.95    | 0.88    |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                       | 1.0     | 1.0     |
| Cl        | 19.75        | HAST          | 5.08                     | 0.635              | 10,541       | $1.6 \times 10^{-4}$           | 10.8         | <i>146</i>                       | 1.0     | 1.1     |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.21: Comparison of simulated active  $S$  and  $D$  for different pipe outer diameters with all other factors constant

| Salt Type | $E$<br>(wt%) | Pipe Material | $D_{pipe,outer}$<br>(cm) | $t_{pipe}$<br>(cm) | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>1</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>1,2</sup> | rel $S^3$ | rel $D^3$ |
|-----------|--------------|---------------|--------------------------|--------------------|--------------|--------------------------------|--------------|----------------------------------|-----------|-----------|
| F         | 19.75        | HAST          | 5.08                     | 0.635              | 10,955       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                       | 1.0       | 1.0       |
| F         | 19.75        | HAST          | 10.16                    | 0.635              | 11,937       | $1.5 \times 10^{-2}$           | 7.4          | <i>242</i>                       | 1.1       | 3.3       |
| F         | 19.75        | HAST          | 20.32                    | 0.635              | 13,374       | $1.4 \times 10^{-2}$           | 20.0         | <i>100</i>                       | 1.2       | 9.0       |
| F         | 19.75        | HAST          | 30.48                    | 0.635              | 14,101       | $1.4 \times 10^{-2}$           | 33.3         | <i>63</i>                        | 1.3       | 15.1      |
| Cl        | 19.75        | INC           | 5.08                     | 0.635              | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                       | 1.0       | 1.0       |
| Cl        | 19.75        | INC           | 10.16                    | 0.635              | 10,271       | $1.6 \times 10^{-2}$           | 17.4         | <i>88</i>                        | 1.0       | 1.7       |
| Cl        | 19.75        | INC           | 20.32                    | 0.635              | 10,921       | $1.6 \times 10^{-2}$           | 29.3         | <i>56</i>                        | 1.1       | 2.9       |
| Cl        | 19.75        | INC           | 30.48                    | 0.635              | 11,132       | $1.6 \times 10^{-2}$           | 40.0         | <i>41</i>                        | 1.1       | 4.0       |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio compared to  $d_{pipe,outer} = 5.08$  cm

Table 6.22: Comparison of simulated active  $S$  and  $D$  for different pipe thicknesses with all other factors constant

| Salt Type | $E$ (wt%) | Pipe Material | $D_{pipe,outer}$ (cm) | $t_{pipe}$ (cm) | $S$ (cps) | $\sigma_S$ (%) <sup>1</sup> | $D$ (cps) | $\sigma_D$ (%) <sup>1,2</sup> | rel $S$ | rel $D$ |
|-----------|-----------|---------------|-----------------------|-----------------|-----------|-----------------------------|-----------|-------------------------------|---------|---------|
| F         | 19.75     | HAST          | 30.48                 | 0.635           | 14,102    | $1.4 \times 10^{-2}$        | 33.3      | <i>63</i>                     | 1.0     | 1.0     |
| F         | 19.75     | HAST          | 30.48                 | 0.254           | 14,414    | $1.4 \times 10^{-2}$        | 48.8      | <i>44</i>                     | 1.0     | 1.5     |
| Cl        | 19.75     | INC           | 5.08                  | 0.635           | 10,043    | $1.6 \times 10^{-2}$        | 10.1      | <i>148</i>                    | 1.0     | 1.0     |
| Cl        | 19.75     | INC           | 5.08                  | 0.254           | 10,058    | $1.7 \times 10^{-2}$        | 17.1      | <i>88</i>                     | 1.0     | 1.7     |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 6.23: Comparison of simulated active  $S$  and  $D$  for pipes with and without insulation; all fluoride pipe materials are Hastelloy N, all chloride pipe materials are Inconel 625, and all uranium enrichments are 19.75 wt%  $^{235}\text{U}$

| Salt Type | $E$ (wt%) | Pipe Material | $D_{\text{pipe,outer}}$ (cm) | $t_{\text{pipe}}$ (cm) | $S$ (cps) | $\sigma_S$ (%) <sup>1</sup> | $D$ (cps) | $\sigma_D$ (%) <sup>1,2</sup> | rel $S$ <sup>3</sup> | rel $D$ <sup>3</sup> |
|-----------|-----------|---------------|------------------------------|------------------------|-----------|-----------------------------|-----------|-------------------------------|----------------------|----------------------|
| F         | 5.08      | 0.635         | True                         | True                   | 10,956    | $1.6 \times 10^{-2}$        | 2.2       | <i>738</i>                    | 1.0                  | 1.0                  |
| F         | 5.08      | 0.635         | True                         | False                  | 10,956    | $1.6 \times 10^{-2}$        | 2.2       | <i>738</i>                    | 1.0                  | 1.0                  |
| F         | 5.08      | 0.635         | False                        | False                  | 10,956    | $1.6 \times 10^{-2}$        | 2.2       | <i>738</i>                    | 1.0                  | 1.0                  |
| Cl        | 5.08      | 0.635         | True                         | True                   | 10,043    | $1.6 \times 10^{-2}$        | 10.1      | <i>148</i>                    | 1.0                  | 1.0                  |
| Cl        | 5.08      | 0.635         | True                         | False                  | 10,043    | $1.7 \times 10^{-2}$        | 10.1      | <i>148</i>                    | 1.0                  | 1.0                  |
| Cl        | 5.08      | 0.635         | False                        | False                  | 10,043    | $1.6 \times 10^{-2}$        | 10.1      | <i>148</i>                    | 1.0                  | 1.0                  |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

<sup>3</sup>Relative ratio compared to insulation and aluminum jacket on

# Chapter 7

## Feed Monitoring System Options

This chapter outlines some of the potential radiation-based feed measurement options that could be used to meet the safeguards facility-level technical objective of quantifying  $^{235}\text{U}$  in unirradiated, fresh fuel being added to the reactor system. These options are based on results described in Chapter 6.

### 7.1 Assessment of Radiation Monitoring Measurement Methods

Table 7.1 summarizes the results in Chapter 6, describing the information gained by the measurement and the parameters of the feasible application scenarios for both the fluoride and chloride salts.

Note that none of the cases studied directly meets the technical objective of quantifying the  $^{235}\text{U}$  or total uranium added to the reactor system in initial or makeup feed salt. In reality, for the IAEA to independently verify the quantities of nuclear material, it will likely have to combine data from multiple measurements. One option would be to take measurements of the material as it is being added to the system (e.g., in the piping) such that there is enough data to independently verify the nuclear material masses at that point within the process flow.

Table 7.1: Feasibility of measurement methods analyzed for fresh fuel salt monitoring for safeguards purposes

|          | Method                                       | Information gained           | F salt feasibility                                       | Cl salt feasibility                                                       |
|----------|----------------------------------------------|------------------------------|----------------------------------------------------------|---------------------------------------------------------------------------|
| $\gamma$ | 186 keV photopeak                            | $m_{235U}$ in visible volume | all $E$ , $D$ , all $t$                                  | HALEU(except large $D$ ),<br>5%, 2%(in HAST)                              |
|          | 1,001 keV photopeak                          | $m_{238U}$ in visible volume | 2%, HALEU(large $D$ )                                    | NU, some HALEU                                                            |
|          | 186 keV/1,001 keV                            | $E$                          | 2%, HALEU(large $D$ )                                    | some HALEU                                                                |
| $n$      | Passive: total n                             | $m_{234U}$ in visible volume | HALEU, but potentially all<br>$E$ , $D$ , $t$ , pipe mat | HALEU(large $D$ ), but po-<br>tentially all $E$ , $D$ , $t$ , pipe<br>mat |
|          | Passive: coincidence                         | $m_{235U}$ in visible volume | HALEU(large $D$ )                                        | HALEU(very large $D$ )                                                    |
|          | Active with AWCC-like<br>collar: coincidence | $m_{235U}$ in visible volume | none                                                     | none                                                                      |

For example, that may mean combining total neutron counting to get the  $^{234}\text{U}$  content per visible volume, with flow meters and geometries to determine the total volume added, as well as gamma detection (or another method) to determine the  $^{234}\text{U}$  to  $^{235}\text{U}$  ratio. Alternatively, a second option would be to combine data from previous measurements within the facility (e.g., of salt in containers as it sits in storage before being added to the reactor system for irradiation) with in situ measurements as the material is being added to the system. This option likely works well for security MC&A purposes where the uranium enrichment is assumed to be the same as it was when measured upon receipt at the facility. In this case, the facility's MC&A plan may only include measurements of the uranium concentration within the salt, and not the uranium isotopic composition, as fuel enters the process stream. The IAEA may be able to accept similar assumptions in combination with containment and surveillance measures that help maintain confidence in the integrity of previous measurements on the material. These are important considerations because they influence the value of the information obtained by different measurements.

