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**PROCESS INFORMED SAFEGUARDS APPROACH FOR A
PYROPROCESSING FACILITY**

By

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Dedication

This work is dedicated to Dr. Michael Lineberry, without whom this work would not have begun, and to my amazing wife Tam and our daughter Permelia.

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List of Abbreviations

Active Inventory- AI

Argonne National Laboratory- ANL

Basket module- BM

Containment & Surveillance- C/S

Continuity of knowledge- CoK

Destructive Assay- DA

Experimental Breeder Reactor-II- EBR-II

Electrorefiner- ER

Fission Chamber- FC

Formula quantity- FQ

Fuel Conditioning Facility- FCF

Giga-Watt days per Metric ton of initial Heavy Metal- GWd/MTiHM

Heavy metal-HM

High purity germanium- HPGe

Idaho National Laboratory- INL

International Atomic Energy Agency- IAEA

Inventory Difference- ID

Key measurement point- KMP

Korea Atomic Energy Research Institute- KAERI

Light Water Reactor- LWR

Limit of error in inventory difference- LEID

Los Alamos National Laboratory- LANL

Low enriched uranium- LEU

Material Access Area- MAA

Material Balance Area- MBA

Material Unaccounted for- MUF

Materials Controls & Accountability- MC&A

Next Generation Safeguards Initiative Spent Fuel Assay Project- NGSI-SF

Non Destructive Assay- NDA

Non-Nuclear Weapon State- NNWS

Non Proliferation Treaty- NPT

Nuclear Weapon State- NWS

Oxide transfer can- OTC

Passive gamma ray detection- PG

Passive Neutron Albedo Reactivity- PNAR

Physical inventory- PI

Pressurized water reactor- PWR

Protected area- PA

Pyroprocessing Safeguards Performance Model- PSPM

Republic of Korea- ROK

Rokkasho Reprocessing Plant- RRP

Safeguards & Security- S&S

Safeguards Hazards Matrix- SHM

Self- Interrogation Neutron Resonance Densitometry- SINRD

Significant quantity- SQ

Special Nuclear Material- SNM

Spent nuclear fuel- SNF

Standard error in inventory difference- SEID

Standardized, Independent Material Unaccounted For- SITMUF

Thermal Oxide Reprocessing Plant- THORP

Total neutron counting- TN

Transuranic- TRU

Unified Nondestructive Assay- UNDA

US Department of Energy- DOE

US Nuclear Regulatory Commission- NRC

Vital Area- VA

Abstract

Electrochemical reprocessing has been proposed as a spent fuel disposition technique. If implemented on a commercial scale the current safeguard approaches used to satisfy IAEA requirements may not be adequate. To aid in testing of safeguard strategies, two tools were developed; 1) the Pyroprocessing Safeguards Performance Model (PSPM), a MATLAB/Simulink simulation of plant operations capable of calculating inventory differences (ID) for a specified balance period as well as sensitivity studies of detection measurements, and 2) the Safeguard Hazards Matrix (SHM), a risk matrix developed using the calculated ID and probability to detect one significant quantity (SQ) to represent the risk of diversion within a reference facility. Using both tools, the inclusion of process data (using electronic mass balances within the material balance area (MBA) at each processing zone) was tested along with the black box approach and a conventional safeguards approach. The process informed strategy provided the most acceptable scenarios with detection techniques possible within the foreseeable future. A brief overview of reprocessing regulation is given with regulation gaps identified as well.

1. Introduction

This dissertation is focused on the electrochemical reprocessing technique known as pyroprocessing. The technique is known by a few names throughout the literature, electrochemical processing, pyroprocessing, or electrometallurgical processing; for this work it will be referred to as pyroprocessing. The work will provide a summary of the current regulatory environment for a reprocessing facility as well as an overview of current nondestructive assay (NDA) techniques applicable for pyroprocessing. Two tools for evaluating the effectiveness of a safeguards approach have been developed: the Pyroprocessing Safeguards Performance Model (PSPM) and the Safeguards Hazards Matrix (SHM). The PSPM is a MATLAB/Simulink¹ simulation of plant operations capable of calculating inventory differences (ID) for a specified balance period as well as sensitivity studies of detection measurements. The SHM is a risk matrix developed using the calculated ID and probability to detect one significant quantity (SQ) to represent the risk of diversion within a reference facility. Four safeguard strategies were evaluated using both these tools; a black box approach, a conventional approach, and two variations of a process informed approach. The results of these tests show a great benefit can be gained by including some process monitoring data into the safeguards evaluation.

Currently two techniques are used for reprocessing nuclear fuel, aqueous reprocessing and pyroprocessing. Aqueous reprocessing has been employed since the

¹ The Mathworks.Inc., MATLAB and Simulink R2014a, Natick, MA, (2014)

Manhattan Project as a plutonium (Pu) separation process. The process has evolved since then into the common PUREX (Plutonium & Uranium Extraction) process employed at five commercial reprocessing facilities located worldwide. An overview of aqueous reprocessing begins with spent fuel assemblies being chopped and dissolved in nitric acid. The dissolved fuel is then transferred to the accountability tank, where a sample of the solution is taken to determine isotopic composition of the input fuel assembly to very low uncertainty (0.1-0.5 %) (IAEA, Nov 2010) and the mass of the solution is measured. Next, an organic solvent with Tributyl Phosphate (TBP) is introduced to preferentially extract U and Pu. There are different configurations that can be employed depending on the desired products from this point; in most cases U and Pu are each separate products with purity levels of 99.99% or better or in another configuration the U and Pu can be mixed together after separation in a 50:50 mix and then stored (similar to the process at Rokkasho Reprocessing Plant in Japan). The remaining waste, aka raffinate, is highly radioactive and is vitrified and formed into a waste glass to be sent to a repository. By adjusting the solvent concentrations, the other actinides could be separated as well or they could all be extracted together in one step. Several extraction stages results in a high purity product, which is an attractive attribute for a potential proliferator. It must be noted that an aqueous facility is significant in size – a four square kilometer complex, and contains tens of kilometers of piping - thus it is very difficult to track material throughout the facility at any moment in time.

