

Semi-Empirical Modeling and Implementation of Hybrid K-Edge Densitometry for Pyroprocessing Applications

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Degree

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I can do all things through Christ which strengtheneth me.

Philippians 4:13

Abstract

The United States is currently looking at options for handling of spent nuclear fuel. Currently, there are $\approx 70,000$ metric tons of spent fuel in storage in the US alone. Pyroprocessing is a possible method for spent nuclear fuel reprocessing which was proven to work at Argonne National Laboratory. This masters thesis showcases a method for empirically modeling hybrid k-edge densitometry, one of the numerous possible safeguards needed for a reprocessing facility. This is accomplished by using MCNP to perform 54 sets of 2-stage simulations for KED and XRF, respectively. The end results are empirical linear functions for the magnitude of the k-edge drop of uranium and plutonium, as well as empirical functions for the XRF peaks for uranium and plutonium. These two empirical functions are functions of uranium concentration and plutonium to uranium mass fraction ratio, respectively.

The semi-empirical functions are then implemented into the Sandia National Laboratory Separation and Safeguards Performance Model EChem (SSPM Echem) Simulink model. The SSPM is a Simulink model which can use modular safeguards functions for methods such as HKED, passive gamma, and passive neutron detection. These types of safeguards modules help to evaluate how much the standard error of inventory difference (SEID) is affected by additional safeguards, providing a quantifiable value. The empirical functions as well as multiple representative figures and tables are presented, showcasing the ability of the Simulink module to correctly predict the KED drops as well as the XRF peaks. The ultimate goal is to combine the KED and XRF into HKED measurements to get a value for the mass fraction of plutonium in salt. Plots showcasing the differences between the HKED module output and SSPM's internal mass tracking are shown.

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Chapter 1

Introduction

The United States currently has approximately 70000 MTU of spent nuclear fuel (SNF) sitting either in intermediate length storage via spent fuel casks (heavily shielded concrete containers), or in spent fuel pools [6]. Most reactor sites are at or near their limit for pool storage, and above ground cask storage is becoming commonplace. The United States federal government has responsibility for commercially produced spent nuclear fuel, per 10 CFR 961.11 [7].

As such, the United States is currently investigating a number of possible solutions for spent nuclear fuel. The most widely known route is via direct disposal at a facility such as Yucca Mountain. This route would involve simply taking the spent fuel at reactor sites, repackaging it, and then disposing of it in a geologic repository. Some consider this route to be wasteful, not utilizing the full potential of nuclear fuel. The reason for this is that spent fuel is composed of $\approx 96\%$ uranium [8], and reprocessing the SNF would offer multiple benefits.

Reprocessing is the method by which spent nuclear fuel is recycled, typically by either aqueous or electrochemical means. This is already being done in multiple countries, and was originally developed in the US for defense purposes. One of the key international issues with reprocessing is the handling of plutonium and enriched uranium, the latter of which coming from breeder and/or research reactors, in the spent fuel. For material accountancy and safeguards purposes, being able to reliably track and quantify plutonium masses throughout a reprocessing facility is crucial.

To this goal, one of the in-situ safeguards techniques, hybrid k-edge densitometry, is being looked at. HKED is already in use at aqueous based reprocessing facilities such as Rokkasho-mura Reprocessing Plant in Japan [9]. Some safeguards methods such as passive gamma or neutron can provide information on isotopes such as ^{137}Cs , but do not provide the plutonium mass fraction in the salt. HKED offers an opportunity to determine the absolute density of plutonium in the salt with a non-intrusive measurement.

This thesis attempts to use a pre-existing MCNP model for HKED to develop empirical functions for use in the Safeguards System Performance Model, covering a wide range of PWR fuel and burnup conditions. SSPM is a Simulink based flowsheet model, keeping track of mass flows throughout a pyroprocessing facility. The model has the ability to be augmented with additional Simulink modules. SSPM is described in more detail in chapter 2.

HKED is potentially very beneficial for pyroprocessing, because pyroprocessing involves a much harsher environment, where the end products are uranium ingots, uranium/transuranic mixed ingots, and ceramic waste. There are not many good ways to determine plutonium content while a pyroprocessing facility is online. HKED provides one option for this. Pyroprocessing involves very high temperatures, and does not have the exact same safeguards environment that aqueous based reprocessing does. In aqueous reprocessing, plutonium is typically separated from the uranium, and can be accounted for on its own, using methods as simple as mass balances. In pyroprocessing, the fact that the plutonium is contained within the U/TRU ingot means that it would be beneficial to verify that no plutonium is missing outside of tolerances before this stage. In addition to this, there is residual plutonium in the salt used in pyroprocessing, making mass balances for plutonium very difficult without frequent facility clean-outs. This would add considerable cost and inefficiency to pyroprocessing.

For the sake of nomenclature, when the term mass fraction is used in this paper, it is referring to the absolute mass of an element with respect to the total mass of the fuel-salt mixture.

1.0.1 Motivation

The motivation from this project stems from the fact that pyroprocessing presents potentially different safeguards possibilities at different locations than aqueous reprocessing does. As such, it was determined that UT Knoxville would develop a suite of Simulink modules for the SSPM. These modules would represent different safeguards techniques, and try to simulate outputs of the techniques in a novel approach to pyroprocessing safeguards. These techniques range from gamma and neutron passive emission, to HKED. The goal is to eventually use the results of the different modules together to keep account of the inventory difference (ID). By using multiple modules where they make sense to use, this provides a more robust method of material accountability and safeguards. The goal is to combine the outputs of these modules to lower the overall uncertainty of items such as the ID.

As part of a literature review process, it was determined that passive gamma and neutron would be easy to implement, the (α, n) portion of passive neutron would require some work but was doable, and HKED would require some work, but would be possible to implement into SSPM. As such, it was determined that Cooper would be in charge of developing a semi-empirical set of relations for HKED, using the previously built HKED MCNP6 model from Matt Cook. This model is described in detail in chapter 3. The key measurement that HKED provides is the absolute density of plutonium in salt, or using MCNP terms, the mass fraction of plutonium in salt.

From a pyroprocessing perspective, the most sensible place to locate a module for HKED would be the electrorefiner, where there is a fuel-salt mixture. Locations such as the ingot storage or fuel-chopping do not make sense as HKED relies on shining x-rays through a sample solution.

1.1 Reprocessing

The first benefit of reprocessing is utilization of more of the potential energy of the nuclear fuel. Reprocessing the fuel allows the majority of the uranium to be used as fuel a second time, if also using fast spectrum reactors. A second benefit is the reduced amount of SNF, both in terms of total mass, and potentially, heat and volume, that would be going to a

repository. The potentially statement is due to several factors including if fast reactors are used with the assumption of full recovery of actinides [10]. The majority of the highly radioactive isotopes will have half lives of less than 50 years, which implies that partitioning the waste could be beneficial.

1.1.1 PUREX

The United States has been reprocessing SNF since the Manhattan project, although not for the purpose of recycling commercial fuel since the 1978 decision by former president Carter. The United States has used the Plutonium Uranium Solvent Extraction (PUREX) process for decades to recover plutonium from specially burned nuclear fuel for the purpose of harvesting nuclear weapons material. This process involves multiple steps of reducing and oxidizing the uranium and plutonium, to produce an immiscible mixture, whereby uranium and plutonium are selectively extracted using compounds such as tri-*n*-butyl-phosphate (TBP), which facilitates solvent extraction by forming an organic compound with uranium and plutonium. This process is well-understood, and there is significant experience with it. However, there are drawbacks. A significant amount of byproduct nuclear waste is produced via this method, and it can not be used on very fresh SNF, as there are radiation limits to this technique. Specifically, TBP can suffer from radiolysis in high radiation fields. It is also not suitable to sodium bonded fuel [10], which is illustrated by the EBR-II fast reactor at INL. The last drawback is the large footprint required for a PUREX facility. This is illustrated by the Savannah River and La Hague sites. Savannah River is a aqueous facility in the US that was used for reprocessing nuclear fuel for defense purposes. The facility is 800 km², and has more than 10000 employees. La Hague has a listed capacity of 1700 metric tons of spent fuel per year. Both of these sites are large scale, with La Hague being capable of reprocessing roughly half of the spent fuel that the US generates per year, for reference purposes. Pyroprocessing facilities can be built at a smaller scale, such as Argonne National Laboratory's proposed 100 metric ton per year facility [10].

1.1.2 Pyroprocessing

An alternative method of reprocessing that has been proven to work is electrochemical reprocessing, also known as pyroprocessing. This method utilizes the fact that the Gibbs Free Energy of chloride formation of uranium, plutonium, and other transuranics have slight differences, and as such can be separated via adjusting the voltage going through a molten salt mixture. The method of pyroprocessing is described as follows.

The fuel is first received into the facility and chopped mechanically. Oxide fuels are then optionally sent for voloxidation to increase processing speed. The fuel is then sent to an electroreducer to be converted to metallic form. Afterwards, the electrorefiner separates the spent fuel according to differences in Gibbs Free Energy. From there, the uranium and U/TRU metals are sent for ingot processing. The salt is recycled and cleaned, with fission products being encapsulated in glass. This process is described in detail in the following subsections, and shown in fig [1.1](#).

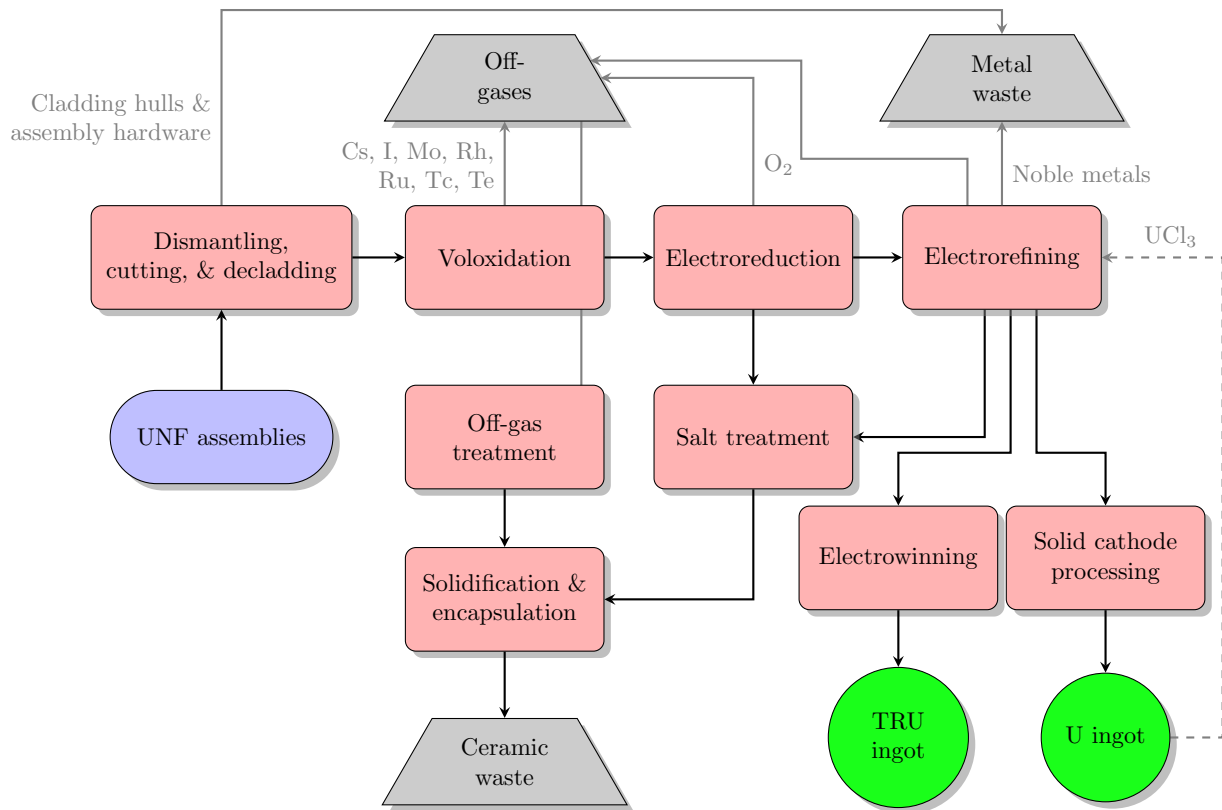


Figure 1.1: This flow sheet illustrates the pathways of pyroprocessing. The uranium is extracted along with transuranics in the electrorefiner, and the remaining waste is processed into ceramic waste. Salt is recycled prior to this. [2]

1.1.3 Fuel Rod Processing

As Figure 1.1 shows, the process starts with the spent fuel rods being extracted and then being chopped. The fuel pellets are then extracted, and oxide fuels are then optionally sent to the voloxidizer. Metallic form fuel bypasses the voloxidizer entirely. The zirconium fuel rod cladding is sent for metal waste disposal. This part is completely mechanical. The fuel pellets will then need to be oxidized via voloxidation [11].