This research focused on providing a feasibility study of a limited number of radiation detection methods that could be used to quantify, or monitor,  $^{235}\text{U}$  and total uranium added to an MSR system in the form of initial or makeup fuel salt. Future computational work could model different detector designs to optimize expected signatures. For example, the work described in Chapter 5 and 6 includes a half-collar detector surrounding the salt-filled pipe when active neutron interrogation was explored. A full collar, similar to that modeled for passive neutron detection, could be used instead. Table 7.2 includes the singles and doubles rates, and their associated relative errors, for half-collar as well as full-collar detector, with all other parameters being equal. Although a full-collar detector increases the efficiency of the detector and the doubles count rates, the very large relative uncertainties still make this detector type and geometry a poor choice for material accountancy of fresh fuel in piping.

Table 7.2: Singles and doubles rates (and relative uncertainties) for active neutron counts with a half- and full-collar detector surrounding a 5.08 cm outer diameter pipe of 0.635 cm thickness

| Salt Type | $E$<br>(wt%) | Pipe Material | Full/Half Collar | $S$<br>(cps) | $\sigma_S$<br>(%) <sup>2</sup> | $D$<br>(cps) | $\sigma_D$<br>(%) <sup>2</sup> |
|-----------|--------------|---------------|------------------|--------------|--------------------------------|--------------|--------------------------------|
| F         | 19           | HAST          | Full             | 16,102       | $1.3 \times 10^{-2}$           | 4.68         | <i>513</i>                     |
| F         | 19           | HAST          | Half             | 10,956       | $1.6 \times 10^{-2}$           | 2.2          | <i>738</i>                     |
| Cl        | 19           | INC           | Full             | 15,270       | $1.3 \times 10^{-2}$           | 22.3         | <i>102</i>                     |
| Cl        | 19           | INC           | Half             | 10,043       | $1.7 \times 10^{-2}$           | 10.1         | <i>148</i>                     |
| F         | NU           | HAST          | Full             | 16,090       | $1.3 \times 10^{-2}$           | 0.23         | <i>10,570</i>                  |
| F         | NU           | HAST          | Half             | 10,940       | $1.6 \times 10^{-4}$           | 0.12         | <i>13,400</i>                  |
| Cl        | NU           | INC           | Full             | 15,181       | $1.4 \times 10^{-4}$           | 1.36         | <i>1,660</i>                   |
| Cl        | NU           | INC           | Half             | 9,946        | $1.7 \times 10^{-2}$           | 0.63         | <i>2,350</i>                   |

<sup>1</sup>60 min count time

<sup>2</sup>Uncertainties in *italics* indicate the associated  $D$  do not meet feasibility criteria of a signal  $4\sigma$  greater than background

Table 7.1 only analyzes the detectors modeled for this research. Other detector types or geometries that use the same methods may have different applications and feasibilities for use for safeguards and security applications. This table is also limited to unirradiated, uranium-bearing salt applications. Separate analyses would have to be done for salts with other nuclear materials (e.g., thorium, plutonium). However, the tool kit developed for this research can be modified such that the source definitions in the MCNP input files are changed out with the rest of the pipeline remaining the same. This would allow analysis of other fuel salts. If the detector geometries or types of detectors are changed out, this tool kit can also be used to analyze irradiated salts in pipes or tanks.

Although the AWCC-like detector modeled as a collar around the pipe caused undesirable relative uncertainties, coincidence counting with active neutron interrogation is still a promising methodology for material accountancy in fresh fuel salts that would benefit from additional R&D. For applications where long count times (e.g., several hours) would be feasible due to operations, this detector setup may be reasonable. For example, if fresh fuel salt with achaleu levels of enrichment was sitting in a tank inside an inaccessible area and tank quantities were only changed in periods less frequent than several hours, this detector setup may be useful. This type of measurement scenario may occur if MSR vendors load their fresh fuel into tanks before adding them to the reactor system, or keep makeup salt in tanks only making additions to the system in batches.

Additionally, other detector models could be analyzed to determine their feasibility before future experiments with uranium-bearing salt in piping. Most fast neutron coincidence counting instruments have relied on detection of thermal neutrons, but the IAEA has more recently been relying on the use of fast-neutron detectors. When counting fast coincident neutrons, induced fission neutrons can be detected shortly after a fission event (i.e., within nanoseconds). Thus, gate widths can be reduced from microseconds to nanoseconds, significantly reducing the accidentals counts [74, 95]. The fast neutron coincidence collar (FNCL), developed by the IAEA, uses 12

liquid scintillator detectors (EJ-309) and an AmLi source to interrogate LWR fuel assemblies (see Figure 7.1) [74]. A liquid scintillator-based coincidence counter should be explored for use with active neutron interrogation to quantify  $^{235}\text{U}$  in unirradiated uranium-based fuel salt.

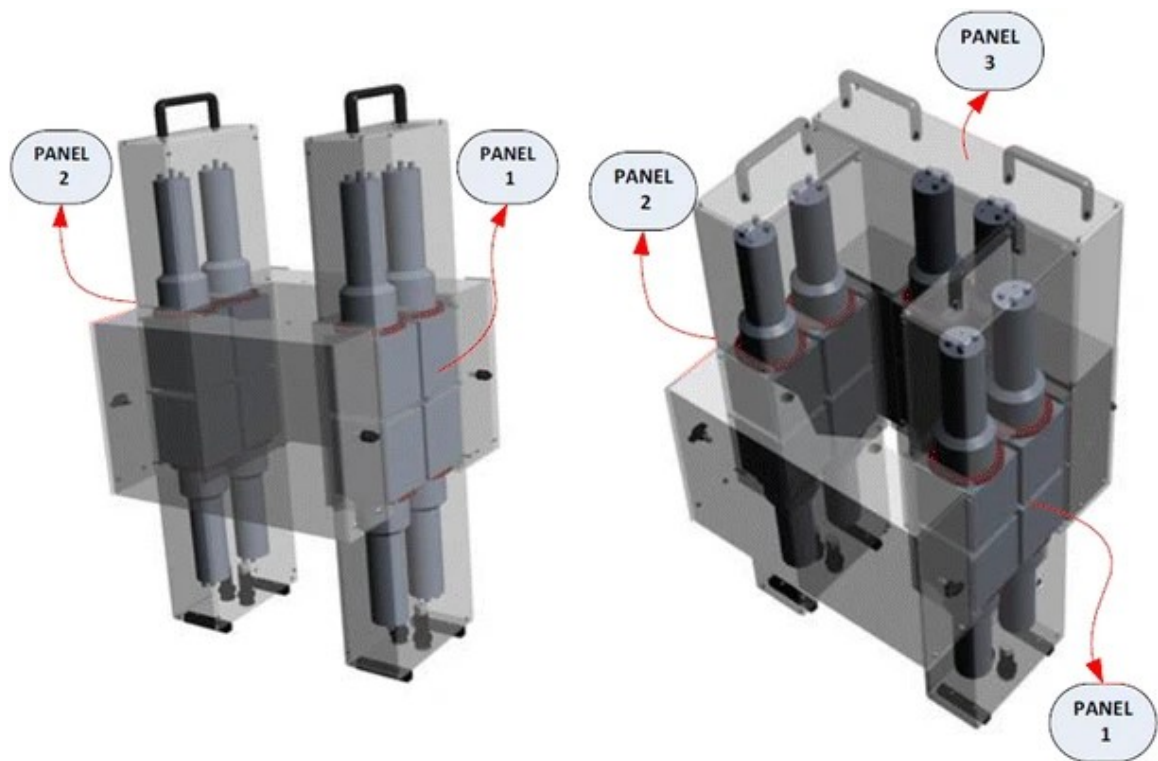


Figure 7.1: The IAEA's fast neutron coincidence collar [74]

# Chapter 8

## Conclusions

Organizations and countries around the world are pursuing advanced nuclear reactor designs as a way to meet climate change goals and capitalize on features that improve fuel efficiency, reduce waste, and incorporate passive safety features. Among others, the IAEA, the US National Academies, and the US NNSA have stated the need for safeguards and security by design in these advanced reactors [34, 36, 37]. Liquid-fueled MSRs are the most distinct from existing reactors implementing MC&A for safeguards or security purposes. While there are challenges because of how distinct liquid-fueled MSRs are from other facility-types, MC&A at liquid-fueled MSR facilities is not insurmountable. There is a path to effective safeguards and security implementation in potential future liquid-fueled MSR facilities, leveraging MC&A. However, MC&A at these facilities will require resources for planning, integration into designs, and implementation in future facilities. It is likely that MC&A costs for safeguards and security applications will be higher than those for other reactor facilities, but this may not be true compared to other bulk facilities. Given the differences in fuel cycles for each proposed advanced reactor type, the author recommends that MC&A costs and safeguards challenges always be considered in context with the proposed fuel cycle options. Liquid-fueled MSR MC&A challenges and costs should consider enrichment, conversion, fuel synthesis, reactor, irradiated salt management, and waste

facilities. Comparisons only across reactor types gives an incomplete, and, in the author's opinion, inaccurate, perspective.