Similar to the aqueous process, there are a few variants of pyroprocessing. These variants are based upon the input spent fuel stock rather than the desired output products. Argonne National Laboratory (ANL) initially developed melt refining (the predecessor to

the modern pyroprocess) in the early 1960's for reprocessing of metal fueled fast reactors, such as the Experimental Breeder Reactor-II (EBR-II). An engineering scale facility (10 MT/yr throughput) was co-located with the EBR-II facility originally known as the Fuel Cycle Facility (FCF). The melt refining process was a simple reprocessing technique in which the spent fuel was melted and about 5-10% of the fission products were removed. After which the melt was recast into new fuel pins. The process was demonstrated from 1965 to 1969 and 44,000 fuel pins were recast and irradiated in EBR-II; demonstrating a closed fuel cycle. (Koch, 2008) Lessons learned from the initial melt refining operation led to the development of the modern pyroprocess. In 1994, EBR-II's operation was ended and FCF (in the process of being refurbished at the time) was selected to process the spent fuel before being sent to interim storage and finally a geological repository. FCF became the demonstration facility for the pyroprocess in 1995 and has been in operation since that time. Pyroprocessing variants are dependent upon input fuel chemical composition. FCF processes using PYRO-A, the input fuel is metallic, rather than PYRO-B where the input fuel is an oxide (such as those in current LWRs). PYRO-B has an extra step to convert the oxide fuel into a metallic form as compared to PYRO-A. This work deals with PYRO-B but its conclusions are applicable to PYRO-A as well. Throughout the rest of this dissertation, pyroprocessing will refer to PYRO-B; unless specifically noted.

The Republic of Korea (ROK) has specifically identified pyroprocessing as the technology of greatest benefit to them. If they continue in this effort then International Atomic Energy Agency (IAEA) safeguards will need to be implemented for this technology which IAEA has no experience regulating. This will most likely require an

international effort similar to the Large Scale Reprocessing (LASCAR) project (IAEA, 1992) developed after Japan first decided to build the Rokkasho Reprocessing Plant (RRP), a large scale (800 MT/yr throughput) aqueous reprocessing facility. As part of this strategy, inventory periods were shortened from the traditional 30-day inventory to 15-day virtual inventories. A study by Lineberry demonstrated this shortened inventory period is possible for RRP but if it is applied to fuels with higher Pu loading (i.e. fast reactor fuels) then even an inventory every day would be insufficient to meet current detection goals (Lineberry, 2011). This study shows that a new approach is needed if larger throughput or fast reactor reprocessing facilities are to be built in the future.

A new concept to safeguards is the implementation of a safeguard system during the conceptual design phase of the facility; this is known as 3S by Design: Safety, Security, and Safeguards (IAEA, 2013). Historically, safety has been well investigated during the design stage and to some extent security as well, but safeguards (within a Nuclear Weapons State (NWS) such as the US specifically) have been an add-on feature typically as the facility is being constructed. When applying 3S by Design, facility designers begin to integrate each of these important areas much earlier than previously considered. Regulators are also included earlier in the proposed facility's plans for each of these. Considering and planning for safety, security, and safeguards in the facility prior to construction will provide adequate space for all instrumentation. Another advantage of adding safeguards in the design stage is that the regulator has an increased amount of time to verify that the particular strategy for the facility is appropriate and whether further measures are warranted. To accommodate these changes while the facility is still on paper is relatively inexpensive but to change the structure once

construction has begun can be very costly.

1.1 Purpose of Research

This dissertation develops two tools for evaluating a potential safeguard strategy at a pyroprocessing facility and makes recommendations based upon their results. The proposed technique for evaluating safeguard risks at a facility is the Safeguard Hazards Matrix. The SHM is plotted as the probability of detection of one significant quantity of material against the calculated ID. This approach is analogous to deterministic hazard analysis performed for nuclear safety. A MATLAB/Simulink simulation model has also been created based upon a reference pyroprocessing facility, the Pyroprocessing Safeguards Performance Model (PSPM), and several proposed safeguard strategies are evaluated. Once the risk is determined for a strategy then additional measures can be implemented to reduce the risk to an acceptable regulatory level. A final recommendation for a facility materials, controls, and accountability (MC&A) program is made based upon the results obtained using both the SHM and PSPM.

2. Regulation Review

Before a new safeguard strategy can be suggested, current regulations regarding reprocessing facilities need to be understood. For this work three likely regulators of a future facility were studied: International Atomic Energy Agency (IAEA), US Nuclear Regulatory Commission (NRC), and US Department of Energy (DOE).

2.1 IAEA

Each State, which is a signatory party to the Non Proliferation Treaty (NPT), known as Member States, agrees to bring into force safeguards agreements with the IAEA. A caveat, though, the NWS which include: US, Russia, China, UK, and France, are not required to have a safeguards agreement with the IAEA; though all now have voluntary agreements with the IAEA, these are not comprehensive and facilities related to defense are often excluded from their scope. The goal of IAEA safeguards is:

The timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection (IAEA, 2002).

To implement safeguards, a material accountancy program and containment and surveillance (C/S) programs must be in place. Each Member State enters into a safeguards agreement with the IAEA and states the objectives and facilities that will be subjected to this agreement. The IAEA has a limited work force so non-nuclear weapon

States (NNWS) are prioritized for inspections. If identical reprocessing facilities were built in a NWS and a NNWS the facility in the NNWS would be subject to much more rigorous inspections to verify weapons material is not being misused. The NWS on the other hand could simply not include the facility in its voluntary offer with the IAEA and would not be subject to inspections.

Currently two reprocessing facilities, both in Japan, are under IAEA safeguards: RRP and a smaller facility at Tokai. RRP is in a protracted startup mode while Tokai is in the process of shutting down. However, there are four reprocessing facilities operating in weapon States: Thermal Oxide Reprocessing Plant (THORP) in UK, UP 2 & 3 in France, and Mayak in Russia. Since NWS have voluntary offers with the IAEA, the State is able to choose which facilities to allow inspectors in and the extent of those inspections, thereby lowering a facility's safeguards burdens and ultimately lowering its operating cost compared to a comparable facility in a NNWS. The safeguards burdens imposed on NNWS can be alleviated if the state regulations of a NWS are as rigorous as IAEA safeguards, but that is not a factor controlled by the IAEA.