1.1.4 Voloxidation

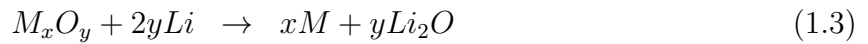
Voloxidation stands for volumetric oxidation, and it serves two purposes. The first is to release and capture some of the fission gasses, and the second is to prepare the oxide fuel for electroreduction. Voloxidation is the process of controlled addition of oxygen and heat to produce a controlled oxidation reaction within the fuel pellets, and is performed by flowing oxygen through a voloxidation chamber, producing the following reaction.



The result of this is that the fuel material is converted to a less dense, higher surface area powder to be sent to electroreduction. This allows for a faster rate of the spent fuel conversion to metallic form, similar to faster dissolution in aqueous methods [12]. Several items of note for this process are the air flow rate and the temperature of the system. The oxygen flow rate should be such that it produces the oxidizing reaction without being so high as to carry off fuel particles into the off gas stream. The temperature must be maintained above 500 °C and below 1000 °C. Temperatures above 1000 °C can cause undesirable sintering of particles. An interesting item to note, is that the voloxidation process results in elimination of a fraction of the fission products. It eliminates the majority of the technetium and ruthenium, while eliminating between $\frac{1}{5}$ to $\frac{1}{3}$ of the cesium [11].

1.1.5 Electroreduction

After the material has been pulverized and converted to U_3O_8 , the material must then be electroreduced, to yield pure metallic material, rather than an oxide. The electrolyte used is $LiCl - Li_2O$ at 650 °C, with a platinum rod anode. The cathode of this cell is the fuel powder, contained within magnesia membrane walls, so ions can traverse the membrane to react. The reaction that takes place is shown as Equations (1.2) and (1.3) [11].



Some of the fission products such as cesium and barium (alkaline elements) will dissolve into the salt as metal chlorides, as it is electrochemically favorable for them to do so. These fission products can be disposed of when the salt is treated. The electroreduction process is extremely effective, reducing uranium and plutonium to metals at rates of ≈ 99 and 96 %, respectively. The salt then has to be separated from the transuranic metals. This is done by evaporation at temperatures of around 1200 °C. During this process, some actinides can be contained in the waste streams of fission products and salt [11].

1.1.6 Electrorefiner

The spent fuel is then inserted into an anodic basket to be placed into the electrorefiner. At the electrorefiner, the fuel is dissolved into the molten salt with an applied voltage. The voltage applied will pull off components of the fuel in a particular order, corresponding to the Gibbs Free Energy. The lathanides will dissolve into the salt first, then the uranium and transuranics will go into the salt. The noble metals (iron, zirconium, cadmium, etc) will stay in the chopped fuel basket, as they have higher Gibbs Free Energy, are more resistant to forming chloride species. After uranium and transuranics are dissolved into the salt, the voltage is then lowered, allowing the reaction to proceed in reverse. The uranium will plate out first as dendrites on the solid cathode. After uranium collection on the solid cathode has saturated, the voltage is lowered again, this time pulling out the transuranics, along

with some of the residual uranium, into a liquid cadmium cathode. After these two steps, the cathodes are sent off to collect the uranium and transuranics, eventually producing solid metal ingots [11, 10].

The salt will over time accumulate fission products and trace amounts of uranium and transuranics. This is a key difference between aqueous and pyroprocessing methods, and produces a significant difference in applicable safeguards. For aqueous based reprocessing, it can safely be assumed that when flushing material from one place to another, material accountancy can be strong, and plutonium can be tracked this way. Because pyroprocessing is a batch based process with plutonium being accumulated in the salt over time, this implies that a standard mass balance of plutonium input versus ingot mass would not work. Some plutonium may be contained within the salt of the electrorefiner as an example. In reality it would be exceptionally hard without a complete system clean to determine the plutonium content via mass balance alone. This highlights the need for HKED and other measurement modules.

Every so often, the salt must be purified, with the fission products being captured in zeolite for storage/disposal. Throughout this process, whenever possible, off-gasses (such as tritium and xenon) are captured for disposal [11].

1.2 Hybrid K-Edge Densitometry

1.2.1 Description of HKED

Hybrid K-Edge densitometry is a nuclear safeguards technique that is based on two separate techniques k-edge densitometry (KED) and x-ray fluorescence (XRF), and combines them together into one measurement device. This technique has been proven to work for aqueous based reprocessing, and is scheduled to be used in the Rokkasho reprocessing facility in Japan [13].

K-edge densitometry works by using the fact that the mass attenuation coefficient for gamma rays encounters a significant increase around electron shell binding energies. This is because when a gamma ray is above a shell closure energy, it is possible for that gamma to

eject an electron. This extra reaction channel increases the total cross section. The K-shell is of particular interest for several reasons. The first is that it is located at 115.6 keV for uranium, and is very noticeable. The second is that the mass attenuation coefficient for the K-edge is a very sharp, distinct rise and fall, compared to for example the L-edge, which consists of a doublet state. These two factors combine to make for a very clean and easy measurement.

The K-edge drop occurs when a gamma ray passes through a material, such as molten salt, and encounters uranium. To perform this measurement, a gamma ray source is shown at a molten salt sample, with a detector behind the sample. As uranium concentration in the salt is increased, a noticeable decrease in gamma ray intensity occurs around 115.6 keV in the detector pulse height spectrum. This is due to the uranium mass attenuation coefficient at 115.6 keV being significantly higher than the nearby energies, and preferentially blocking transmission of gammas at that energy. This is shown in [1.2](#). Plutonium's K-edge is located at 121.8 keV [[14](#)]. However, for pressurized water reactor fuel, plutonium makes up a small mass fraction of the fuel, and the plutonium k-edge is barely visible.

X-ray fluorescence occurs as a result of the ejection of K-shell electrons due to photon interaction. This electron ejection causes a higher orbit electron to fall and take the vacant spot, emitting a characteristic x-ray with an energy representing the difference between the two shell binding energies. These binding energies are unique to each element. A list of common x-rays emitted from uranium and plutonium k-shell ejections is listed in [1.1](#) [[5](#)].

XRF works by relating the concentration of uranium to some other actinide, typically plutonium. The intensity of x-ray peaks from actinides is proportional to their mass in concentration. As such, if the uranium and plutonium x-ray intensities are known, it is possible to express the uranium to plutonium mass as a ratio. If the uranium to plutonium mass fraction ratio is known from XRF, and the uranium mass concentration is known from KED, then it is possible to combine the two measurements to determine the absolute density of plutonium in the salt.

Uranium

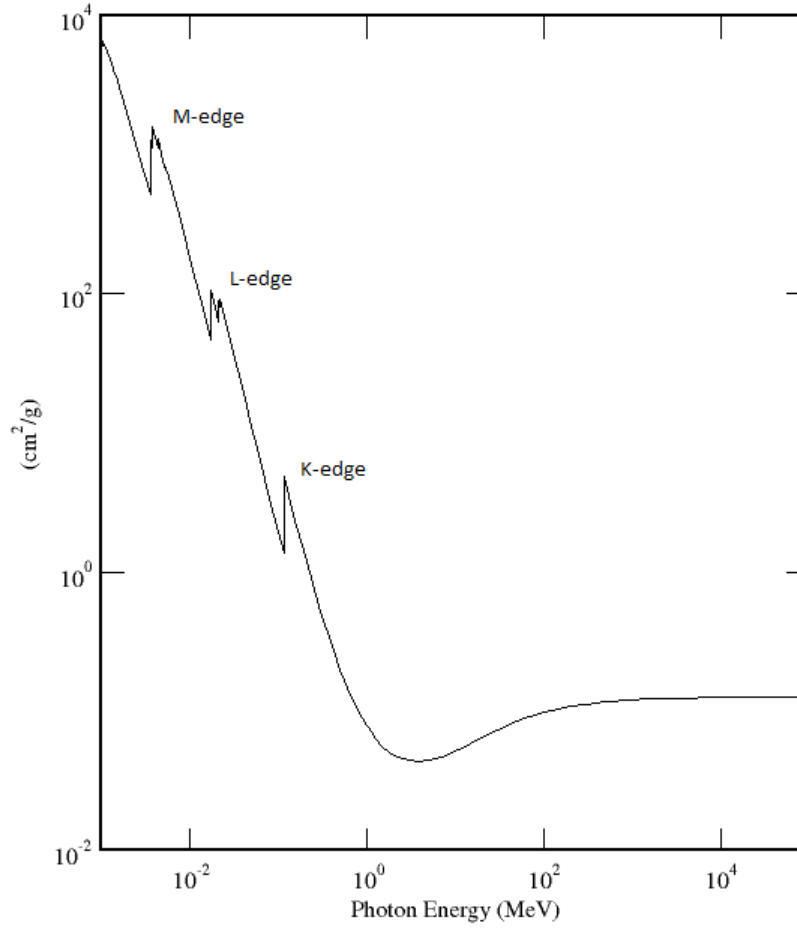


Figure 1.2: Mass Attenuation Coefficient for Uranium [3]

Table 1.1: X-ray Energies Emitted By Uranium and Plutonium K-shell Ejections (keV) [1]

Element	$K_{\alpha 1}$	$K_{\alpha 2}$	$K_{\beta 1}$	$K_{\beta 2}$	$K_{\beta 3}$	$K_{\beta 4}$
Uranium	98.43	94.65	111.3	114.5	110.4	114.8
Plutonium	103.7	99.52	117.2	120.6	116.2	120.9

Chapter 2

SSPM Description

2.1 Goals of SSPM

A Simulink-based model of the PUREX flowsheet (SSPM: Safeguards System Performance Model) was originally developed by SNL for the modeling of safeguards for the PUREX process. To facilitate the modeling and simulation of a pyroprocessing facility, a Simulink model named the Separation and Safeguards Performance Model Echem (SSPM-Echem) was developed from the previous SSPM. This model follows in the footsteps of the original PUREX SSPM, but with key differences due to the nature of pyroprocessing. Several of these differences are the fact that aqueous uses and accountability tank, whereas pyroprocessing does not. Pyroprocessing typically does not have routine frequent flushing of the plant, and as such actinides can build up in the salt [4].

Some of the guidance for developing safeguards for a reprocessing plant can be gleaned from the safeguards principles being implemented by the Rokkasho plant. Some of the guidelines for the safeguards are as follows [15].

1. Defined Material Balance Areas (MBAs) for nuclear material accounting
2. Key Measurement Points (KMPs) for measuring flow and inventory of material
3. Defined strategic points for containment, surveillance, and verification measures

4. Statistical evaluation of the material balance to determine Material Unaccounted For (MUF) or Inventory Difference (ID)

2.2 SSPM Layout

SSPM models safeguards performance in a hypothetical pyroprocessing facility by using Simulink blocks in a batch based (as opposed to continuous) flow. The flow of material throughout the facility is tracked with a vector of 99 elements which correspond to the first 99 atomic elements. The mass of material moving through various stages such as electroreduction or electrorefining is handled using separation fractions, rather than actual electrochemistry. The yearly throughput of the facility being modeled would be ≈ 100 MTU per year. This corresponds to roughly 1 fuel assembly per day [4].