This research proposed a methodology to prioritize the areas of MC&A that are most in need of research effort. The author identified safeguards-relevant design features for liquid-fueled MSR facilities and outlined their relevancy for both IAEA safeguards and security. Then, MC&A facility-level technical objectives were defined throughout the facility. The author identified the need to further evaluate technical methods to quantify nuclear material in unirradiated fuel salt entering the reactor system as the highest priority technical objective based on the (i) time until the challenge is expected, (ii) largest gap compared to existing technologies and practices, and (iii) diversion or misuse implications if a diversion/misuse does occur.

A modeling tool kit was developed to generate radiation signatures from fuel salts representative of the nearest-term liquid-fueled MSR concepts in the US and assess the feasibility of using passive gamma spectrometry, passive neutron total neutron counting, passive neutron coincidence counting, and coincidence counting with active neutron interrogation for MC&A of uranium in feed fuel salt. This toolkit uses SCALE/ORIGEN to generate passive source terms from salts with different chemical and isotopic compositions of unirradiated, uranium-bearing fuel salt. These source terms are used as source definitions in Monte Carlo transport, analyzing the simulated detector response to these emissions in NaI and HPGe gamma detectors and a moderated  $^3\text{He}$  tube collar modeled outside of the piping, representing in situ measurements of material in process streams. Additionally, a moderated  $^3\text{He}$  tube half-collar detector was modeled to analyze the response to active neutron interrogation by an AmLi source placed on the opposite side of the pipe.

A broad range of design parameters representing current liquid-fueled MSR design concepts was assessed to determine the feasibility of using these modeled detectors to quantify nuclear material in salt as it sits (or potentially moves) within a pipe. The parameters assessed include a fluoride-based salt ( $\text{LiF}\text{--}\text{BeF}_2\text{--}\text{UF}_4\text{--}\text{ZrF}_4$  (65–29.1–0.9–5 mol%)), a chloride-based salt ( $\text{NaCl}\text{--}\text{UCl}_3$  (66–34 mol%)); four uranium enrichments

(natural uranium, 2 wt%  $^{235}\text{U}$ , 5 wt%  $^{235}\text{U}$ , and 19.75 wt%  $^{235}\text{U}$ ); two pipe materials (Hastelloy N and Inconel 625), multiple pipe outer diameters ranging from 2.54 to 20.38 cm; multiple pipe thicknesses ranging from 0.254 to 2.54 cm; and the optional inclusion of insulation material and an aluminum jacket external to the pipe to reduce the temperature. Simulated detector responses in the gamma and neutron detectors were assessed to determine which radiation-based methods seem the most promising for future use in material accountancy. Gamma results were assessed based on whether the 186 and 1,001 keV gamma peaks were well-defined and whether the relative uncertainties were low enough such that peak areas would be at least  $4\sigma$  above background. Passive neutron results were assessed for total neutron counting based on whether Singles count rates were high enough and relative uncertainties low enough such that the mass would likely be  $4\sigma$  above background (which is assumed to be equal to the accidentals count rate). Both passive and active neutron results were assessed for coincidence counting based on whether Doubles count rates were high enough and relative uncertainties low enough such that the fissile masses would likely be above minimum detectable limits.

In general, no measurement technique worked well for both salt types and all design parameters assessed. However, gamma 186 keV photopeak area measurements seem promising to quantify the  $^{235}\text{U}$  mass in the volume visible to the detector. Additional work needs to be done to assess how this information could be related to the overall  $^{235}\text{U}$  mass in initial and makeup fuel salt. Additionally, passive total neutron counting seems promising for fuel salts using HALEU enrichment levels near 20 wt%  $^{235}\text{U}$  to quantify  $^{234}\text{U}$  mass in the volume visible to the detector. Passive neutron coincidence counting and coincidence counting with active neutron interrogation may be useful in large diameter pipes with HALEU. This research modeled a moderated detection collar similar to the IAEA's AWCC in thermal mode. This detector system is not ideal for active neutron interrogation in this measurement application because of the extremely high accidentals rates that caused high relative

uncertainties. A fast neutron detection system, potentially similar to the IAEA's FNCL should be explored for feasibility.

While this work looked specifically at applications in liquid-fueled MSR facilities, these results are directly extendable to material accountancy approaches in other facility types or process streams that will also handle unirradiated, uranium-bearing fuel salts. These may include process streams for salt synthesis or any waste processing streams containing unirradiated salt. The value of this toolkit and methodology is also extendable to evaluate use of radiation detection for monitoring unirradiated fuel salt (or salt components) in tanks at MSR, fuel synthesis, or waste facilities through minor modifications of the pipe geometry to model that of a tank. This toolkit and methodology could be extended to look at irradiated salts in piping or tanks through modifications to the source term. Other publications describe models of various liquid-fueled MSR designs that can be used to generate inventories of irradiated salts at various locations within the reactor system [96, 97]. These inventories could define the source term in the existing pipeline and alternative detector types and geometries could be analyzed.

While liquid-fueled MSR vendors are currently in early design phases, mostly in conceptual design phases, this is an ideal time for developers to integrate recommendations that will facilitate safeguards and security implementation in the future. However, advanced reactor developers would greatly benefit from specific recommendations more similar to design criteria. The high-level guidance documents currently available are informative and help set the foundation for an understanding of international safeguards, but additional quantitative R&D assessments that lead to technical recommendations could lead to notable design changes. The research methodology described here is an important step in identifying high-priority technical objectives that need additional R&D and developing data-driven recommendations for industry and the IAEA on methods for material accountancy in these novel facility types.

## 8.1 Future Work

Future work should be done looking at other detection systems for neutron coincidence counting with neutron interrogation. For example, a liquid scintillator-based coincidence collar should be explored with active neutron interrogation. A nickel or stainless steel reflector could be placed around the side of the AmLi (or other neutron source) that is not facing the pipe to reflect neutrons back into the salt and increase the interrogating source count rate.

Additionally, the neutron- and gamma-based measurement options analyzed within this research should be compared against other radiation-based measurements. For example, further research should continue to look at alpha spectrometers submerged in fuel salt to verify uranium enrichment of fresh fuel salt [98–100]. Additionally, prompt gammas from the active neutron interrogation may provide useful for determining uranium enrichment and uranium isotopic masses. These techniques should be compared against measurement techniques that are not based on penetrating radiation (e.g., optical spectroscopy, electrochemical sensors). This comparison should be done using a structured approach and figures of merit defined in Section 6.1. The author proposes cost, measurement time, measurement precision, level of intrusiveness, burden on the operator, anticipated maintenance requirements, and reliability be considered as figures of merit when assessing multiple options to meet a technical safeguards objective.

This research did not incorporate experimental validation of the computational results because there are currently limited options for measurements on pipes that contain uranium-bearing liquid molten salt. In the future, as more test uranium-bearing molten salt loops come online, these computational results should be compared with experimental results. Densities of salts and assumptions, including the isotropic emission of radiation (i.e., homogeneous salt) and lack of residual material buildup on pipe walls, should be modified, if needed, as more research is completed on these topics.

Additionally, these results and recommendations aimed at assessing whether different measurement techniques were feasible. Further work needs to be done to consider how the information gained from each measurement could be combined with information from other measurements to meet the overall MC&A goal of e.g.,  $^{235}\text{U}$  and total uranium mass in initial and makeup salt added to a reactor system. Optimization could be done to determine the most appropriate detector designs for different specific liquid-fueled MSR designs, if unique detection systems are desired.

Lastly, this work assumed that molten salt could be stationary within a pipe or would provide a constant signature over a period of time. This may not be realistic or feasible in a real facility environment. Additional measurement techniques that take into account small amounts of makeup salt being added (such that there is a short period of time where salt is filling a pipe) or better address salt moving within a pipe should be explored. For example, delayed neutron and gamma signatures from detectors placed downstream of a neutron interrogation source should be explored.