An important concept in IAEA material accountancy is the material unaccounted for (MUF). MUF must be less than the mass of nuclear material needed to fabricate a crude weapon, known as a Significant Quantity (SQ).

$$MUF = inputs + BI - outputs - EI \quad (1)$$

BI and EI refer to the beginning and ending inventory for the balance period respectively. For Pu, the SQ is 8 kg and for low enriched U it is 75 kg of U²³⁵. Materials are also divided into categories: “direct-use” in a weapon and “indirect use”, both aimed at

achievement of a timeliness goal. Pu and enriched U >20% in U-235 are considered direct-use while low enriched U (LEU) and Th are indirect-use. The detection goal for unirradiated, direct use material is 30 days, irradiated, direct use material is three months, and indirect use material is one year. A measurement system must also have a 95% detection probability to detect a diversion of a SQ and a 5% false alarm probability. Operating experience from bulk handling facilities (such as reprocessing plants) has shown MUF to be a standard normal distribution, as described by equation 2.

$$f(x) = \frac{1}{\sqrt{2\pi}} e^{-\frac{(1-x)^2}{2}} \quad (2)$$

Integration of equation 2 with x ranging from $-\infty$ to ∞ results in a probability of 1. If the probability desired is instead 0.95 the resulting x upper limit value is 1.65, this x value corresponds to both the detection probability and alarm probability requirements. Since both the detection probability and alarm probability are the same, a double counting arises. To assure detection of a loss of one SQ, equation 3 must hold true (Burr, 2013).

$$\sigma_{\text{MUF}} \leq \frac{\text{SQ}}{3.33} \quad (3)$$

For Pu and LEU σ_{MUF} , must be less than 2.402 and 22.7 kg respectively; σ_{MUF} is also known as the standard error in inventory difference (SEID) by other regulatory bodies. (IAEA, 2002)

To complement material accountancy, containment and surveillance (C/S) provisions are also implemented as “complimentary” provisions. One C/S strategy is “continuity of knowledge” (CoK) where material is tracked throughout the facility to verify tampering is not occurring. With a batch scheme it would be useful to track the batch from initial chopping to storage. Some preliminary research was done to implement

this at the FCF and analyze how it could be implemented in a pyroprocess plant (Bean, 2010).

Safeguards for large aqueous reprocessing plants such as RRP are a challenge because of the difficulty of measuring Pu within the extensive piping system and the nature of the large footprint of the facility. A pyroprocessing plant does not have the complexity of extensive piping and tank systems as an aqueous plant; however, it does not have a homogenous input measurement point where the fuel assembly content can be initially measured accurately. However, as throughputs increase, even with a homogenous input measurement point, timeliness and material detection goals cannot be achieved with current detection abilities. This is the case at RRP; therefore, the enhanced safeguards applied should be reviewed for insight into alternative safeguards strategies for any pyroprocessing facility. At RRP, enhanced safeguards measures were implemented to assure “Operations as Declared” (Johnson, 2010). These enhanced safeguards measures rely on preserving CoK for material within each Material Balance Area (MBA) using C/S measures. Over 70 closed circuit cameras are used throughout the process to monitor material flows in process and storage areas (Johnson S., 1997).

Physical protection is not explicitly mentioned in safeguarding SNM by the IAEA; however, an effective physical protection strategy uses C/S measures to accomplish its goal. The IAEA also recommends standards for physical protection but the member States retain an inalienable right to their own determination of physical protection. As such the physical protection recommendations are not enforced by the IAEA and left to member States. Thus, security of special nuclear material (SNM) is the

focus of C/S but the exact measurement techniques to be used are left up to the discretion of the member States.

2.2 NRC

Nuclear material is defined by what it consists of and how much is present. SNM is defined as plutonium, uranium 233, or uranium enriched in the isotopes of 233 or 235 (10CFR50.2, 2007). It is broken up into strategic importance levels: special nuclear material of low strategic importance (Category 3), special material of moderate strategic importance (Category 2), and strategic special nuclear material (Category 1). Categories are determined based on an effective kg, also referred to as a formula quantity, of material in an inventory period. A formula quantity (FQ) is defined as:

$$FQ = (\text{grams of } U^{235}) + 2.5 * (\text{grams of } U^{233} + \text{grams of Pu}) \quad (4)$$

Category 1 is defined as $FQ > 5 \text{ kg}$, category 2 as $1 \text{ kg} < FQ < 5 \text{ kg}$, and category 3 as $FQ < 1 \text{ kg}$ but greater than “reportable quantities”

The NRC MC&A requirements for a facility are found in 10 CFR 74. This section of the regulation outlines the performance of any MC&A system for each of the categories. The facility will contain formula quantities of SNM, however 10 CFR 74.51 states that the section, which describes the performance of an MC&A program for FQ facilities, is not applicable to irradiated fuel reprocessing facilities (10CFR74.15a, 2002). There is not another section with this part 74, or any other part of CFR, that describes the MC&A program objectives and performance criteria for a reprocessing facility.

The NRC has conducted an investigation into regulatory gaps associated with licensing a reprocessing facility. 18 gaps were identified and a framework is being

pursued to resolve these gaps by 2032 (Nuclear Regulatory Commission, 2009). The NRC has received letters of interest from five industry leaders regarding a rulemaking on reprocessing facilities.

Regulation on the protection of SNM is found in 10 CFR 73. The physical protection of SNM is applicable for both fixed sites and transportation of SNM. Only the fixed site regulations will be discussed further for the facility, but transportation safety will be important in a license application. The NRC uses a strategy known as “defense-in-depth” for physical protection of SNM. This strategy requires at least three physical barriers (walls, fences, sealed containers, etc.) be placed between the general public and the SNM. A complementary idea used with this strategy is the restricting of personnel movements. This is accomplished using security zones with monitored access control points. These security zones are exclusion zone, protected areas (PA), isolation zone, material access areas (MAA), and vital areas (VA). Nesting of these security zones help to accomplish defense-in-depth. The exclusion zone is an area removed from public use, sometimes known as the site boundary; however, highways or other roads can cross this zone. The PA is enclosed by two physical barriers and contains an intrusion detection capability between the two barriers. Access to this area should be limited and all personnel entering the area should be searched to ensure prohibited material or objects are not entering the facility. The isolation zone is located just outside the perimeter of the PA and is large enough so that guards can observe personnel activities on either side of the PA. Parking lots should be located outside of the isolation zone. A MAA lies within the PA and contains any areas where SNM can be accessed or is processed. A VA is also located within a PA and contains equipment that has been deemed vital to the protection

of the public from radiological exposure. VAs are determined based upon the threat assessment of the facility. Threat assessments are detailed analyses on the types of threats a facility is required to defend against. As such, this information is classified and site-specific, and therefore actual threat assessment is not part of this work. A threat assessment would be required in any license application for a facility under consideration.

SNM must be located within an MAA in a secured container, either locked within the processing vessel or within a vault with tamper evident seals, or under the supervision of at least two guards at all times. Stored Category 1 material is required to be located within a vault with intrusion detection when not accessible and guards must be present when the vault is accessible.

2.3 DOE

The DOE approach to safeguarding SNM is done under two programs that a facility must implement: MC&A, and safeguards and security (S&S). The DOE has the most experience with regulating a pyroprocessing facility of the three regulatory bodies; DOE has been operating FCF since 1995.