Several assumptions are made in the model. Of these, one is the U product being 100% U, and containing no impurities. The second assumption is that the U/TRU mixed product is composed of 70% U, and 30% TRU. These values can be changed, but were not for this project [4].

One of the concepts of the SSPM is to have 3 material balance accountancy areas, and one process monitoring section. The mass flow starts with first MBA, with fuel being stored to be chopped, and then voloxidized, the same as the process described in [chapter 1](#). The next MBA handles the electroreduction and electrorefining, followed by drawdown and ingot formation. Afterwards, the salt is recycled back to the electroreducer and electrorefiner. The third MBA handles keeping inventory of the mass flows.

A figure illustrating the layout of the SSPM model is shown in [2.1](#). While the results of this masters project were implemented into SSPM, the actual workings of SSPM are not the topic of this thesis. The modeling strategy for safeguards modules, in particular HKED, will be discussed however.

[illegible]

Figure 2.1: Illustration of SSPM-ECHEM flowsheet [4]

2.3 Modeling Strategy

One of the goals for the project is to develop safeguards modules for SSPM. An advantage of SSPM is that as a Simulink model that tracks mass flows, it can be outfitted with modular safeguards functions with ease. The first thing that was done on the project was evaluating the safeguards options that would be available to model. Many different safeguards methods were evaluated, and several were selected based on feasibility with respect to the information being tracked in SSPM. These include but are not limited to options such as passive gamma, passive neutron (including (α, n) and spontaneous fission), and HKED signatures. Once a functionalized safeguards method has been developed, it can then be implented into SSPM via function blocks. As such, the main work of this thesis is the parameterization of the HKED model developed by Cook, and the implementation of the semi-empirical model into the SSPM-Echem.

The bigger goal of the overall project with regards to the SSPM is to provide multiple modules to simulate measurements, while combining the results of these modules to attempt to provide lower overall uncertainty. This in theory provides higher confidence regarding key safeguards checks such as inventory difference of plutonium, and confidence that the facility is operating in a normal safe manner.

Chapter 3

Methods

This chapter will cover the approach taken to implement HKED safeguards into the SSPM. An introduction to the ORIGEN modeling of spent fuel will be the first step, then an introduction to the MCNP model will be the second topic. This will be followed by methods used to decrease runtime and automate generation of the MCNP simulations.

3.1 Used Fuel Source Terms

Spent nuclear fuel is primarily composed of very lightly enriched uranium, with around 5% fission products, and 1-2% plutonium. However, burnup, length of operating cycles, downtimes between cycles, and original fuel enrichment, among other factors, can cause the isotopics of spent fuel to vary from one discharge to another. This implies that using a single spent fuel composition for HKED modeling could be misrepresentative, and lead to incorrect XRF peak ratios, for example. To rectify this, the first part of this project was to identify common spent fuel enrichments and burnups. This was based off of a Los Alamos National Laboratory empirical relationship relating burnup to original enrichment for commercial PWR fuel [16].

It is beneficial to describe the ORNL SCALE code before continuing. SCALE is, among other things, a reactor physics code which includes multiple sub-packages. One of these packages is the Oak Ridge Isotope Generation (ORIGEN) code. This code solves the Bateman equations to handle depletion cases for nuclear fuel. This code allows researchers

to predict discharge isotopic compositions based off of reactor power, cycle length, and downtimes.

The range of burnups was restricted to the low point of 20 GWd/MTU, and high point of 60 GWd/MTU. This were chosen based off of typical US spent fuel composition, assuming an average burnup of 45 GWd/MTU. Fuel rods on the edges of reactors will not have as high of burnup due to reactor operators optimizing the reactor neutronics, and could have values significantly lower than the average 45 GWd/MTU. Values below 20 GWd/MTU are typically reserved for special cases such as research reactors, failed fuel assemblies, or weapons material generation. This can be shown with trivial depletion calculations. Values above 60 GWd/MTU typically don't occur in the US due to the fuel generating neutron poisons over time and depleting the U-235 in the fuel. Another reason for the majority of SNF being under 60 GWd/MTU is that the NRC licenses fuel to a maximum region-average and batch-average discharge burnup [17].

With this in mind, the enrichment and burnup values chosen for the spent fuel source terms are listed in Table 3.1.

Once the space of enrichment and burnup values was determined, a Python script was written by Jonathan Mitchell and Steven Skutnik to automate production of ORIGAMI (an alternative input format to ORIGEN) input decks. A change that was made to this script by Michael Cooper was to switch the reactor cycle lengths and number of cycles from the original 12 months and 5 cycles, to the values of 18 months and 3 cycles. This change was based on the fact that the United States reactors typically operate on 18 month cycles [18]. The Python ORIGAMI generation script was then used to generate the ORIGEN input to use in the SCALE [19].

Now that the source term generation has been described, a discussion of the HKED MCNP model is next.

Table 3.1: Spent Fuel Compositions For Source Terms

Burnup $\frac{GWd}{MTU}$	Enrichment	Cooling Time (Years)
20	1.96%	5, 15
20	2.17%	5, 15
20	2.39%	5, 15
25	2.26%	5, 15
25	2.51%	5, 15
25	2.76%	5, 15
30	2.55%	5, 15
30	2.83%	5, 15
30	3.11%	5, 15
35	2.81%	5, 15
35	3.13%	5, 15
35	3.44%	5, 15
40	3.07%	5, 15
40	3.41%	5, 15
40	3.75%	5, 15
45	3.31%	5, 15
45	3.68%	5, 15
45	4.05%	5, 15
50	3.55%	5, 15
50	3.94%	5, 15
50	4.34%	5, 15
55	3.77%	5, 15
55	4.19%	5, 15
55	4.61%	5, 15
60	3.99%	5, 15
Continued on next page		

Table 3.1 – continued from previous page

Burnup $\frac{GWd}{MTU}$	Enrichment	Cooling Time (Years)
60	4.44%	5, 15
60	4.88%	5, 15

3.2 MCNP Modeling of HKED

3.2.1 MCNP Model

A MCNP model was constructed by former UT PhD student Matt Cook as part of his dissertation project [5]. This model contains the geometries needed to properly describe a HKED measurement apparatus such as the one shown in Figure 3.1. This MCNP model works by performing two-stage simulations for KED and XRF scenarios separately. Since the measurements are not interdependent, this allows for the KED and XRF to be handled separately for simulation, then the results can later be corroborated. This aids in the ease of building and troubleshooting the model. The KED and XRF measurements use separate beamlines to detectors.

The model is a two stage model, with the first stage employing heavy variance reduction, and the second stage being analogue transport. The second stage of the model does not employ variance reduction, and is solely photon transport from the end of the beamline to the detector, to generate a F8 pulse height tally. As mentioned above, since the KED and XRF measurements do not have to be ran simultaneously, the model is thus split into a KED version and XRF version, for simplification, troubleshooting, and runtime. Another reason would be the way that MCNP handles DXTRAN spheres. DXTRAN is the MCNP terminology for deterministically trasnsporting particles from one area to another. It is typically used in scenarios where Monte Carlo transport would lead to extremely few particles reaching the target area. An example of this would be a sharp corner of a hallway, or thick shielding. This method of variance reduction can lead to unreliable answers at worst when

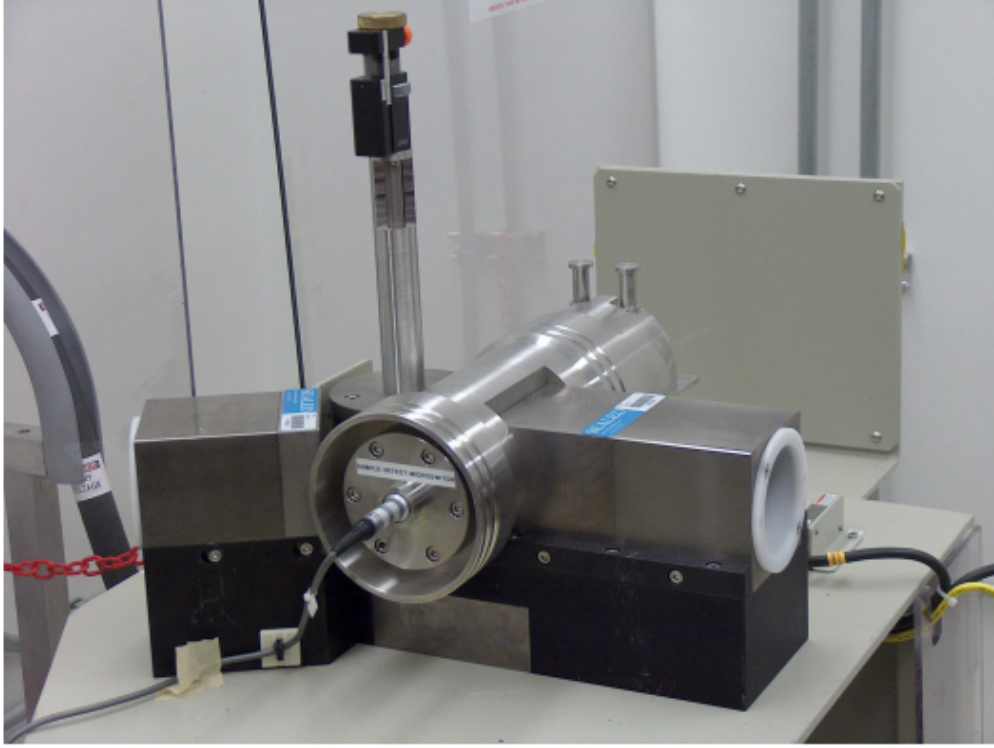


Figure 3.1: HKED system without outer lead shielding. Both XRF (left) and KED (right) component beamlines are exposed and germanium detectors have been removed [5]

multiple spheres are used. Extreme care must be taken when using multiple DXTRAN spheres, so that ordering of the spheres is correct.

The model operates on several principles. The stage-1 simulations are used to generate gamma energy distributions exiting the beamline, which are to be used for the source file for the stage-2 simulations. The stage-2 simulations determine the pulse height response using F8 tallies. This effectively means that the stage-1 simulations guide the stage-2 simulations by building a more focused gamma source term. The gamma source exiting the beamline, which is going from the salt sample to the detector, has very few particles reaching the detector due to the narrowness of the beamline[5].

The overall variance reduction techniques employed in the XRF model were as follows [5, 20]:

1. **Forced collisions:** All particles entering the sample are required to interact and weighted appropriately. This is done to maximize the number of reactions taking place

in the model. Increasing the number of reactions results in better statistical certainty for a set number of particles.

2. **MCNP DXTRAN Sphere:** Particles are forced to travel to the sphere volume, and are weighted appropriately. This method uses deterministic transport to transport particles to the desired location. It is typically used in either very tight geometry such as narrow beamlines or sharp corners, or in thick shielding scenarios. For the HKED model, a DXTRAN sphere is used to deterministically guide the particles to the edge of the beamline, improving statistics.
3. **Spatial weight window mesh:** Particles are weighted and controlled via splitting and Russian roulette based on position. This is done to increase the total number of particles in a particular area, resulting in better statistics. This is tied to the energy weight windows in MCNP.
4. **Energy weight window mesh:** Particles are weighted and controlled via splitting and Russian roulette based on energy. This is done to increase the number of particles within a particular energy range, resulting in better statistics for a target energy range. This is tied to the spatial weight windows in MCNP.

3.2.2 Preliminary Work Performed On Model

The first step that was done was running the KED portion of the model and checking the output with no modifications. The resulting stage-2 spectrum did not match the results published in Cook's dissertation, so the MCNP input decks and Python processing script used were reviewed. This resulted in the realization that the Cd-109 check source was not being implemented into the stage-2 source term. This Cd-109 source was being used for normalization, and as such, its absence was causing the spectrum to be incorrect. Once this was remedied, along with correcting MCNP surface references, a comparable pulse height spectra was obtained for the sample case of EBR fuel using the stage-2 simulation output.