# Bibliography

- [1] Third Way, “2022 Advanced Nuclear Map: Charting a Breakout Year,”  
[https://www.thirdway.org/graphic/  
2022-advanced-nuclear-map-charting-a-breakout-year](https://www.thirdway.org/graphic/2022-advanced-nuclear-map-charting-a-breakout-year), accessed: 2022-02-18. 1
- [2] —, “2021 Update: Map of the Global Market for Advanced Nuclear,”  
[https://www.thirdway.org/memo/  
2021-update-map-of-the-global-market-for-advanced-nuclear](https://www.thirdway.org/memo/2021-update-map-of-the-global-market-for-advanced-nuclear), accessed:  
2022-02-18. 1
- [3] Generation IV International Forum, “Generation IV Systems,”  
[https://world-nuclear.org/information-library/nuclear-fuel-cycle/  
nuclear-power-reactors/generation-iv-nuclear-reactors.aspx](https://world-nuclear.org/information-library/nuclear-fuel-cycle/nuclear-power-reactors/generation-iv-nuclear-reactors.aspx), accessed:  
2021-04-10. 1
- [4] —, “Generation IV Systems,”  
[https://www.gen-4.org/gif/jcms/c\\_59461/generation-iv-systems](https://www.gen-4.org/gif/jcms/c_59461/generation-iv-systems), 2023. 2
- [5] International Atomic Energy Agency, “Advanced Reactor Information  
System,” <https://aris.iaea.org/sites/MSR.html>, accessed: 2022-02-10. 3
- [6] K. Hogue, P. Gibbs, M. Dion, and W. Poore III, “Domestic Safeguards  
Material Control and Accountancy Considerations for Molten Salt Reactors,”  
Oak Ridge National Laboratory, Tech. Rep. ORNL/SPR/150504, 2 2021.  
[Online]. Available: <https://www.osti.gov/biblio/1797666> 4, 7, 15

- [7] A. Swift, K. Hogue, T. Folk, and J. Cooley, “Safeguards Technical Objectives for Thorium Molten Salt Reactor Fuel Cycles,” in *2020 Institute for Nuclear Materials Management Annual Meeting*, 8 2020. [Online]. Available: <https://www.osti.gov/biblio/1763719> 3, 15, 26
- [8] J. H. Shaffer, “Preparation and Handling of Salt Mixtures for the Molten Salt Reactor Experiment,” Oak Ridge National Laboratory, Tech. Rep. ORNL-4616, 1 1971. [Online]. Available: <https://www.osti.gov/biblio/4074869> 3
- [9] AlphaTech Research Corp., “An Affordable, Clean-Energy Future for Everyone, Everywhere,” <https://alphatechresearchcorp.com/info/affordable-clean-energy.php>, date accessed: June 27, 2023. 3, 26
- [10] Government of India Department of Atomic Energy, Bhabha Atomic Research Centre, “Indian Nuclear Power Program,” <https://www.barc.gov.in/randd/artnp.html>, date accessed: June 27, 2023.
- [11] Copenhagen Atomics, “Technology,” <https://www.copenhagenatomics.com/technology/>, date accessed: June 1, 2023. 3
- [12] Exodys Energy, <https://www.exodysenergy.com/>, date accessed: June 1, 2023.
- [13] Flibe Energy, <https://flibe.com/energy/>, date accessed: June 1, 2023.
- [14] CNRS, “Molten Salt Fast Reactor,” *IAEA Advanced Reactor Information System*, accessed 6/1/2023. [Online]. Available: <https://aris.iaea.org/PDF/MSFR.pdf> 3, 26, 30
- [15] Kairos Power, “Technology,” <https://kairospower.com/technology/>, date accessed: June 1, 2023.

- [16] Moltex FLEX, “FLEX Reactor,” <https://www.moltexflex.com/flex-reactor/>, date accessed: June 27, 2023.
- [17] Moltex Energy, “Moltex SSR (Stable Salt Reactors) a Roadmap to Thorium @ ThEC2018,” <https://www.youtube.com/watch?v=n-dXB3sunx0>, May 2019. 26
- [18] —, “Moltex Clean Energy,” <https://www.moltexenergy.com/>, date accessed: June 1, 2023. 26
- [19] Natura Resources, “Molten Salt Research Reactor,” <https://naturaresources.org/molten-salt-research-reactor/>, date accessed: June 1, 2023.
- [20] World Nuclear News, “Draft Design for Molten-Salt Research Reactor Plant in Russia,” <https://www.world-nuclear-news.org/Articles/Draft-design-for-molten-salt-research-reactor-plan>, January 2023. 3
- [21] Seaborg Technologies, “Our Technology,” <https://www.seaborg.com/the-reactor>, date accessed: June 1, 2023. 26
- [22] World Nuclear News, “Operating Permit Issued for Chinese Molten Salt reactor,” <https://www.world-nuclear-news.org/Articles/Operating-permit-issued-for-Chinese-molten-salt-re>, June 2023. 3
- [23] H. Xu, “Progress of TMSR in China,” <https://threeconsulting.com/mt-content/uploads/2021/04/chinatmsr2018.pdf>, September 2018, accessed: September 25, 2022.
- [24] TerraPower, “Molten Chloride Fast Reactor Technology,” <https://www.terrapower.com/our-work/molten-chloride-fast-reactor-technology/>, date accessed: June 1, 2023. 3

- [25] Terrestrial Energy, “Status Report—IMSR-400,” *IAEA Advanced Reactor Information System*, accessed 6/1/2023. [Online]. Available: <https://aris.iaea.org/PDF/IMSR400.pdf> 26
- [26] ThorCon, “Design,” <https://thorconpower.com/design/>, date accessed: June 1, 2023. 3
- [27] Wielenga Innovation Foundation, “Wielenga Innovative Static Salt Reactor,” <https://www.wifound.org/nuclear-reactor>, 2023. 3
- [28] US Department of Energy, Office of Nuclear Energy, “Energy Department’s Advanced Reactor Demonstration Program Awards \$30 Million in Initial Funding for Risk Reduction Projects,” <https://www.energy.gov/ne/articles/energy-department-s-advanced-reactor-demonstration-program-awards-30-million-initial>, accessed: 2021-04-10. 4
- [29] V. Bytchkov and J. Cooley, “IAEA Safeguards System: Implementing the State-Level Concept,” [https://media.nti.org/documents/IAEA\\_Safeguards\\_System-Implementing\\_the\\_State-Level\\_Concept.pdf](https://media.nti.org/documents/IAEA_Safeguards_System-Implementing_the_State-Level_Concept.pdf), accessed: 2021-11-20. 8
- [30] “Atomic Energy Act of 1954,” title I, Chap. 2, Sec. 11.aa. 8
- [31] US Nuclear Regulatory Commission, “Domestic Safeguards,” <https://www.nrc.gov/security/domestic.html>, 2020/3/11. 8
- [32] —, “Material Control and Accounting of Special Nuclear Material,” code of Federal Regulations, Title 10, Part 74. 8
- [33] Hogue, Karen et al., “MC&A Initiatives for Licensing of New Reactor Technologies,” Invited Presentation at the 2023 Nuclear Materials Management and Safeguards System Annual User’s Meeting, June 2023. 8

- [34] International Atomic Energy Agency, “Development and Implementation Support Programme for Nuclear Verification 2022—2023,” International Atomic Energy Agency, Tech. Rep. STR-399, January 2022. 9, 116
- [35] —, “Enhancing Capabilities for Nuclear Verification Resource Mobilization Priorities,” International Atomic Energy Agency, Tech. Rep. STR-400, January 2022. 9
- [36] US Department of Energy, National Nuclear Security Administration, “Remarks by NNSA Administrator Jill Hruby at the INMM/ESARDA Joint Annual Meeting,” <http://energy.gov/nnsa/articles/remarks-nnsa-administrator-jill-hruby-inmmesarda-joint-annual-meeting>, May 2023. 10, 116
- [37] National Academy of Engineering and National Academies of Sciences, Engineering, and Medicine, *Laying the Foundation for New and Advanced Nuclear Reactors in the United States*. Washington, DC: National Academies Press, 2023. [Online]. Available: <https://nap.nationalacademies.org/catalog/26630/laying-the-foundation-for-new-and-advanced-nuclear-reactors-in-the-united-states> 10, 116
- [38] US Department of Energy, National Nuclear Security Administration, Office of Nuclear Materials Integration, NA-532, “Nuclear Materials Management and Safeguards System (NMMSS) Users Guide—Rev. 2.1,” 2017. 13
- [39] A. Worrall, B. R. Betzler, G. Flanagan, D. E. Holcomb, J. Hu, D. N. Kovacic, A. L. Qualls, and L. G. Worrall, “Molten Salt Reactors and Associated Safeguards Challenges and Opportunities,” *Proceedings from the 2018 IAEA Safeguards Symposium*, 11 2018. [Online]. Available: <https://www.osti.gov/biblio/1630525> 15