The DOE defines attractiveness of a material for its use in a nuclear weapon, as well as the quantity of material present, to determine the safeguards approach, known as a graded approach. Attractiveness levels range from A to E; level A is an assembled nuclear weapon, level B consists of pure products that might require some work to create a nuclear weapon, level C consists of high grade materials such as fuel elements and assemblies that are unirradiated, level D consists of low grade materials such as uranium

enriched between 20% and 50% in ^{235}U , and level E consists of all other materials that contain SNM above reportable quantities. Within each attractiveness level there are four categories, I, II, III, IV, determined by the amount of material; the requirements for each can be seen in Appendix B.

The S&S program consists of the typical elements of physical protection to protect the material from external threats, while the MC&A program focuses more on internal diversion attempts. To effectively implement the S&S program, a strategy similar to the NRC has been developed using multiple levels of increased security to provide sufficient barriers to delay and detect theft. These areas are similar to the NRC security zones and are nested so that the more restrictive zones lie completely within a less restrictive zone.

The MC&A program for nuclear material is based around inventory difference (ID) and limit of error of inventory difference (LEID), using IAEA terminology these are MUF and σ_{MUF} respectively. DOE requires $\text{ID} < 2 * \text{LEID}$ and $\text{LEID} < 2\%$ active inventory (AI). There is an additional requirement that ID also must be less than a Category II amount of SNM; for attractiveness levels C and B this is 6 kg and 2 kg of Pu respectively. To account for SNM throughout the facility, MBAs will be set up and material crossing an MBA boundary must be monitored and accounted for. Physical inventories for MBAs are based on the category of material and type of operations performed within it. For process MBAs of Category I or II material a Physical Inventory (PI) must be done at least every 60 days, while Category III or IV MBAs are every six months. For non-processing MBAs a PI is done every six months for Category I or II and every two years for

Category III or IV. Other accountable nuclear materials in an MBA are inventoried every two years.

The DOE has more defined areas each with increased barrier protection for SNM than the NRC, though both employ the “defense in depth” philosophy. While both the DOE and NRC require a well-defined MC&A program, only DOE has regulations and performance criteria in place for a reprocessing facility; NRC has criteria established for research facilities and enrichment facilities but these are not and cannot be applied to large bulk handling facility such as a reprocessing plant. If attractiveness B material is produced in the U/TRU product than the requirement that more than 2 kg of material difference be investigated will be more restrictive than the IAEA requirement of 2.40 kg.

3. Reference Facility Description

A reference pyroprocessing facility will be assumed to have an annual LWR spent nuclear fuel throughput of 100 MTiHM/yr. The facility design is based on reprocessing PWR 17x17 fuel assemblies which have an initial enrichment of 4.5% and burn-up of ~50 GWd/MTiHM. The assemblies will be discharged from the reactor for ~10 yr before being reprocessed. To determine the initial isotopic distribution within the spent fuel assemblies an ORIGEN calculation was performed using the above parameters. Figure 1 provides a flow diagram for the process implemented at the reference facility (Benedict). The arrows represent chemical forms of the materials to and from each process; solid blue are solids, blue with a white stripe are solids and salt, and red are salt.

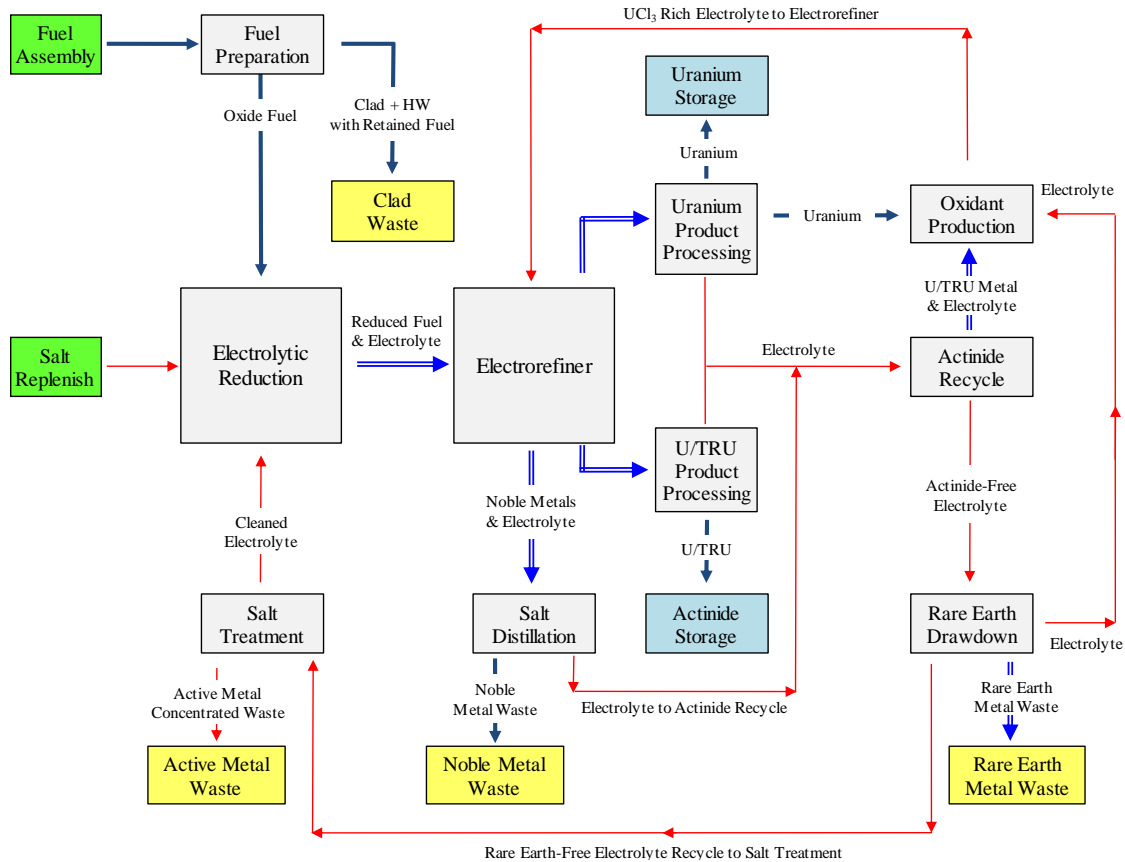


Figure 1 Pyroprocess flow diagram for LWR input fuel

The pyroprocess has a number of areas of interest with regard to safeguards application. The process will now be described in reference to these key areas. The spent fuel receiving and storage area encompasses the storage capacity of the facility, and houses the receiving equipment for spent fuel shipped from other sites. The air-atmosphere hot cell contains Non-Destructive Assay (NDA) equipment that will verify the assembly contents to determine the Shipper/Receiver difference and provide the initial safeguard measurement. Within this air cell, the assembly is disassembled and fuel pellets are placed in oxide transfer cans (OTC). From the air cell, the OTCs are moved into the argon atmosphere process hot cell and loaded into the processing basket module;