For the XRF portion of the model, it was run as is, and the stage-2 output analyzed. As was the case with the KED portion, the unmodified input decks did not result in an

appropriate spectrum. Upon review of the XRF output, it was realized that extremely few stage-1 particles were reaching the F2 tally used to generate the gamma source term for the stage-2 simulation. The XRF input decks were then examined, where it was revealed that the DXTRAN variance reduction had been commented out. Once this part of the code was repaired, the model was run again, with the resulting spectra matching those published in Cook’s dissertation [5].

3.2.3 Generation of MCNP Outputs for Modeling

It was originally assumed that a large number of spent fuel compositions and MCNP simulations would be required to correctly empirically model the k-edge drop and XRF ratios. While this turned out to not be the case, the original implementation of MCNP output generation will now be discussed.

Originally, Tables 3.1 were used as a reference domain for generating the spent fuel mass fractions. Each of the spent fuel discharge conditions highlighted in 3.1 and ?? were simulated in ORIGEN. The resulting ORIGEN outputs were then post-processed in Python to obtain the spent fuel elemental concentrations. These concentrations were used to determine the relative amounts of uranium, neptunium, plutonium, and americium in the spent fuel. Cook’s MCNP model had assumed an actinide mass fraction in the salt of $\approx 7\%$. To modify the MCNP input decks to represent the spent fuel conditions in 3.1 and ??, the relative amounts of uranium and other actinides from the ORIGEN spent fuel source terms were then proportionally assigned to the 7% mass fraction of total actinides. Assigning the spent fuel source terms this way ensures that the optical thickness of the fuel-salt mixture is kept nearly constant. The total number of input decks was 216, composed of 27 stage-1 runs, 27 stage-2 runs, for 2 discharge times, and for KED and XRF.

Of note, for commercial spent fuel, there is an extremely small presence of any actinides other than uranium and plutonium, with uranium being vastly more abundant than plutonium. This will cause the plutonium k-edge drop to be barely visible. Due to this consideration, only uranium k-edge fitting was attempted for this project. An example KED spectra is shown in figure 3.2.

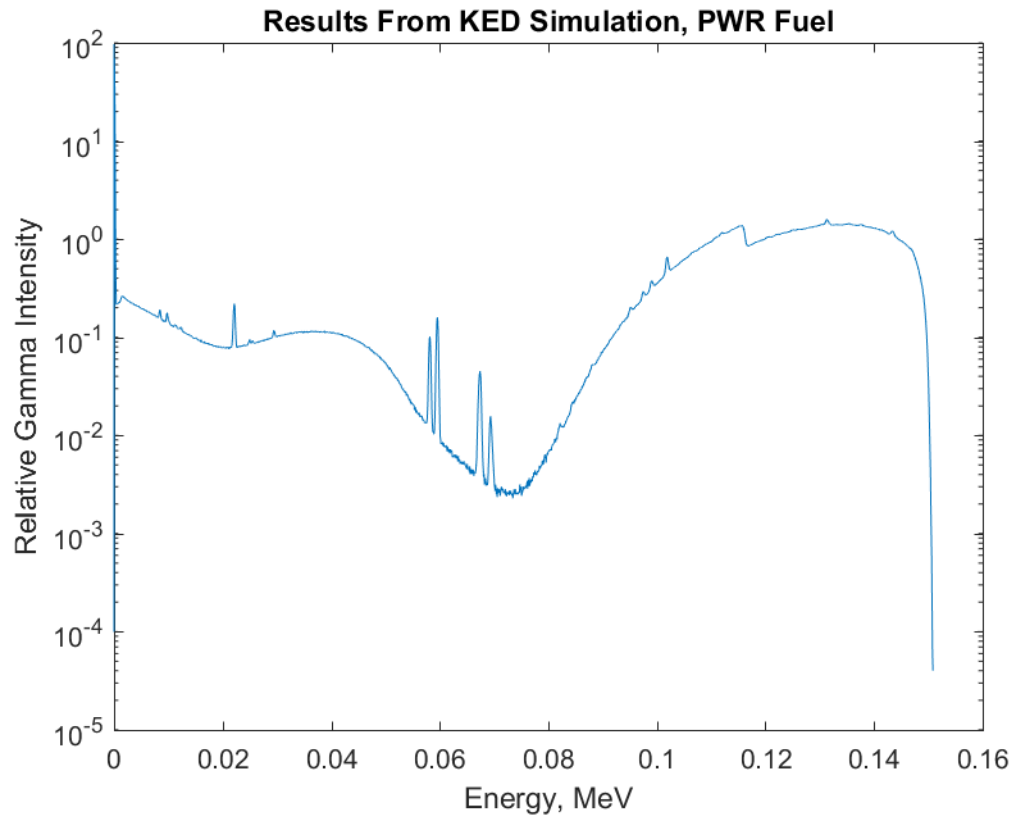


Figure 3.2: KED Spectrum for a Representative PWR Fuel Sample

Once the input decks for MCNP were created, the stage-1 and stage-2 simulations were performed sequentially. For the stage-1 simulations, 250 million particles were used per run. For the stage-2 simulations, 100 million particles were used per run. This would turn out to be a very grave mistake due to stage-1 error influencing stage-2 tallies, as will be discussed in the section “second attempt”.

3.2.4 Potential Issues With MCNP Modeling

The overall task of modeling HKED with MCNP works very well. However, there are several issues, one of which is due to the stochastic nature of Monte Carlo, and the other is due to Cook’s method of implementation.

The first issue, which is tied to the second issue, is statistical uncertainty. Monte Carlo is a stochastic process, with the uncertainty varying as $N^{-0.5}$. Therefore, in order to get low uncertainty in the F8 pulse height tallies, it is necessary to run many particles, if few are reaching the detector. The reality is that due to the geometry of HKED, and the beamline’s narrowness, an extremely small percentage of the original particles reach the detector, on the order of 1 in 10 million per energy channel of the stage 1 tallies. As such, the original method described in Chapter 3, using 250 million particles for stage-1 simulations, and 100 million for stage-2, is not what was used for the final results. This will be described in the next subsection.

For the HKED models, both KED and XRF, the uncertainty listed in the output of the S2 tally is not necessarily correct. This is because of the way the simulation works. The stage-1 simulation uses a DXTRAN sphere to guide the particles to the end of a beamline, where where particle flux is tallied via an F2 tally ($\frac{particle}{cm^2}$) to generate a source term for the stage-2 simulation. Therefore, there is intrinsic uncertainty in the stage-2 source term, resulting from the stage-1 simulation results. In a sense, since the stage-1 simulations are guiding the stage-2 simulations, if the stage-1 simulation has a large uncertainty, it can bias the source term energy distribution used by the S2 simulations. This will not be reflected in the output of the S2 F8 tally, as it is only concerned with particles traveling from the beamline to the microcell target, without any care for how many particles actually reach the beamline. As a result, even though the S2 MCNP output will list uncertainties on the order

of 1-5% for KED, and less than 1% for XRF, these values are not completely trustworthy, and they can be very misleading. One of the main underlying facts is that the stage-1 energy distribution for particles is being used for the stage-2 simulations. Biases due to high variance carry over directly to the stage-2 results.

3.2.5 Second Attempt

As was mentioned in the previous subsection, the first attempt to perform the KED modeling with the assumption of a linear fit was a failure. This stemmed from the method of implementation of the MCNP model, with its usage of a stage-1 and stage-2 simulation. For the first attempt, using 250 million particles resulted in the stage-1 tallies having very high uncertainties in the region of interest (100-125 keV). This high uncertainties were translating over to the stage-2 tallies, resulting in plots with very shaky features. Of other note is that when a linear fit was applied for the k-edge drop, the linear fit had a relatively wide spread.

The solution to the problems encountered in the first attempt was to increase the particle count in the stage-1 tallies to 10 billion. As this represents more than an order of magnitude more particles, the CPU hours runtime for these extended simulations was much higher. As such, it was decided to use 5 ultra-high quality simulations rather than the original 54 spent fuel compositions. This was decided based on several factors. Of the original 54 data points, many of them are extremely similar in composition. There is negligible difference between 5 year cooled fuel and 15 year cooled fuel for elemental uranium and plutonium concentrations, for example. The 5 new simulations would be based on spent fuel compositions spanning the range of uranium to plutonium ratios encountered in the electrorefiner of the SSPM model. This would allow each data point to be meaningful without oversaturating the domain that is desired to be simulated. The other reason is that should the hypothesis of a linear fit for KED be correct, then the linear fit to the 5 quality runs should be nearly exactly linear, and it indeed is, as shown by [3.3](#). The results of the first attempt at this project, along with the second attempt, are discussed in the results section.

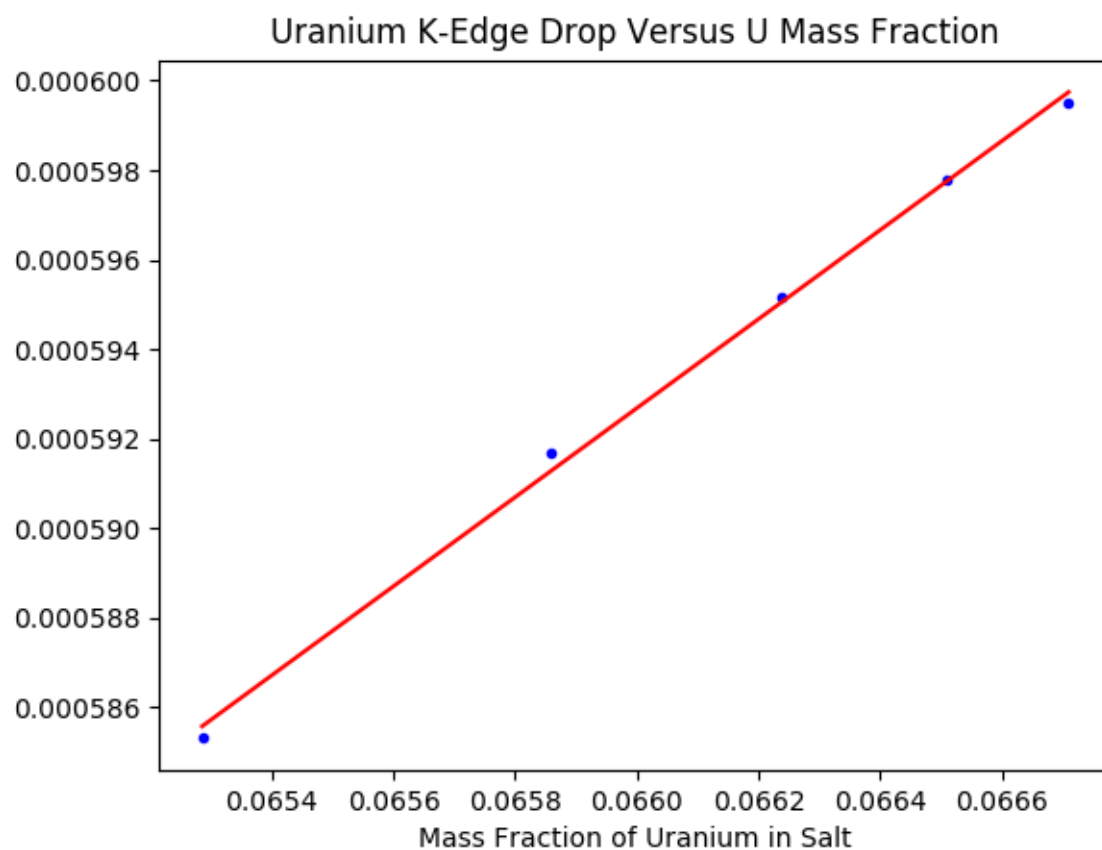


Figure 3.3: KED Fitting Using 5 Data Points. This plot spans an uranium fraction ranging from 0.0654 to 0.0666. Note error bars are too small to be visible for KED.