- [40] N. Dunkle and O. Chvala, “Safeguards by Design Considerations for Modular Molten Salt Reactors,” *Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting*, 8 2021. 15
- [41] M. Dion, M. Greenwood, K. Hogue, S. O’Brien, L. Scott, and G. Westphal, “Material Control & Accountancy for Molten Salt Reactors: FY2021 Report,” *Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting*, 10 2021. [Online]. Available: <https://www.osti.gov/biblio/1840163> 19, 20, 50
- [42] 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. [Online]. Available: <https://doi.org/10.1080/00295450.2020.1724728> 24
- [43] A. Lines, S. Bryan, H. Felmy, and S. Branch, “On-line Monitoring for Molten Salt Reactor MC&A: Optical Spectroscopy-Based Approaches,” *Pacific Northwest National Laboratories*, no. PNNL-31955, September 2021. 25
- [44] C. E. Moore, W. H. Doniger, and N. C. Hoyt, “Assessment of Flow-Enhanced Sensors for Actinide Quantification in MSRs,” *Argonne National Laboratory*, no. ANL/CFCT-22/34, September 2022. 25
- [45] S. Chirayath, R. Aoyagi, and H. Sagara, “Feasibility Study of Nuclear Material Accounting In A Molten Salt Fast Reactor to Develop a Safeguards Approach,” in *Proceedings from the 2020 Institute of Nuclear Materials Management Annual Meeting*, 2020. 25
- [46] M. Cook, “Hybrid K-edge Densitometry as a Method for Materials Accountancy Measurements in Pyrochemical Reprocessing,” Ph.D. dissertation, University of Tennessee, Knoxville, 2015. [Online]. Available: [https://trace.tennessee.edu/utk\\_graddiss/3329](https://trace.tennessee.edu/utk_graddiss/3329) 25

- [47] A. Stubsgaard, “Development Update from Copenhagen Atomics,”  
<https://www.youtube.com/watch?v=hW3YBF5N1-E&list=PLisa2eqmVBFbCTd0-tbSjD6xgR514dmgw&index=21>, October 2022, 2022 Molten Salt Workshop. 26
- [48] S. Patel, “China Approves Commissioning of Thorium-Powered Reactor,”  
<https://www.powermag.com/china-approves-commissioning-of-thorium-powered-reactor/>, November 2022.
- [49] J. Latkowski, “TerraPower’s Molten Chloride Fast Reactor (MCFR),” 7 2021.
- [50] D. Walter, “Molten Chloride Fast Reactor Program Overview,” Presented at the 2022 Molten Salt Reactor Workshop, October 2022. 29, 30
- [51] D. Wilson, “ThorCon: Status 2022,” <https://www.youtube.com/watch?v=WJHlsvKvh4c&list=PLisa2eqmVBFbCTd0-tbSjD6xgR514dmgw&index=20>, October 2022, presented at the 2022 MSR Workshop. 26
- [52] W. Wieselquist and R. A. Lefebvre, “SCALE 6.3.1 User Manual,” Oak Ridge National Laboratory, Tech. Rep. ORNL/TM-SCALE-6.3.1, 2 2023. [Online]. Available: <https://www.osti.gov/biblio/1959594> 29, 35
- [53] R. C. Robertson, “MSRE Design & Operations Report Part 1 Description of Reactor Design,” Oak Ridge National Laboratory, Tech. Rep. ORNL-TM-0728, 1 1965. [Online]. Available: <https://www.osti.gov/biblio/4654707> 29, 30, 51
- [54] G. L. Fredrickson, G. Cao, R. Gakhar, and T. S. Yoo, “Molten Salt Reactor Salt Processing—Technology Status,” Idaho National Laboratory, Tech. Rep. INL/EXT-18-51033, 8 2018. [Online]. Available: <https://www.osti.gov/biblio/1484689> 30
- [55] D. Brown, M. Chadwick, R. Capote, A. Kahler, A. Trkov, M. Herman, A. Sonzogni, Y. Danon, A. Carlson, M. Dunn, D. Smith, G. Hale, G. Arbanas,

- R. Arcilla, C. Bates, B. Beck, B. Becker, F. Brown, R. Casperson, J. Conlin, D. Cullen, M.-A. Descalle, R. Firestone, T. Gaines, K. Guber, A. Hawari, J. Holmes, T. Johnson, T. Kawano, B. Kiedrowski, A. Koning, S. Kopecky, L. Leal, J. Lestone, C. Lubitz, J. M. Damian, C. Mattoon, E. McCutchan, S. Mughabghab, P. Navratil, D. Neudecker, G. Nobre, G. Noguere, M. Paris, M. Pigni, A. Plompen, B. Pritychenko, V. Pronyaev, D. Roubtsov, D. Rochman, P. Romano, P. Schillebeeckx, S. Simakov, M. Sin, I. Sirakov, B. Sleaford, V. Sobes, E. Soukhovitskii, I. Stetcu, P. Talou, I. Thompson, S. van der Marck, L. Welser-Sherrill, D. Wiarda, M. White, J. Wormald, R. Wright, M. Zerkle, G. Aerovnik, and Y. Zhu, “ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data,” *Nuclear Data Sheets*, vol. 148, pp. 1–142, 2018, special Issue on Nuclear Reaction Data. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0090375218300206> 31
- [56] J. Mcfarlane, P. A. Taylor, D. E. Holcomb, and W. Poore III, “Review of Hazards Associated with Molten Salt Reactor Fuel Processing Operations,” Oak Ridge National Laboratory, Tech. Rep. ORNL/TM-2019/1195, 6 2019. [Online]. Available: <https://www.osti.gov/biblio/1543201> 31
- [57] D. Holcomb, personal communication, June 2023. 31
- [58] D. Reilly, N. Ensslin, H. Smith Jr., and S. Kreiner, *Passive Nondestructive Assay of Nuclear Materials*. Los Alamos National Laboratory, 3 1991. [Online]. Available: <https://www.osti.gov/biblio/5428834> 32, 46, 47, 48, 69, 86
- [59] H. R. von Gunten, A. Grütter, H. W. Reist, and M. Baggenstos, “Ground-State Spontaneous-Fission Half-Lives of Uranium Isotopes,” *Phys. Rev. C*, vol. 23, pp. 1110–1112, Mar 1981. [Online]. Available: <https://link.aps.org/doi/10.1103/PhysRevC.23.1110> 32

- [60] N. Soppera, E. Dupont, and M. Bossant, *JANIS Book of alpha-induced cross-sections (2012)*, 06 2012. 34
- [61] D. P. Griesheimer, A. T. Pavlou, J. T. Thompson, J. C. Holmes, M. L. Zerkle, E. Caro, and H. Joo, “In-line ( $\alpha$ ,n) Source Sampling Methodology for Monte Carlo Radiation Transport Simulations,” *Nuclear Engineering and Technology*, vol. 49, no. 6, pp. 1199–1210, 2017, special Issue on International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering 2017 (M&C 2017). [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S1738573317303017> 35
- [62] W. B. Wilson, R. T. Perry, W. S. Charlton, T. A. Parish, and E. F. Shores, “SOURCES: a code for calculating ( $\alpha$ ,n), spontaneous fission, and delayed neutron sources and spectra,” *Radiation Protection Dosimetry*, vol. 115, no. 1-4, pp. 117–121, 12 2005. [Online]. Available: <https://doi.org/10.1093/rpd/nci260> 35
- [63] W. Charlton, R. Perry, and W. Wilson, “Benchmarking of the Los Alamos neutron production rate code SOURCES-3A,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 140, no. 1, pp. 1–8, 1998. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0168583X98000123>
- [64] G. E. McMath and G. W. McKinney, “MCNP6 Enhancements to Alpha Particle Production and Transport,” *ANS Winter Meeting and Nuclear Technology Expo*, no. LA-UR-14-25169, 2014. [Online]. Available: [https://mcnp.lanl.gov/pdf\\_files/TechReport\\_2014\\_LANL\\_LA-UR-14-25169\\_McMathMcKinney.pdf](https://mcnp.lanl.gov/pdf_files/TechReport_2014_LANL_LA-UR-14-25169_McMathMcKinney.pdf) 35
- [65] P. Schubert, “Letter to Director, Office of Nuclear Reactor Regulation US Nuclear Regulatory Commission,” 2020. [Online]. Available: <https://www.nrc.gov/docs/ML2024/ML20241A071.pdf> 45