two OTCs are contained in a basket module. Once two basket modules are filled they are reduced to metal from their initial oxide form via electrolytic reduction and then moved to the electrorefiner (ER). The ER contains a lithium chloride-potassium chloride (LiCl-KCl) salt where the actinides including transuranic elements (TRU) and some fission products form stable chlorides. A voltage is applied across the anode and cathode, which induces the electro-transport of the uranium onto cathode rods. The U dendrite deposits on the cathode rod is scrapped off semi-continuously throughout the 72-hour processing of the basket module and transferred to the U processor. The TRU dissolved in the salt deposit on solid cathodes while the uranium transport is occurring (Laplace, 2008). As part of this transport, some uranium will also co-deposit with the TRU as well as a small fraction of rare earth elements (~0.1%). A U/TRU cathode is collected at the end of the basket module's processing. Boiling off the adhering salts purifies each product. The purified ingots are then moved from the process cell to their respective storage locations, the U/TRU vault and a U storage area. The adhering salts are collected along with some salt from the ER after three basket modules are processed. The salt's actinide composition is drawn down first. This is combined with uranium metal to produce the oxidant (UCl_3) needed to facilitate electro-transport in the ER. Once the actinides are drawn down then the rare earth wastes elements are drawn out. In the following step, the rare earth free salt is sent to a storage tank until another rare earth free salt batch is processed. This actinide and rare earth free salt has the active metals removed via salt distillation. The cleaned salt is then recycled into the electrolytic reduction vessel.

An analytical laboratory will also be needed on the facility site to analyze material samples taken at designated key measurement points to confirm that no Special Nuclear

Material (SNM) is being diverted and help resolve and detect differences. The samples will be on the order of a few grams and will be analyzed with destructive analysis (DA) and/or NDA techniques.

The pyroprocess allows for partitioning of elements based upon chemical properties; therefore, isotopic compositions of the input element are assumed to remain unchanged after processing. The U product therefore consists of uranium, enriched in ^{235}U , of 1%; the same enrichment of the UO_2 within the SNF input assembly. Therefore it will constitute Category 3 material (using NRC terminology) because it is classified LEU. The U/TRU product consists of Pu and U with an approximate ratio of 2.4:1. If the ingot mass is estimated as 8 kg, this is roughly equivalent to 15.9 FQ (using NRC terminology), therefore it will be classified as Category 1 material. The U/TRU material will drive the security and safeguard concerns for any facility design.

4. Non-Destructive Assay Overview

4.1 Technology

The current safeguard strategy for FCF relies on detailed depletion calculations of the input spent fuel assemblies. However, an IAEA inspector will most likely not be able to perform these calculations for each assembly processed; therefore NDA devices will be needed throughout the facility which can give accurate estimations (~1-5% uncertainty) of material compositions. Current NDA technology is not capable of these measurement accuracies, thus an extensive effort worldwide has been focused on updating NDA measurement capabilities. One such effort has the goal of determining the Pu content in a spent fuel assembly to a few percent accuracy (~1-2%), the Next Generation Safeguards Initiative Spent Fuel Nondestructive Assay Project (NGSI-SF) primarily located at Los Alamos National Laboratory (LANL) (Tobin, 2013). The NGSI-SF efforts identified 14 potential technologies and conducted a brief study of each system. Four integrated systems were suggested from the initial 14 technologies. The integrated systems proposed used four different techniques, which could complement each other and provide an estimate of elemental Pu mass. This can be achieved using Pu_{eff} described as:

$$Pu_{eff} = C_1 * U_{235} + Pu_{239} + C_2 * Pu_{241} \quad (5)$$

In equation 5, C_1 and C_2 represent the weighting of net neutron production within the fissile material; see equation 6 (Conlin, 2010).

$$C_1 = \frac{(\overline{\nu_{U235}} - 1)\sigma_{f,U235}}{(\overline{\nu_{Pu239}} - 1)\sigma_{f,Pu239}}$$

$$C_2 = \frac{(\overline{\nu_{Pu241}} - 1)\sigma_{f,Pu241}}{(\overline{\nu_{Pu239}} - 1)\sigma_{f,Pu239}}$$
(6)

In equation 6, $\nu-1$ refers to the net number of neutrons emitted, excluding the neutron inducing the fission event, and σ_f the microscopic fission cross section.

The NGSF-SF program is not specifically for reprocessing applications, therefore not all techniques investigated are applicable; only those promising for use in the reference facility will be discussed further.

The fissile content of an assembly (total of individual masses: ^{235}U , ^{239}Pu , and ^{241}Pu) can be estimated provided the assembly burnup, initial enrichment before irradiation, and cooling time since irradiation are known. The reactor operator before shipping to a reprocessing plant reports these quantities, where an independent verification must also occur, known as shipper/receiver difference. Two techniques currently used in NDA are passive gamma ray detection (PG) and total neutron counting (TN). PG uses relative intensities of gamma rays from fission product within the assembly to verify burnup or cooling. Two isotopes commonly used are ^{137}Cs and ^{154}Eu . TN also is another well-known measure to determine burnup using coincidence counting. Each of these techniques is integrated with other techniques so that the elemental Pu mass can be determined.

The most promising system for assaying spent fuel in an air environment is the Passive Neutron Albedo Reactivity / Self- Interrogation Neutron Resonance Densitometry (PNAR/SINRD). PNAR measures the amount of fissile material in the

assembly by measuring a ratio of neutron multiplicity between a "bare" fission chamber (FC) and one behind a cadmium shield, known as the Cd ratio (Conlin, 2010). The Cd ratio can be used to determine Pu_{eff} as,

$$Pu_{eff} = Cd\ ratio * fissile\ mass\ of\ assembly \quad (7)$$

The fissile mass in equation 7 is based upon the burnup determined using PG and TN.

SINRD uses FCs with filters sensitive to specific energy regions of the fissile isotopes enabling a mass to be determined for ^{235}U and ^{239}Pu (Hu, Fall 2012). Using equation 5, the mass of ^{241}Pu can be determined. An elemental Pu estimate is then achieved using a burnup code by knowing the burnup, initial enrichment, and the respective amount of ^{239}Pu and ^{241}Pu .