3.2.6 KED Fitting

After the MCNP simulations were completed, the MCNP spectral results from F8 tallies were extracted via Python scripts. These simulations were then fitted using SciPy's `curve_fit` procedure. This fits the data using according to a user supplied function, and the weights are determined by the MCNP uncertainties. The output is a linear fit of the form in (3.1). The method of KED fitting will now be discussed.

The KED region is fit before and after the K-edge drop, producing two positive slope linear fits. These linear fits are then extrapolated to where the K-edge energy is located. Then, a top and bottom value for the K-edge drop are obtained from these two fits, and this is used as the K-edge drop.

$$y = mx + b \tag{3.1}$$

For the k-edge drop, it is beneficial to observe the region being fit, so that the reader will understand the approach. This is shown in figure 3.4. The slight jaggedness of the fitting region is mostly due to stage-1 uncertainty bleeding into the stage-2 runs, as mentioned before. Increasing the particle count of the MCNP simulations greatly improved this fitting process.

The K-edge drop result for each of the KED simulations is then used in another linear fit, with this fit being the uranium mass fraction versus the magnitude of the K-edge drop. This results in a highly linear fit, which was expected since the range of uranium masses is relatively small and an exponential can be approximated as linear over a very small x range. The error propagation for the KED fitting is shown in (3.2) and (3.3).

$$\sigma_f = ((\frac{df}{dx_1} * \sigma_{x_1})^2 + (\frac{df}{dx_2} * \sigma_{x_2})^2 + \dots)^{0.5} \tag{3.2}$$

Equation (3.2) is the basic formula for propagating uncertainty in a multi-variable function. In the case of the Python Scipy fitting, both a and b are fit parameters, with uncertainty. x is assumed to be completely certain, as it is the mass fraction that is known a

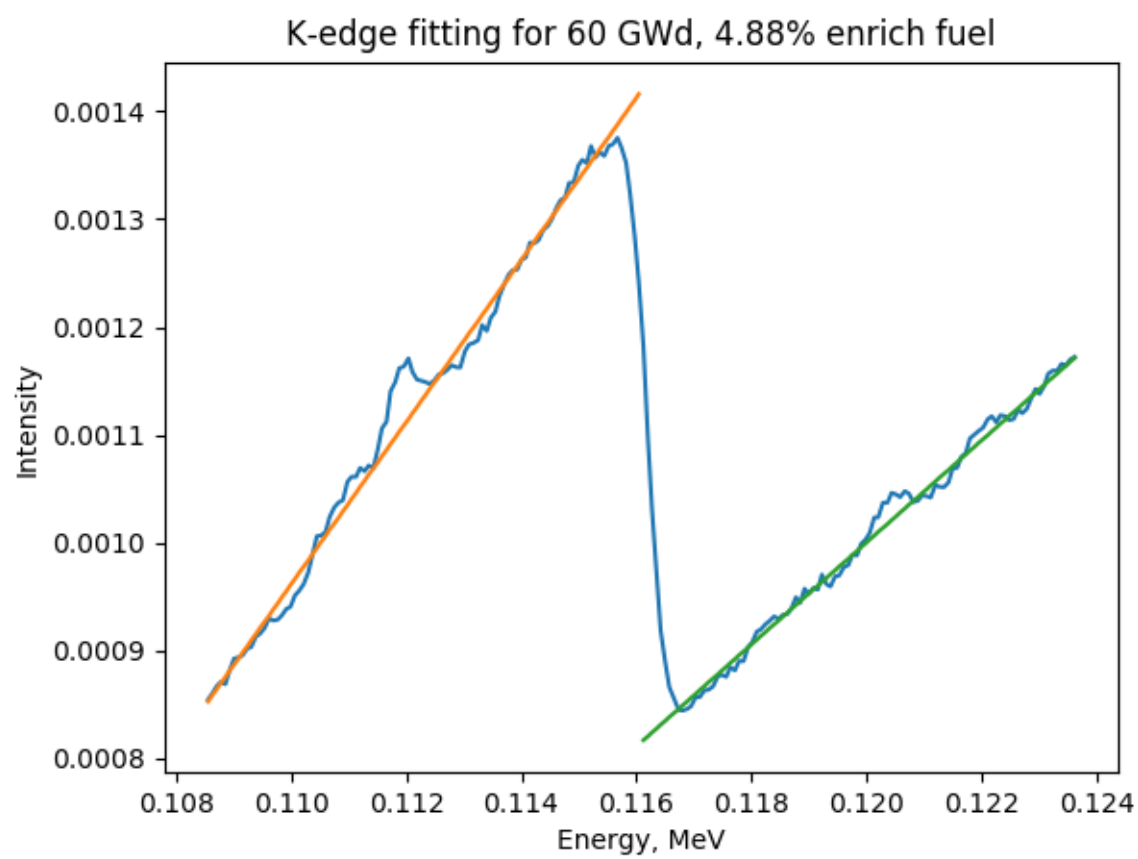


Figure 3.4: KED Fitting Region

priori from the original spent fuel compositions, and the MCNP energy bins are also assumed to be certain. This is then used for (3.3) to calculate the absolute uncertainty for each K-edge drop.

$$\begin{aligned}
y_1 &= a_1x + b_1 \\
y_2 &= a_2x + b_2 \\
\frac{dy_1}{da_1} &= x \\
\frac{dy_1}{db_1} &= 1 \\
\frac{dy_2}{da_2} &= x \\
\frac{dy_2}{db_2} &= 1 \\
\sigma_{left} &= ((\frac{dy_1}{da_1} * \sigma_{a_1})^2 + (\frac{dy_1}{db_1} * \sigma_{b_1})^2)^{0.5} \\
\sigma_{right} &= ((\frac{dy_2}{da_2} * \sigma_{a_2})^2 + (\frac{dy_2}{db_2} * \sigma_{b_2})^2)^{0.5} \\
\sigma_{drop} &= (\sigma_{left}^2 + \sigma_{right}^2)^{0.5}
\end{aligned} \tag{3.3}$$

Once the K-edge drops and their absolute uncertainties are known, these are then linearly fit with Scipy. The resulting linear fit and uncertainty are shown in the results section.

3.2.7 XRF Fitting

Although the fitting of XRF peaks with Gaussian functions was done by J. Mitchell, the correct uncertainty propagation and application to the fitting procedure was done by Cooper and will be discussed as part of this thesis as follows [21]. The only high quality XRF peaks that could be obtained from the MCNP simulations were for the k_{α_1} XRF peaks of uranium and plutonium.

Assuming the following, that the XRF ratio is the ratio of two Gaussian peaks, and that the peaks are independent of each other, it is possible to do basic uncertainty propagation to determine the uncertainty of the XRF ratio. This uncertainty can then be used for confidence

bounds in the SSPM. Equation (3.4) shows the ratio of two Gaussian peaks centered on their mean values. (3.4) is based on work by J. Mitchell [21].

$$\begin{aligned}
\gamma &= \left(\frac{A_1}{(2 * \pi * \sigma_1^2)^{0.5}} \right)^{-1} * \left(\frac{SB * A_2}{(2 * \pi * \sigma_1^2)^{0.5}} \right) \\
\gamma &= \left(\frac{SB * A_2}{A_1} \right) * \left(\frac{V_1}{V_2} \right)^{0.5} \\
\gamma &= XRF \frac{Pu}{U} \text{ peak ratio} \\
A_1, A_2 &= \text{Gaussian amplitude parameters (fitted)} \\
\sigma_1, \sigma_2 &= \text{Standard deviations of Gaussians (fitted)} \\
SB &= \text{Correction terms for peak fitting} \\
V_1, V_2 &= \sigma_1^2, \sigma_2^2
\end{aligned} \tag{3.4}$$

Using equation (3.4), the correct uncertainty propagation is as follows. A and V are fitted parameters with uncertainties that must be propagated to get the correct XRF ratio uncertainty.

$$\begin{aligned}
\gamma &= \left(\frac{SB * A_2}{A_1} \right) * \left(\frac{V_1}{V_2} \right)^{0.5} \\
\frac{d\gamma}{dA_1} &= SB * \left(\frac{-A_2}{A_1^2} \right) * \left(\frac{V_1}{V_2} \right)^{0.5} \\
\frac{d\gamma}{dA_2} &= SB * \frac{V_1^{0.5}}{A_1 * V_2^{0.5}} \\
\frac{d\gamma}{dV_1} &= SB * \left(\frac{A_2}{A_1} \right) * 0.5 * \left(\frac{1}{V_1 * V_2} \right)^{0.5} \\
\frac{d\gamma}{dV_2} &= SB * \left(\frac{A_2}{A_1} \right) * 0.5 * \left(\frac{V_1^{0.5}}{V_2^{1.5}} \right) \\
\sigma_\gamma &= \left(\sigma_{A_1}^2 * \frac{d\gamma}{dA_1}^2 + \sigma_{A_2}^2 * \frac{d\gamma}{dA_2}^2 + \sigma_{V_1}^2 * \frac{d\gamma}{dV_1}^2 + \sigma_{V_2}^2 * \frac{d\gamma}{dV_2}^2 \right)^{0.5}
\end{aligned} \tag{3.5}$$

Now that the methods for finding KED and XRF values and uncertainties has been shown, a proper display of results, and discussion of those results, can commence. The

combination of KED and XRF into HKED, as well as the propagation of uncertainty for that, will also be discussed in the results section.

Chapter 4

Results

4.1 Overview

This section will discuss the results from the semi-empirical modeling of HKED. The first results discussed will be for the KED fitting, then the XRF fitting, then the combined HKED fitting. The uncertainty propagation for combining the KED and XRF results will be discussed. The HKED combined calculation implementation into the SSPM will shown, with plutonium mass fractions compared to actual mass fractions. Machine uncertainty is propagated into the HKED combined calculation by propagating 1% machine uncertainty into each of the KED and XRF measurements.

4.2 KED Results

The semi-empirical modeling of the K-edge drop from KED compared to uranium mass fraction resulted in a very linear fit shown in equation 4.1. This fit is also shown in figure 3.3. In particular, 3.3 shows that the fit is extremely good, with all points showing agreement. There is a small negative constant offset from the zero point, which will be of note for the SSPM implementation, but is inconsequential for the theory side. The KED relative uncertainty, without machine uncertainty, is 4.485%. This is higher than experimental KED values with aqueous methods, which can be sub-1%.

The main way to reduce the model uncertainty would be to run more MCNP particles. However, this is not easy to do with current UTK computer resources, as running 10 billion particles for the stage-1 second attempt required a significant part of the UTK nuclear engineering computer cluster for a period of close to 24 hours. Since Monte Carlo uncertainty goes as $N^{-0.5}$, it would require 100 times more hours to achieve a factor of 10 decrease in uncertainty on the stage-1 results. Modifying the bin sizes for the MCNP simulations, or possibly looking at other optimizations might reduce the number of particles required, but it would still likely require extensive computer run time for the simulations.

$$\begin{aligned} y_{KED} &= 9.94477827 \cdot 10^{-3} \cdot x - 6.36704256 \cdot 10^{-5} \\ \sigma_{KED} &= 4.485\% \end{aligned} \tag{4.1}$$

The quality of the K-edge fitting can be seen from figure 4.1. This figure shows the very linear appearance of the k-edge drop over the PWR fuel domain. The dispersion of the data points from the fitted line is much less than for the first attempt at modeling, showing that the MCNP uncertainty from the stage-1 simulations was altering the particle distributions reaching the stage-2 simulations. The difference between 4.1 and 4.2 is stark. Figure 4.2 has a wide dispersion of the data points owing to bad stage-1 uncertainties and subsequent particle distributions for stage-2 simulations.

The KED uranium mass fraction calculations as well as K-edge drops calculated with SSPM will be shown in the “Application to SSPM” section.