- [66] International Atomic Energy Agency, *Safeguards Techniques and Equipment: 2011 Edition*. International Atomic Energy Agency, 2011, no. International Nuclear Verification Series No. 1 (Rev. 2). 46, 48, 63
- [67] R. Venkataraman, personal communication, July 2023. 46, 74
- [68] S. E. Skutnik and O. Chvala, “Development of an MC&A toolbox for liquid-fueled molten salt reactors with online reprocessing: Final Report,” 10 2022. [Online]. Available: <https://www.osti.gov/biblio/1958864> 46
- [69] M. Bootharajan, K. Swaminathan, and C. Venkata Subramani, “Assay of actinides in molten salt matrix using K-edge densitometry,” *Proceedings of the 12th DAE-BRNS national symposium on nuclear and radiochemistry*, pp. 226–227, 2015. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0141391008002516> 46
- [70] R. Venkataraman, F. Bronson, V. Abashkevich, B. Young, and M. Field, “Validation of in situ object counting system (isocs) mathematical efficiency calibration software,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 422, no. 1, pp. 450–454, 1999. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0168900298011152> 47
- [71] Mirion Technologies, “Model S573 ISOCS Calibration Software,” [https://mirionprodstorage.blob.core.windows.net/prod-20220822/cms4\\_mirion/files/pdf/spec-sheets/c40166s573-isocs-calibration-software.pdf](https://mirionprodstorage.blob.core.windows.net/prod-20220822/cms4_mirion/files/pdf/spec-sheets/c40166s573-isocs-calibration-software.pdf), Apr 2013. 47
- [72] T. D. Reilly, E. R. Martin, J. L. Parker, L. G. Speir, and R. B. Walton, “A continuous in-line monitor for uf6 enrichment,” *Nuclear Technology*, vol. 23, no. 3, pp. 318–327, 1974. [Online]. Available: <https://doi.org/10.13182/NT74-A15924> 47

- [73] Statement of Gene Aloise, “DOE’s Depleted Uranium Tails Could Be a Source of Revenue for the Government,” 2011. [Online]. Available: <https://www.gao.gov/assets/gao-11-752t.pdf> 47
- [74] J. S. Beaumont, T. H. Lee, M. Mayorov, C. Tintori, F. Rogo, B. Angelucci, and M. Corbo, “A Fast-Neutron Coincidence Collar Using Liquid Scintillators for Fresh Fuel Verification,” *Journal of Radioanalytical and Nuclear Chemistry*, vol. 314, p. 803–812, 11 2017. [Online]. Available: <https://doi.org/10.1007/s10967-017-5412-x> 48, 113, 114, 115
- [75] Werner, C.J. (editor), “MCNP Users Manual—Code Version 6.2,” Los Alamos National Laboratory, Tech. Rep. LA-UR-17-29981, 2017. 48
- [76] G. Van Rossum and F. L. Drake, *Python 3 Reference Manual*. Scotts Valley, CA: CreateSpace, 2009. 48
- [77] A. M. Wheeler, V. Singh, L. F. Miller, and O. Chvala, “Initial calculations for source term of Molten Salt Reactors,” *Progress in Nuclear Energy*, vol. 132, p. 103616, 2021. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0149197020303619> 50
- [78] J. Latkowski, personal communication, July 2022. 51
- [79] D. Andersson and B. Beeler, “Ab initio molecular dynamics (aimd) simulations of nacl, ucl<sub>3</sub> and nacl-ucl<sub>3</sub> molten salts,” *Journal of Nuclear Materials*, vol. 568, p. 153836, 2022. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0022311522003221> 51
- [80] G. van Oudenaren, J. Ocadiz-Flores, and A. Smith, “Coupled structural-thermodynamic modelling of the molten salt system NaCl–UCl<sub>3</sub>,” *Journal of Molecular Liquids*, vol. 342, p. 117470, 2021. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0167732221021942> 51

- [81] R. Hu, G. Hu, M. Gorman, J. Fang, T. Mui, D. O’Grady, T. Fei, and R. Salko, “FY21 SAM Developments for MSR Modeling,” Argonne National Laboratory, Tech. Rep. ANL/NSE-21/74, 2021. 52
- [82] S. E. Beall, P. N. Haubenreich, R. B. Lindauer, and J. R. Tallackson, “MSRE Design and Operations Report—Part V—Reactor Safety Analysis Report,” Oak Ridge National Laboratory, Tech. Rep. ORNL-TM-732, 8 1964. [Online]. Available: <https://www.osti.gov/biblio/4034157> 52
- [83] Special Metals, “Inconel Alloy 625,” <https://www.specialmetals.com/documents/technical-bulletins/inconel/inconel-alloy-625.pdf>, 2013, date accessed: December 6, 2021. 52, 53
- [84] K. Robb, “Status and Direction of Molten Salt Loops at ORNL,” [https://msrworkshop.ornl.gov/wp-content/uploads/2021/11/23-Robb\\_MSRworkshop3\\_crop.pdf](https://msrworkshop.ornl.gov/wp-content/uploads/2021/11/23-Robb_MSRworkshop3_crop.pdf), October 2021, presented at the 2021 MSR Workshop. 52
- [85] —, personal communication, January 2023. 52, 54
- [86] HAYNES International, “HASTELLOY N alloy,” [https://www.haynesintl.com/alloys/alloy-portfolio\\_/Corrosion-resistant-Alloys/hastelloy-n-alloy/principle-features](https://www.haynesintl.com/alloys/alloy-portfolio_/Corrosion-resistant-Alloys/hastelloy-n-alloy/principle-features), 2023. 53
- [87] ZRCI Refractory Composites, “SB-2000 Blanket,” <https://www.zrci.com/material/sb-2000-blanket-3/>, 2019, date accessed: January 6, 2023. 54, 55
- [88] McMaster-Carr, “Aluminum Pipe Insulation Jacketing Sheet,” <https://www.mcmaster.com/9669K51/>, date accessed: January 6, 2023. 54
- [89] L. E. Smith and A. R. Lebrun, “Design, modeling and viability analysis of an online uranium enrichment monitor,” in *2011 IEEE Nuclear Science Symposium Conference Record*, 2011, pp. 1030–1037. 56, 58

- [90] T. Packer, M. Wormald, C. Charlier, W. Bush, and I. Cherradi, “Review of the current status of CEMO installed at gas centrifuge enrichment plants,” International Atomic Energy Agency, Tech. Rep. IAEA-SM-351/169, 1997. [Online]. Available: [https://inis.iaea.org/collection/NCLCollectionStore/\\_Public/30/014/30014884.pdf?r=1](https://inis.iaea.org/collection/NCLCollectionStore/_Public/30/014/30014884.pdf?r=1) 56
- [91] L. E. Smith, A. R. Lebrun, and R. Labella, “Potential Roles for Unattended Safeguards Instrumentation at Centrifuge Enrichment Plants,” *Journal of Nuclear Materials Management*, vol. XLII, no. I, Fall 2013. 58
- [92] J. Araujo, W. Waldman, and M. De Paoli, “Thermal Properties of High Density Polyethylene Composites with Natural Fibres: Coupling Agent Effect,” *Polymer Degradation and Stability*, vol. 93, no. 10, pp. 1770–1775, 2008. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0141391008002516> 63
- [93] J. S. Hendricks, M. T. Swinhoe, and A. Favalli, *Monte Carlo N-Particle Simulations for Nuclear Detection and Safeguards*. Washington, DC: Springer Cham, 2022. 64, 85
- [94] M. Berger, J. Hubbell, S. Seltzer, J. Chang, J. Coursey, R. Sukumar, D. Zucker, and K. Olsen, “XCOM: Photon Cross Section Database (version 1.5),” <http://physics.nist.gov/xcom>, 2010. 75
- [95] J. Beaumont, T. Lee, M. Mayorov, J. K. Jeon, A. Bonino, M. Grund, M. Dutra, G. Renha, S. H. Ahn, K. H. Kim, J. S. Park, and G. H. Yang, “Field testing of a fast-neutron coincidence collar for fresh uranium fuel verification,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 962, p. 163682, 2020. [Online]. Available: <https://www.sciencedirect.com/science/article/pii/S0168900220302461> 113