The detector is comprised of three sections, section one contains the "bare" FC for the PNAR ratio, section two contains ion chambers for PG and TN, and section three contains the SINRD filtered FCs as well as the cadmium shielded (cadmium shield is 1mm thick and 320 mm tall) FC surrounded by polyethylene for the PNAR ratio. The SINRD FCs, in section three, are wrapped in thin filters of gadolinium (0.025mm) + hafnium (0.5 mm), and cadmium (3.0mm) (Hu, 2013).

The proposed system for assay of a 17X17 PWR assembly is shown in Figure 2. An XZ view of the entire detector (left), XY view of section one (center) and XY view of section three with FC embedded in poly (right) are shown. Materials are shown as: red- SNF assembly, yellow- polyethylene, cyan- FC, green-ion chamber, orange- cadmium, and purple- aluminum housing.

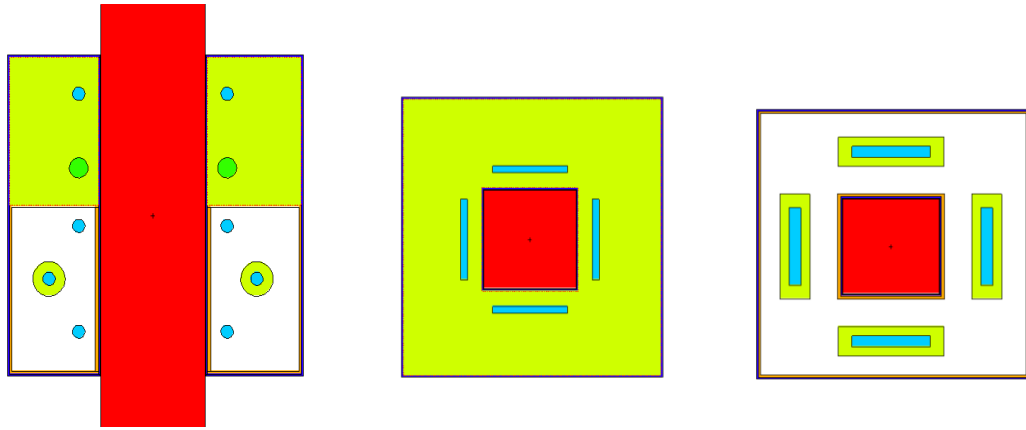


Figure 2 PWR PNAR/SINRD detector.

A PNAR/SINRD device is currently being tested in Japan at the Fugen test reactor; however, results have not been published at the time of writing (Tobin, 2013). A disadvantage of this technique is its insensitivity to the central pins within the assembly. The majority of the PNAR signal comes from the fissile material nearest the FC; therefore, a potential proliferator could remove up to eight inner fuel pins without detection (Conlin, 2010), corresponding to a 3.0% mass diversion with a higher percentage of Pu than the outer pins.

To enhance the confidence that pins have not been removed from the assembly, a neutron tomography system could be implemented to complement the PNAR/SINRD system. To create a neutron tomography image a neutron beam is passes through an object, to create a projection profile. The neutron beam is then rotated to additional positions to create multiple projections profiles. Integrating all of the projections profiles creates the tomography image. A simple example using three projection profiles and the resulting tomography image is shown in Figure 3 (Pope C. , 2011).

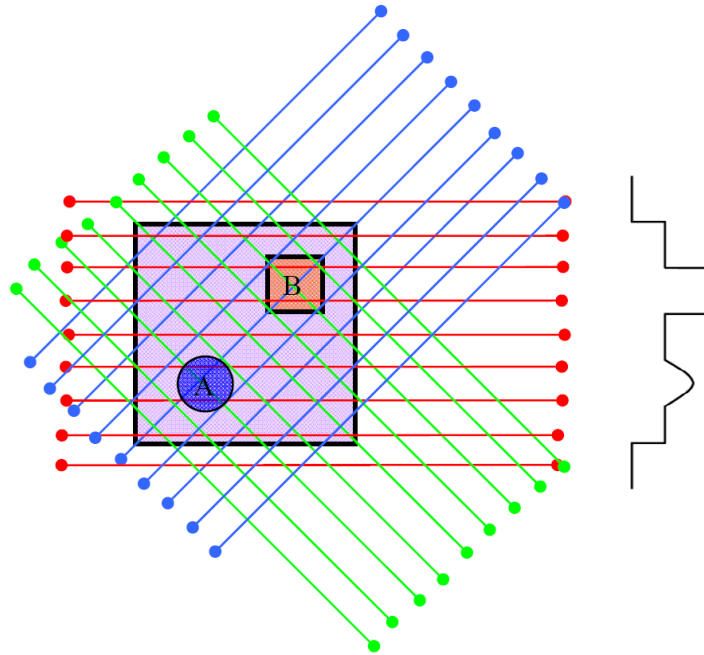


Figure 3 Tomography example with three projection profiles

The projection difference image technique generates a tomography image based upon the differences between known and unknown projections. Applying this technique to SNF assemblies, difference between a standard assembly and those about to be processed can be discovered. Using a 14 MeV neutron source, simulations have been shown to detect subtle material differences between the two generated images, for example substituting irradiated fuel pins with depleted uranium or fresh fuel pins (Pope C. , 2011). This technique is an excellent qualitative check to couple to the quantitative assay of fissile material estimate from PNAR/SINRD. A proposed system incorporating both a PNAR/SINRD detector and tomography-imaging suite is shown in Figure 4.