4.3 XRF Results

The modeling of the XRF $k-\alpha_1$ ratio as a function of plutonium to uranium mass fraction also resulted in a very linear fit, albeit without having to do a second attempt with increased particles. This fit is shown in equation 4.2, and figure 4.3. Again, there is a small negative constant offset from the zero point which will be talked about in the SSPM implementation. The XRF relative uncertainty, without machine uncertainty, is 1.2261%.

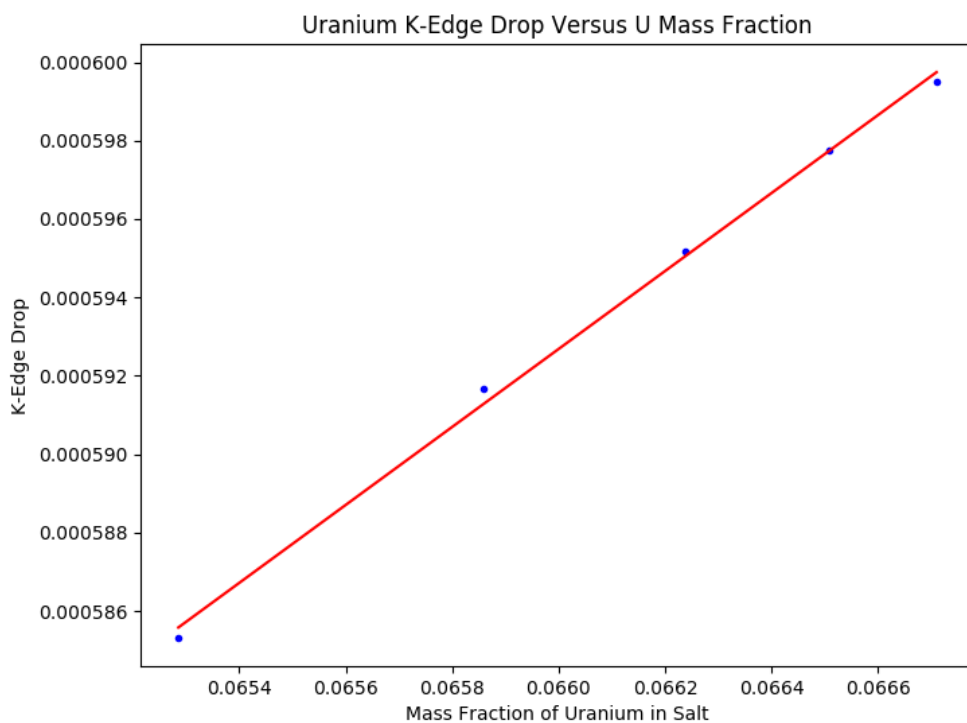


Figure 4.1: This figure shows the second attempt KED fitting. Note the narrow spread of data points from the fitted line. This plot is shown again here for convenience sake.

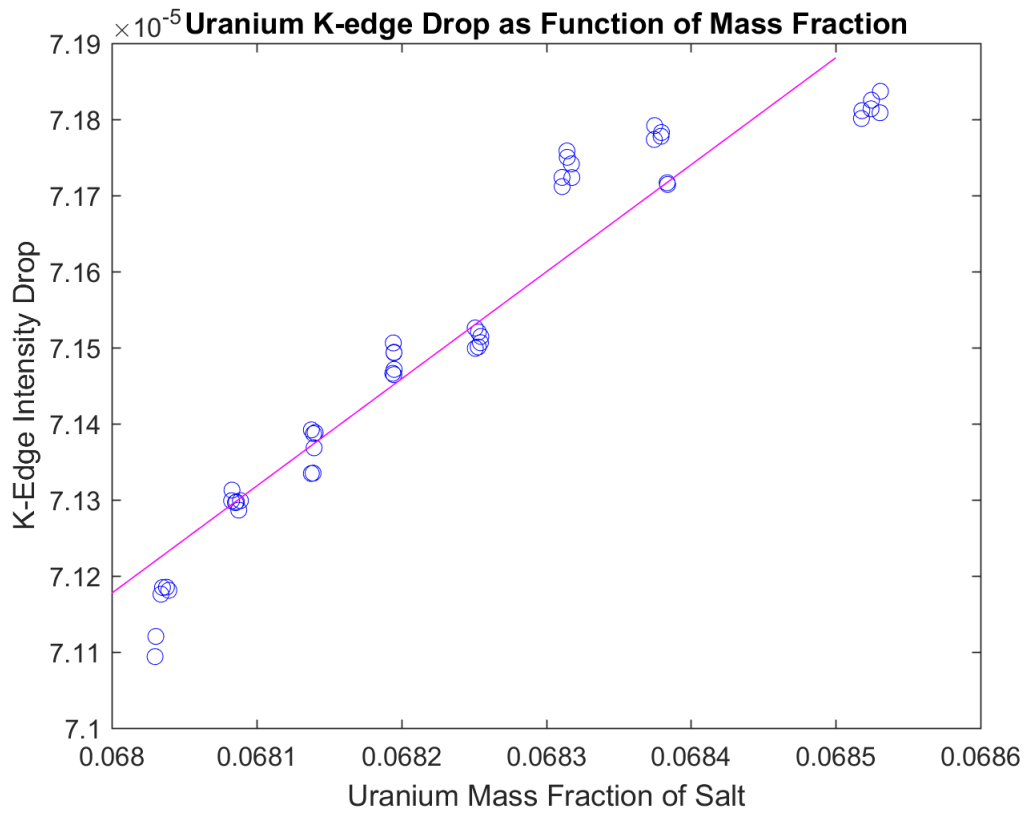


Figure 4.2: This figure shows the original KED fitting. Note the wide spread of data points from the fitted line.

$$\begin{aligned}
y_{XRF} &= 1.10472410 \cdot x - 6.36704256 \cdot 10^{-5} \\
\sigma_{XRF} &= 1.2261\%
\end{aligned}
\tag{4.2}$$

4.4 HKED Combined Calculation Results

To get the desired value which is the plutonium mass fraction, it is possible to combine the KED uranium mass fraction results with the XRF plutonium to uranium mass fraction ratio results. This is shown in equation 4.3. As shown in this thesis, it is possible to empirically relate the uranium mass fraction with the K-edge drop, and the XRF K- α_1 x-ray ratio with the plutonium to uranium mass fraction ratio. Since this is possible, it is also possible to determine the mass fraction of plutonium, which is of concern for safeguards purposes. It is assumed that from a statistics perspective, the XRF and KED measurements are uncorrelated. This is true due to the fact that XRF is a relative measurement. The absolute density of uranium in the salt is not of importance to XRF, but rather the relative uranium to plutonium ratio is.

$$\begin{aligned}
Pu_{Mass\ Fraction} &= (KED_{U\ Mass\ Fraction} \cdot XRF_{Mass\ Fraction\ Ratio}) \\
XRF_{Mass\ Fraction\ Ratio} &= \frac{Pu_{Mass\ Fraction}}{U_{Mass\ Fraction}}
\end{aligned}
\tag{4.3}$$

Combined HKED results showing the plutonium mass fraction in the electrorefiner as a function of time will be presented in the SSPM modeling results section.

4.4.1 Propagated Uncertainty

As mentioned before, for realistic expectation of HKED results, machine uncertainty for both KED and XRF must be accounted for. These are both conservatively assumed to be 1% [22]. These are propagated as shown in equations 4.5 and 4.4.

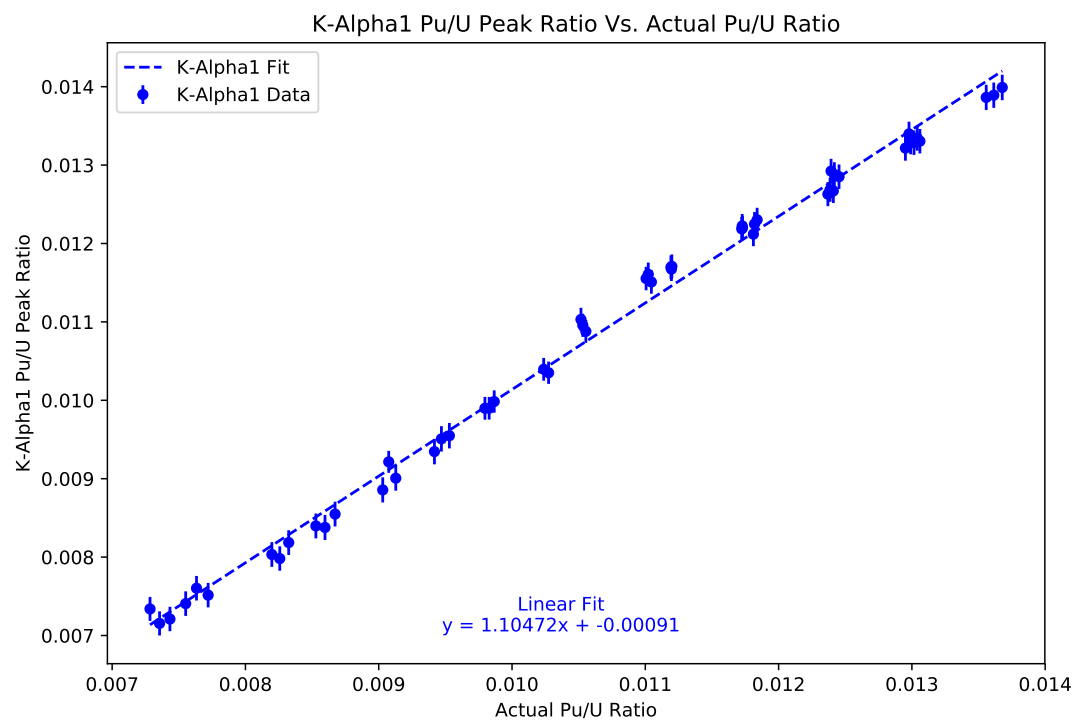


Figure 4.3: XRF K- α_1 Fitting

$$\begin{aligned}
\sigma_{XRF_{Total}} &= (x^2 * 0.012261^2 + 0.01^2 * x^2) \\
Relative \sigma_{XRF_{Total}} &= 1.58\% \\
x &= XRF \text{ peak ratio}
\end{aligned} \tag{4.4}$$

$$\begin{aligned}
\sigma_{KED_{Total}} &= (x^2 * 0.04485^2 + 0.01^2 * x^2) \\
Relative \sigma_{KED_{Total}} &= 4.60\% \\
x &= KED \text{ K-edge drop}
\end{aligned} \tag{4.5}$$

4.5 Application to SSPM

The application of the linear fits for KED and XRF, along with the combined HKED calculation for plutonium mass fraction is straightforward, and explained in this section. The first thing to note again is that SSPM keeps track of mass flows via a 1x100 vector, with each element between 1-99 representing an elemental mass quantity. Units do not matter here as the quantity being calculated is unitless mass fraction. Element 100 is the mass of the salt in the area. For this project, the electrorefiner was the section of interest, and all calculations were done there. In theory, the electrorefiner provides the best promise for HKED measurements due to what is ideally a thorough mixing of fuel and salt.

The application to SSPM did have to have minor adjustments, due to the aforementioned small negative offset of the fits. At time 0, when fuel is still making its way through the SSPM, the electrorefiner may have 0 quantities of either plutonium or uranium. This can cause the predicted values for the uranium K-edge drop and the uranium and plutonium XRF peaks to be incorrect. What should be recorded as 0 predicted plutonium mass is instead recorded as a negative value. To address this, whenever the predicted mass of uranium or the mass fraction ratio of plutonium to uranium is less than 0, the predicted mass from the empirical fits is set to 0.