- [96] M. S. Greenwood, B. R. Betzler, A. L. Qualls, J. Yoo, and C. Rabiti, “Demonstration of the Advanced Dynamic System Modeling Tool TRANSFORM in a Molten Salt Reactor Application via a Model of the Molten Salt Demonstration Reactor,” *Nuclear Technology*, vol. 206, no. 3, 7 2019. [Online]. Available: <https://www.osti.gov/biblio/1543221> 119
- [97] N. Shoman and M. Higgins, “Material Accountancy for Molten Salt Reactors: Challenges and Opportunities,” Sandia National Laboratories, Albuquerque, NM, Tech. Rep. SAND2023-01845R, 2023. [Online]. Available: [https://energy.sandia.gov/wp-content/uploads/2023/04/MSRSG\\_pub\\_nshoman\\_final.pdf](https://energy.sandia.gov/wp-content/uploads/2023/04/MSRSG_pub_nshoman_final.pdf) 119
- [98] E. Lukosi, “Microfluidic alpha spectrometry of uox pwr unf in a molten salt,” *Nuclear Science and Engineering*, vol. 188, no. 3, pp. 294–302, 2017. [Online]. Available: <https://doi.org/10.1080/00295639.2017.1367248> 120
- [99] J. Jarell, “Fabrication and Characterization of a Molten Salt Application Silicon Carbide Alpha Detector,” Ph.D. dissertation, The Ohio State University, 2018. [Online]. Available: [http://rave.ohiolink.edu/etdc/view?acc\\_num=osu1533219324268259](http://rave.ohiolink.edu/etdc/view?acc_num=osu1533219324268259)
- [100] K. C. Goetz, F. K. Reed, N. Taylor, and N. D. Bull Ezell, “Considerations of Radiation-Hardened Electronics for Alpha Detectors in Molten Salt Reactors,” Oak Ridge National Laboratory, Tech. Rep. ORNL/TM-2022/2574, Sep 2022. [Online]. Available: <https://www.osti.gov/biblio/1890315> 120

# Appendix

# Appendix A

## SCALE/ORIGEN Input Files

### A.1 Fluoride-based salt

```
' FLiBe salt with NU [LiF-BeF2-UF4-ZrF4 (65-29.1-0.9-5 mol\%)]
=origen

bounds{
  neutron= [ 0      100  500 1E3
            1098i   10E3 11E6 ] \% 10keV bins, from 10 keV to 11 MeV
  gamma= [ 0      0.01  1
           1298i   1E3  1.3E6 ] \% 1 keV bins, from 1 keV to 1.3 MeV
}

solver{type=CRAM}

\% E = NU
case(FLiBe_NU) \{
  lib{ file="end7dec" }
  mat{ units="grams" iso=[ U-234 = 2.74808E-06
                           U-235 = 0.000369419
                           U-238 = 0.050907565
                           Li-6  = 0.0000109162
                           Li-7  = 0.109150743 % assumes enriched to 99.99\% Li-7
                           Be    = 0.062776361
                           Zr     = 0.109182304
                           F      = 0.667599943] }
  time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
  power = [0 0 0 0]
  print{
    cutoffs=[all=1e-16]
```

```

    neutron {summary=yes spectra=yes}
    gamma{summary=yes spectra=yes}
}

neutron{
    alphan_medium=CASE
    alphan_bins=200    \% use 200 bins for alpha slowing down calculation
    alphan_cutoff=0.0  \% cutoff for alpha,n calculation
    alphan_step=LAST   \% step index in this case
}

gamma{
    sublib=ALL          \% LT, FP, or AC single sublib
                        \% ALL = all sub-libraries
    continuum=yes       \% expand continuum data stored as
                        \%   lines into proper continuua
    immediate=yes       \% load lines for immediate gamma/x-rays
    spont=yes           \% include photons from spont fission
    brem_medium=u02     \% include bremsstrahlung from betas
}

save{ file="FLiBe_NU.f71" steps=all }
}

\% E = 2wt\%
case(FLiBe_2) {
    lib{ file="end7dec" }
    mat{ units="grams" iso=[ U-234 = 9.24223E-06
                             U-235 = 0.001038452
                             U-238 = 0.050232038
                             Li-6  = 0.0000109162
                             Li-7  = 0.109150743 \% assumes enriched to 99.99\% Li-7
                             Be    = 0.062776361

                             Zr     = 0.109182304
                             F      = 0.667599943]}
    time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
    power = [0 0 0 0]
    print{
        cutoffs=[all=1e-16]
        neutron {summary=yes spectra=yes}
        gamma{summary=yes spectra=yes}
    }

    neutron{
        alphan_medium=CASE
        alphan_bins=200    \% use 200 bins for alpha slowing down calculation
        alphan_cutoff=0.0  \% cutoff for alpha,n calculation
        alphan_step=LAST   \% step index in this case
    }

    gamma{

```

```

        sublib=ALL      \% LT, FP, or AC single sublib
                        \% ALL = all sub-libraries
        continuum=yes   \% expand continuum data stored as
                        \%   lines into proper continuua
        immediate=yes   \% load lines for immediate gamma/x-rays
        spont=yes       \% include photons from spont fiss
        brem_medium=u02 \% include bremsstrahlung from betas
    }

    save{ file="FLiBe_2.f71" steps=all }
}

\% E = 5 wt\%
case(FLiBe_5) {
    lib{ file="end7dec" }
    mat{ units="grams" iso=[ U-234 = 2.30968E-05
                             U-235 = 0.002595147
                             U-238 = 0.048661489
                             Li-6  = 0.0000109162
                             Li-7  = 0.109150743 \% assumes enriched to 99.99\% Li-7
                             Be    = 0.062776361

                             Zr     = 0.109182304
                             F      = 0.667599943]}
    time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
    power = [0 0 0 0]
    print{
        cutoffs=[all=1e-16]
        neutron {summary=yes spectra=yes}
        gamma{summary=yes spectra=yes}
    }

    neutron{
        alphan_medium=CASE
        alphan_bins=200    \% use 200 bins for alpha slowing down calculation
        alphan_cutoff=0.0  \% cutoff for alpha,n calculation
        alphan_step=LAST   \% step index in this case
    }

    gamma{
        sublib=ALL      \% LT, FP, or AC single sublib
                        \% ALL = all sub-libraries
        continuum=yes   \% expand continuum data stored as
                        \%   lines into proper continuua
        immediate=yes   \% load lines for immediate gamma/x-rays
        spont=yes       \% include photons from spont fiss
        brem_medium=u02 \% include bremsstrahlung from betas
    }

    save{ file="FLiBe_5.f71" steps=all }
}

```

```

case(FLiBe_19) {
  lib{ file="end7dec" }
  mat{ units="grams" iso=[ U-234 = 9.10623E-05
                           U-235 = 0.01023172
                           U-238 = 0.04095695
                           Li-6  = 0.0000109162
                           Li-7  = 0.109150743 \% assumes enriched to 99.99\% Li-7
                           Be    = 0.062776361

                           Zr     = 0.109182304
                           F      = 0.667599943] }
  time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
  power = [0 0 0 0]
  print{
    cutoffs=[all=1e-16]
    neutron {summary=yes spectra=yes}
    gamma{summary=yes spectra=yes}
  }

  neutron{
    alphan_medium=CASE
    alphan_bins=200    \% use 200 bins for alpha slowing down calculation
    alphan_cutoff=0.0  \% cutoff for alpha,n calculation
    alphan_step=LAST   \% step index in this case
  }

  gamma{
    sublib=ALL          \% LT, FP, or AC single sublib
                      \% ALL = all sub-libraries
    continuum=yes       \% expand continuum data stored as
                      \%   lines into proper continuua
    immediate=yes        \% load lines for immediate gamma/x-rays
    spont=yes           \% include photons from spont fiss
    brem_medium=u02     \% include bremsstrahlung from betas
  }

  save{ file="FLiBe_19.f71" steps=all }
}

end

=opus
title="FLiBe NU sf"
data="FLiBe_NU.f71"
typarams=sfspec
units=particles
end

=opus
title="FLiBe NU (alpha,n)"
data="FLiBe_NU.f71"

```

```

typarams=anspec
units=particles
end

=opus
data="FLiBe_NU.f71"
title="FLiBe NU total n"
typarams=nspectrum
units=particles
end

=opus
title="FLiBe NU photons"
data="FLiBe_NU.f71"
typarams=gspectrum
units=particles
end

=opus
title="FLiBe 2wt\% sf"
data="FLiBe_2.f71"
typarams=sfspec
units=particles
end

=opus
title="FLiBe 2wt\% (alpha,n)"
data="FLiBe_2.f71"
typarams=anspec
units=particles
end

=opus
title="FLiBe 2wt\% total n"
data="FLiBe_2.f71"
typarams=nspectrum
units=particles
end

=opus
title="FLiBe 2wt\% photons"
data="FLiBe_2.f71"
typarams=gspectrum
units=particles
end

=opus
title="FLiBe 5wt\% sf"
data="FLiBe_5.f71"
typarams=sfspec
units=particles