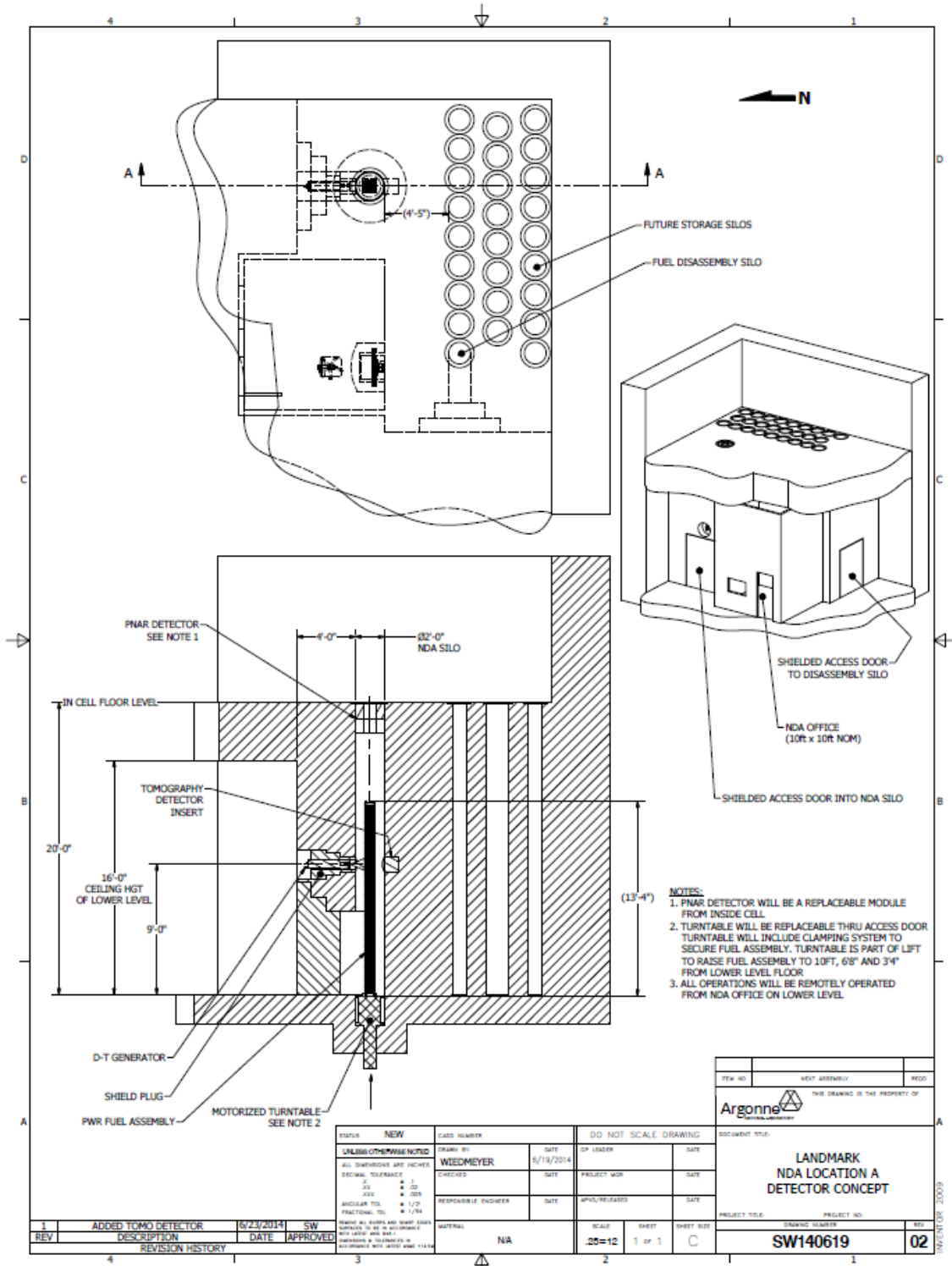


Figure 4 Conceptual design of PNAR/SINRD and Tomography suite

The Korea Atomic Energy Research Institute (KAERI) has also been developing NDA

devices specifically for pyroprocessing applications. KAERI has developed two devices, which show promise, the U/TRU ingot PNAR detector and the Unified NDA (UNDA) detector. The U/TRU ingot PNAR detector is based on the same PNAR technique for spent fuel (without integrating SINRD) and is scaled down to measure the fissile content of the U/TRU ingot (Lee, 2013). As discussed above, this technique can only measure fissile content but it is envisioned to be used in a confirmatory role in the safeguard strategy and not needed to provide Pu isotopic composition. A XY view (left) and YZ view (right) of the detector are shown in Figure 5 (Lee, 2013).

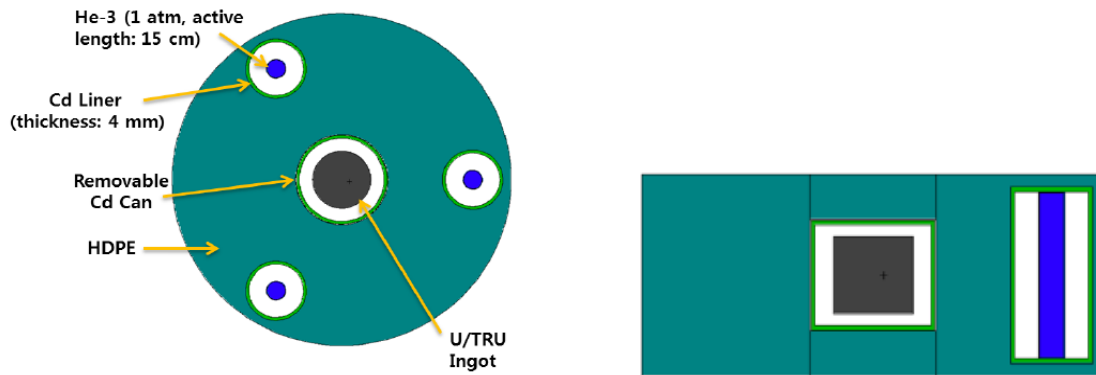


Figure 5 MCNPX Geometry of PNAR detector

The UNDA integrates three measurement techniques into one device: mass balance, gamma ray detection, and neutron detection. A schematic is shown in Figure 6 (Seo, 2012).

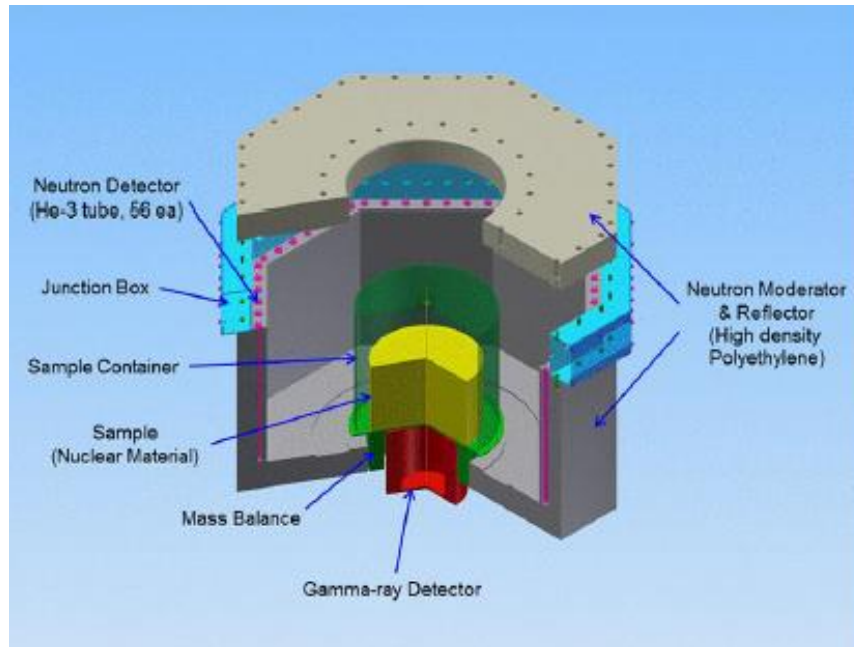


Figure 6 Schematic of UNDA device.