To produce what is assumed to be a more realistic set of results for each SSPM simulation, Gaussian noise is added to the calculated uranium K-edge drop, and the calculated plutonium to uranium XRF ratio. To do this, the uncertainty from (4.5) and (4.4) is used for the Gaussian noise of the K-edge drop and XRF ratio, respectively. These Gaussians have a mean which is the calculated value for each respective calculation. This results in the propagated uncertainty for the combined HKED calculation listed in (4.6).

$$\begin{aligned} \text{Relative } \sigma_{HKED} &= (\sigma_{KED}^2 + \sigma_{XRF}^2)^{0.5} \\ \text{Relative } \sigma_{HKED} &= 4.86\% \end{aligned} \tag{4.6}$$

It should be noted, that for benchmarking purposes, the mass of plutonium reported by SSPM in its mass vector, divided by the total mass in the electrorefiner fuel-salt mixture reported by SSPM, is taken to be a true value. This is then compared to the calculated value of the plutonium mass fraction predicted by the empirical fits.

4.5.1 Modeling Results

The results from the SSPM implementation are very good, with results falling within 5 standard deviations of the mean for all values. For benchmarking purposes, 2 different fuel compositions are used within SSPM. The first is for $\frac{45GWd}{MTU}$, 4% enriched, 5 year cooled spent fuel. The second is for $\frac{33GWd}{MTU}$, 2.6% enriched, 5 year cooled spent fuel. The $\frac{45}{MTU}$ fuel results will be shown and discussed first.

For the $\frac{45}{MTU}$ fuel, the plutonium mass fraction matches very well to the true values of SSPM, as shown by 4.4. The quantified difference between the SSPM values and the calculations is shown in 4.5 and 4.6.

For the $\frac{33}{MTU}$ fuel, the plutonium mass fractions calculated again match the SSPM values within 5 standard deviations, with most cases falling within 3 standard deviations. The results, including quantified percent differences, are shown in figures 4.7, 4.8, and 4.9.

It is interesting to note that the $\frac{33}{MTU}$ fuel case, as compared to the $\frac{45}{MTU}$ fuel case, has a higher mass fraction of plutonium consistently. When comparing this to the original

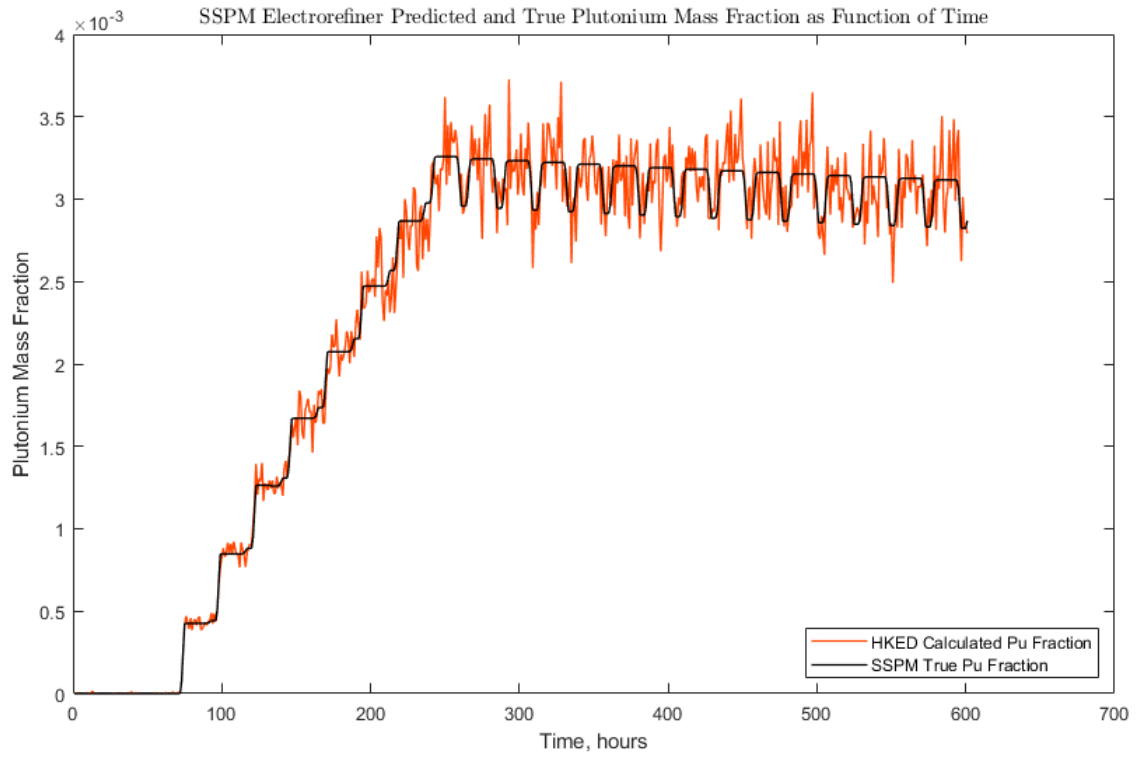


Figure 4.4: This figure shows plutonium mass fractions reported by SSPM and the empirical fits plotted versus time. This figure is using 4.0 % enriched, $45 \frac{GWd}{MTU}$ spent fuel.

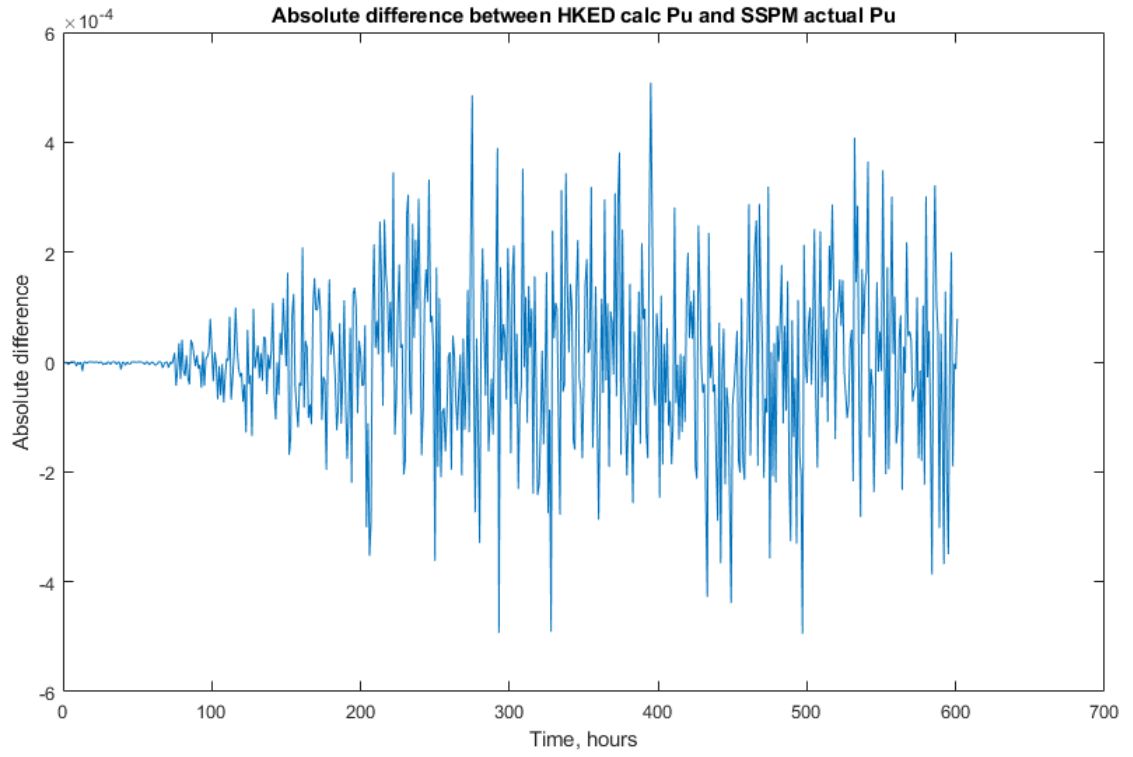


Figure 4.5: This figure shows the absolute difference between the calculated and true values of plutonium mass fraction. This figure is using 4.0 % enriched, $45 \frac{GWd}{MTU}$ spent fuel.

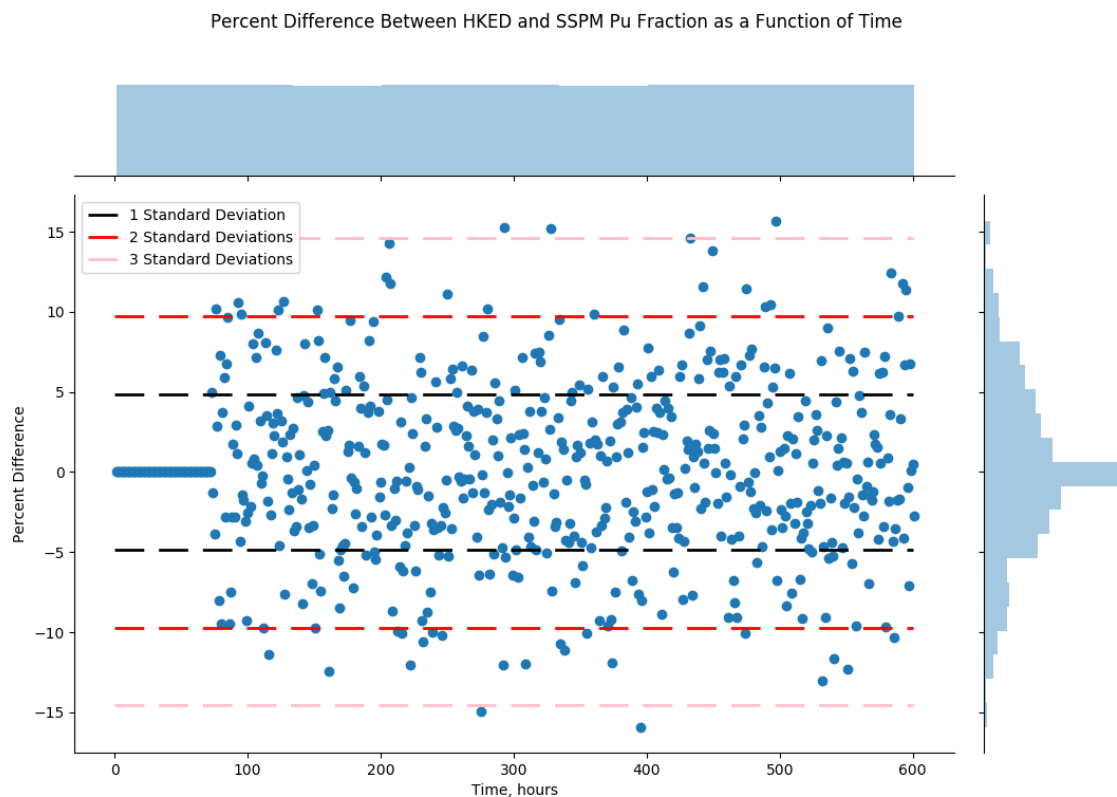


Figure 4.6: This figure shows the percent difference between the calculated and true values of plutonium mass fraction. Note that there is 0 percent difference until around hour 75 due to spent fuel not reaching the electrorefiner until then. This figure is using 4.0 % enriched, 45 $\frac{GWd}{MTU}$ spent fuel.

spent fuel cases from ORIGEN, this cannot be possible. A very likely explanation is that these sample SSPM cases were ran using the old SSPM spent fuel database, and not the data that was developed independently by Michael Cooper and Amanda Bachmann [23]. It is known from previous investigation that there are errors in the old Sandia database for SSPM. Bachmann's database is implemented into the newest version of SSPM, however, the newest version may have issues with mass flows. When it is certain that the new version of SSPM is properly working, the HKED module will be slightly modified to work with this newer version.

To provide another comparison, the results of the HKED calculation without Gaussian noise were compared to the true values from SSPM. These cases could be plotted showing true values versus HKED values, however the plot lines would completely overlap. This is because that since the KED and XRF fit values are calculated using the SSPM mass vector, they must precisely (within rounding error) be the same. Indeed, when the numbers are compared they are nearly identical, proving that the HKED module is correctly implemented into SSPM.