```

```

end

=opus
title="FLiBe 5wt\% (alpha,n)"
data="FLiBe_5.f71"
typarams=anspec
units=particles
end

=opus
title="FLiBe 5wt\% total n"
data="FLiBe_5.f71"
typarams=nspectrum
units=particles
end

=opus
title="FLiBe 5wt\% photons"
data="FLiBe_5.f71"
typarams=gspectrum
units=particles
end

=opus
title="FLiBe 19.75wt\% sf"
data="FLiBe_19.f71"
typarams=sfspec
units=particles
end

=opus
title="FLiBe 19.75wt\% (alpha,n)"
data="FLiBe_19.f71"
typarams=anspec
units=particles
end

=opus
title="FLiBe 19.75wt\% total n"
data="FLiBe_19.f71"
typarams=nspectrum
units=particles
end

=opus
title="FLiBe 19.75wt\% photons"
data="FLiBe_19.f71"
typarams=gspectrum
units=particles
end

```

## A.2 Chloride-based salt

```
' Cl salt with NU, 2, 5, 19.75 [NaCl-UC13 (66-34 mol%)]
=origen

bounds{ % defines tally bins
    neutron= [ 0    100  500 1E3
               1098i  10E3 11E6 ] %    10keV bins, from 10 keV to 11 MeV
    gamma= [ 0    0.01    1
             1298i  1E3  1.3E6 ] %    1 keV bins, from 1 keV to 1.2 MeV
}
solver{type=CRAM}

% E = NU
case(Cl_NU) {
    lib{ file="end7dec" }
    mat{ units="grams" iso=[
        U-234 = 2.75604E-05
                                U-235 = 0.003704893
                                U-238 = 0.51055032
                                Na    = 0.096420973

        Cl-35 = 0.097324063
        Cl-37 = 0.29197219]}
    time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
    power = [0 0 0 0 ]
    print{
        cutoffs=[all=1e-16]
        neutron {summary=yes spectra=yes}
        gamma{summary=yes spectra=yes}
    }

    neutron{
        alphan_medium=CASE
        alphan_bins=200    % use 200 bins for alpha slowing down calculation
        alphan_cutoff=0.0  % cutoff for alpha,n calculation
        alphan_step=LAST   % step index in this case
    }

    gamma{
        sublib=ALL          % LT, FP, or AC single sublib
                           % ALL = all sub-libraries
        continuum=yes       % expand continuum data stored as
                           %   lines into proper continua
        immediate=yes       % load lines for immediate gamma/x-rays
        spont=yes           % include photons from spont fission
        brem_medium=uo2     % include bremsstrahlung from betas
    }

    save{ file="Cl_NU.f71" steps=all }
}
```

```

% E = 2 wt%
case(C1_2) {
  lib{ file="end7dec" }
  mat{ units="grams" iso=[
    U-234 = 9.269E-05
                                U-235 = 0.010414606
                                U-238 = 0.503775478
                                Na    = 0.096420973

    C1-35 = 0.097324063
    C1-37 = 0.29197219]]}
  time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
  power = [0 0 0 0 ]
  print{
    cutoffs=[all=1e-16]
    neutron {summary=yes spectra=yes}
    gamma{summary=yes spectra=yes}
  }

  neutron{
    alphan_medium=CASE
    alphan_bins=200    % use 200 bins for alpha slowing down calculation
    alphan_cutoff=0.0 % cutoff for alpha,n calculation
    alphan_step=LAST   % step index in this case
  }

  gamma{
    sublib=ALL          % LT, FP, or AC single sublib
                        % ALL = all sub-libraries
    continuum=yes       % expand continuum data stored as
                        %   lines into proper continuua
    immediate=yes       % load lines for immediate gamma/x-rays
    spont=yes           % include photons from spont fiss
    brem_medium=u02     % include bremsstrahlung from betas
  }

  save{ file="C1_2.f71" steps=all }
}

% E = 5 wt%
case(C1_5) {
  lib{ file="end7dec" }
  mat{ units="grams" iso=[
    U-234 = 0.000231637
                                U-235 = 0.026026645
                                U-238 = 0.488024491
                                Na    = 0.096420973

    C1-35 = 0.097324063
    C1-37 = 0.29197219]]}
  time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
  power = [0 0 0 0 ]

```

```

print{
    cutoffs=[all=1e-16]
    neutron {summary=yes spectra=yes}
    gamma{summary=yes spectra=yes}
}

neutron{
    alphan_medium=CASE
    alphan_bins=200    % use 200 bins for alpha slowing down calculation
    alphan_cutoff=0.0  % cutoff for alpha,n calculation
    alphan_step=LAST   % step index in this case
}

gamma{
    sublib=ALL          % LT, FP, or AC single sublib
                        % ALL = all sub-libraries
    continuum=yes       % expand continuum data stored as
                        %   lines into proper continuua
    immediate=yes       % load lines for immediate gamma/x-rays
    spont=yes           % include photons from spont fiss
    brem_medium=u02     % include bremsstrahlung from betas
}

save{ file="Cl_5.f71" steps=all }
}

% E=19.75wt%
case(Cl_19) {
    lib{ file="end7dec" }
    mat{ units="grams" iso=[
        U-234 = 0.000913261
                                U-235 = 0.102613592
                                U-238 = 0.410755921
                                Na    = 0.096420973

        Cl-35 = 0.097324063
        Cl-37 = 0.29197219]]}
    time{units="years" t=[ 0.019231 0.083333 1.0 10.0 ] }
    power = [0 0 0 0 ]
    print{
        cutoffs=[all=1e-16]
        neutron {summary=yes spectra=yes}
        gamma{summary=yes spectra=yes}
    }

    neutron{
        alphan_medium=CASE
        alphan_bins=200    % use 200 bins for alpha slowing down calculation
        alphan_cutoff=0.0  % cutoff for alpha,n calculation
        alphan_step=LAST   % step index in this case
    }
}

```

```

    gamma{
        sublib=ALL      % LT, FP, or AC single sublib
                        % ALL = all sub-libraries
        continuum=yes   % expand continuum data stored as
                        %   lines into proper continua
        immediate=yes   % load lines for immediate gamma/x-rays
        spont=yes       % include photons from spont fiss
        brem_medium=uo2 % include bremsstrahlung from betas
    }

    save{ file="Cl_19.f71" steps=all }
}

end

=opus
title="Cl NU sf"
data="Cl_NU.f71"
typarams=sfspec
units=particles
end

=opus
title="Cl NU (alpha, n)"
data="Cl_NU.f71"
typarams=anspec
units=particles
end

=opus
title="Cl NU total n"
data="Cl_NU.f71"
typarams=nspectrum
units=particles
end

=opus
title="Cl NU photons"
data="Cl_NU.f71"
typarams=gspectrum
units=particles
end

=opus
title="Cl 2wt% sf"
data="Cl_2.f71"
typarams=sfspec
units=particles
end

```

```

=opus
title="Cl 2 wt% (alpha,n)"
data="Cl_2.f71"
typarams=anspec
units=particles
end

```

```

=opus
title="Cl 2wt% total n"
data="Cl_2.f71"
typarams=nspectrum
units=particles
end

```

```

=opus
title="Cl 2wt% photons"
data="Cl_2.f71"
typarams=gspectrum
units=particles
end

```

```

=opus
title="Cl 5wt% sf"
data="Cl_5.f71"
typarams=sfspec
units=particles
end

```

```

=opus
title="Cl 5wt% (alpha,n)"
data="Cl_5.f71"
typarams=anspec
units=particles
end

```

```

=opus
title="Cl 5wt% total n"
data="Cl_5.f71"
typarams=nspectrum
units=particles
end

```

```

=opus
title="Cl 5wt% photons"
data="Cl_5.f71"
typarams=gspectrum
units=particles
end

```

```

=opus
title="Cl 19.75wt% sf"

```

```

data="Cl_19.f71"
typarams=sfspec
units=particles
end

=opus
title="Cl 19.75wt% (alpha,n)"
data="Cl_19.f71"
typarams=anspec
units=particles
end

=opus
title="Cl 19.75wt% total n"
data="Cl_19.f71"
typarams=nspectrum
units=particles
end

=opus
title="Cl 19.75wt% photons"
data="Cl_19.f71"
typarams=gspectrum
units=particles
end

```

# Vita

Karen Koop Hogue is an R&D staff member at Oak Ridge National Laboratory in Oak Ridge, Tennessee in the Nuclear Nonproliferation Division. Karen received her BS degree in nuclear engineering from Texas A&M University prior to being commissioned as a US Naval Officer and teaching at Naval Nuclear Power School. Karen also received her MA degree in Nonproliferation and Terrorism Studies from the Middlebury Institute of International Studies at Monterey and has worked previously at Lawrence Livermore National Laboratory and Y-12 National Security Complex.