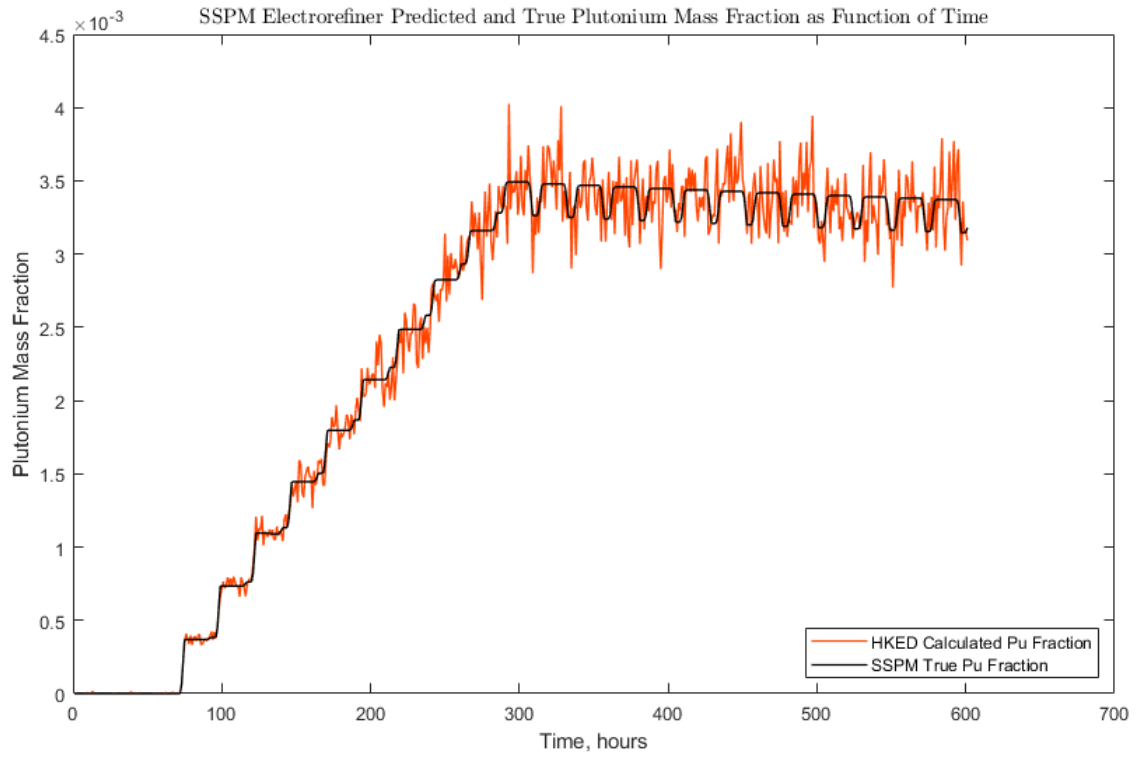


Figure 4.7: This figure shows plutonium mass fractions reported by SSPM and the empirical fits plotted versus time. Values reported as from the SSPM mass vector are considered true. This figure is using 2.6 % enriched, $33 \frac{GWd}{MTU}$ spent fuel.

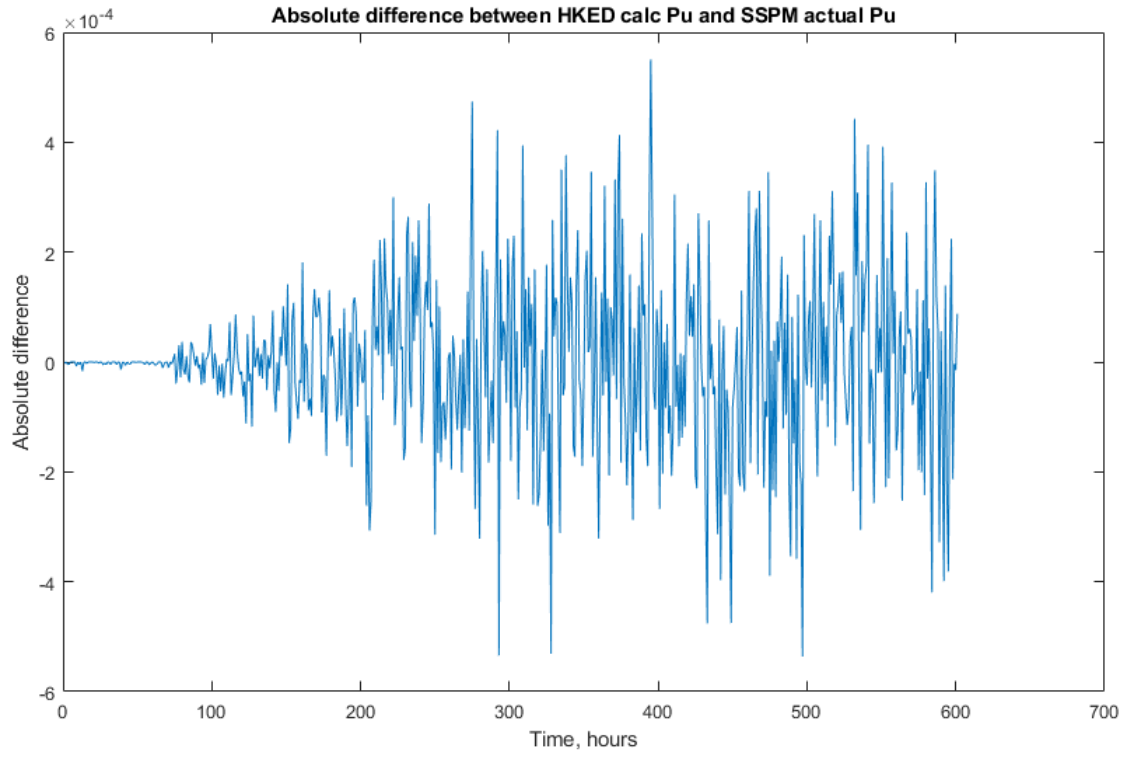


Figure 4.8: This figure shows the absolute difference between the calculated and true values of plutonium mass fraction. This figure is using 2.6 % enriched, $33 \frac{GWd}{MTU}$ spent fuel.

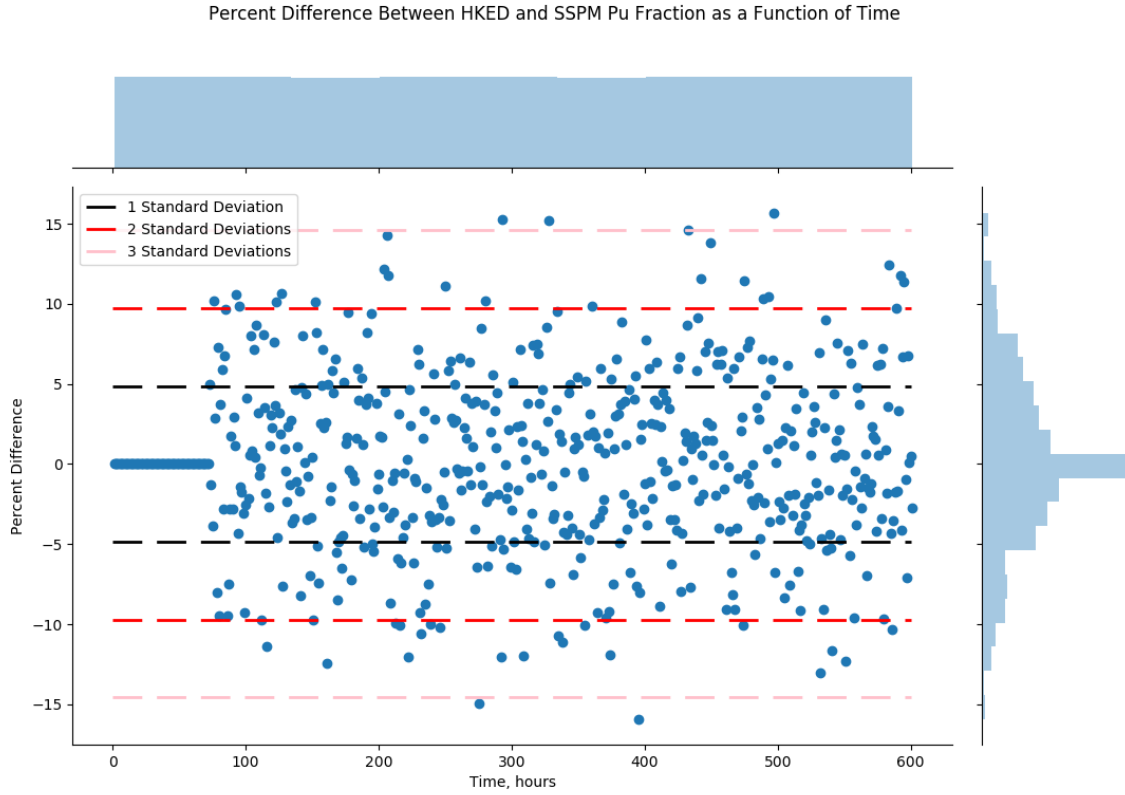


Figure 4.9: This figure shows the percent difference between the calculated and true values of plutonium mass fraction. This figure is using 2.6 % enriched, $33 \frac{GWd}{MTU}$ spent fuel.

Chapter 5

Conclusions

The modeling effort for implementing hybrid K-edge densitometry into the SSPM was successful overall. Initially a large spent fuel data set was generated for empirical modeling, but initial modeling attempts showed that for KED, the stage-1 uncertainties were too high, and were influencing the stage-2 simulations. 5 significantly higher particle count simulations were then performed, resulting in higher quality KED fitting.

Empirical fits were produced for KED and XRF simulation data, which was then combined into the HKED module for SSPM. These fits have quantified uncertainty values. These uncertainty values are then used to add Gaussian based noise to the HKED results. A limited number of test cases were performed with the SSPM module, with the results for the HKED calculated plutonium mass fractions falling within 5 standard deviations of the true values. For the HKED calculations, the overall uncertainty is 4.86%. This value is higher than common experimental uncertainties, however, it is mostly driven by MCNP uncertainty. As discussed in this thesis, in order to drop the HKED uncertainty below 1% would require approximately 50-100 times more computer simulation time with the same hardware used for this project.

When the Gaussian noise is not used, the HKED calculated mass fractions are always within 0.1% of the true values as expected.

The goal for the HKED module construction was to provide a simulation module for SSPM. This is part of an overall goal to include multiple measurement technique simulation modules into SSPM. This will allow for combining the measurements to reduce overall

uncertainty, and provide a greater confidence level of the inventory difference and process monitoring. As such, the next step for the SSPM pyroprocessing modeling is to incorporate the various measurement simulation modules in a data fusion process. Of interest is the effect on overall SSPM simulation confidence of including HKED into the suite of measurement modules. This is planned to be done over summer 2019 and early fall 2019, and is outside the scope of this thesis.

It is the author's hope that the HKED empirical modeling techniques presented in this thesis will be of help to any future researchers working on pyroprocessing safeguards models that wish to include HKED in their simulations.

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Vita

Michael Cooper was born to Kenneth H. Cooper and Lisa M. Cooper in Knoxville, Tennessee on November 10, 1992. Michael has 1 brother, Matthew Cooper, who is an undergraduate at UT. Michael was homeschooled starting in 3rd grade, then started community college at age 16. After his final year of highschool, Michael enrolled at UT for engineering. As a child, Michael was influenced by his father, who was a manager at Y-12 National Security Complex, resulting in a desire to learn as much as possible about nuclear science. While attending UT, Michael realized that he enjoyed working both with physics and computer simulations, and that achieving a PhD in nuclear engineering would allow him to combine the two topics in projects at national laboratories such as LLNL and ORNL.

Michael intends to complete his PhD in nuclear engineering, and then obtain employment at LLNL working nuclear defense related projects.