The UNDA detector's central cavity is 52 cm tall and 1 m in diameter in an octagonal shape. The mass balance is capable of measuring material up to 200 kg (Seo, 2012). The gamma ray detector is a NaI (TI) detector, which will provide basic energy spectrum data about the objects being assayed. The original purpose of this detector was determining enrichment in ^{235}U of depleted uranium samples. To broaden its applicability to a variety of materials, a high purity germanium (HPGe) detector would be better suited to provide more spectral resolution. The UNDA has 56 ^3He neutron detectors to provide the total neutron count for the items. The benefit of this device is gaining three measurements at the same time, while also being versatile enough to handle both the metal and salt materials.

4.2 Possible Assay Locations within a Facility

NDA device placement will vary depending upon on the facility design but will be

needed whenever material is allowed to leave either fuel preparation cell or processing cell. A key consideration for placement of devices should be the necessity for shielding from high radiation sources for each device's electronic systems. For this reason NDA placement and implementation should be considered early in the design process for any pyroprocessing facility. NDA equipment located within transfer locks will see a reduced radiation background compared to the main cell environment so these locations may be ideal for device longevity.

5. Pyroprocessing Safeguards Performance Model

The Pyroprocessing Safeguards Performance Model (PSPM)² is a discrete event model developed for testing of safeguard strategies performance. This model was constructed using SimEvents, a discrete event package used in the MATLAB/Simulink simulation environment. Within the software environment, blocks are connected together to perform specific tasks that could be represented as code segments in a traditional computer coding language such as C++ or Java. The simulation is executed for each time step as specified in the simulation configuration. A discrete event model is used for representing a discrete event rather than a repeating time dependent waveform (i.e. sine function); the pyroprocess is a hybrid discrete-continuous process so this modeling environment was chosen because it is capable of incorporating both aspects into a single representative model.

5.1 Model Description

The PSPM is initialized by the *SNF_source.m* file (see Appendix A.1), which reads the elemental masses of all nuclides present with the reference fuel assembly greater than one gram created using ORIGEN³. The elements are then grouped based upon their chemical behavior throughout the pyroprocess into six groups: actinides, cladding, hardware,

² The PSPM is designed following guidance from ANL expert analysis provided to the author; none of the process details are original to the author. Only the Simulink model based on this guidance is the author's own design.

³Radiation Safety Information Computation Center, ORIGEN2, Version 2.1, Oak Ridge, TN (1991).

ORIGEN is a radiation decay code that can track the decay and production of over 2000 isotopes.

active metals, rare earth metals, and noble metals (Till, 2011). The actinide group is important throughout the process; therefore, each element is assigned its own variable within the PSPM to be tracked. The actinide group contains the elements uranium, neptunium, plutonium, americium, and curium. The cladding & hardware group contains the activation products from the assembly, zirconium, niobium, molybdenum, and selenium. The active metal group contains rubidium, cesium, strontium, barium, samarium, and europium. The rare earth group contains yttrium, lanthanum, cerium, praseodymium, neodymium, promethium, gadolinium, terbium, and dysprosium. Finally the noble metal group contains zirconium and molybdenum (not from activation), technetium, ruthenium, rhodium, palladium, silver, cadmium, indium, tin, antimony, and tellurium. Each element's mass within a group is summed to provide the total mass of the group, which is tracked throughout the PSPM; this applies to all groups except the actinides where each element is tracked individually. It is assumed that the percentage each element, and isotope, contributes to the total mass before processing will be the same after processing. In addition to the above mentioned groups, the bulk salt (LiCl-KCl) mass is also tracked.

Two files can be used to execute the PSPM, *simevent_processflow_rs.slx* or *simevent_processflow_nors.slx*; the *rs* and *nors* in the file name indicate if rejection sampling (see discussion in Section 7.2) is used or not. Both files are identical except for the rejection sampling feature. The model run time is set within the model window of MATLAB, shown in Figure 7, and can be executed once by using the green play button; circled in red.

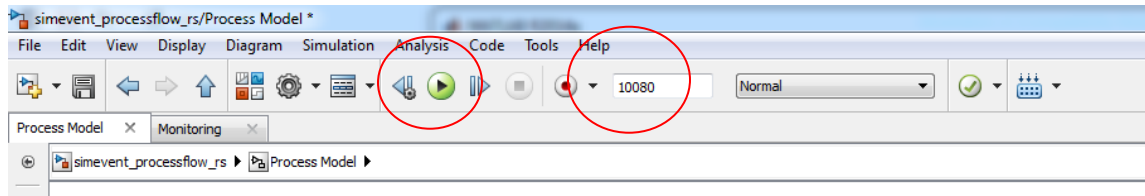


Figure 7 Simulink model run screen

If multiple executions are desired, then *multiruns.m* (see Appendix A.2) can be executed with the *batch* variable set to the number of desired realizations. *SNF_source.m* initialization file must be executed before *multiruns.m*. Each second of model time represents an hour of processing time, i.e. a 720 sec simulation time corresponds to 720 hours (or 30 days) of processing; simulation time does not correspond to clock time for the model to run.

The PSPM is broken up into two major subsystems, process and monitor, each with more complex operations. Figure 8 shows the *process* subsystem; each block shown in this subsystem is itself a subsystem comprised of more complex operations.

The process subsystem begins with the creation of a new entity (representing an assembly entering the air cell) once every 24 hours and assigns it attributes⁴ of the mean masses for the six groups (each actinide is treated as its own attribute) to be propagated throughout the model. The model then assumes only 17 entities are created within a 30 day period. This restriction arises from the assumption that the reference facility will process 200 assemblies per year. A common point of confusion with processing facilities is describing the capacity of the facility, since the facility has a nameplate capacity of 100 MTiHM/year but it is unclear if that number refers to output product or the input product. This work will assume this nameplate capacity refers to the input capacity and a corresponding number of accepted assemblies. A typical PWR assembly is assumed to contain 500 kg of heavy metal (Duderstadt, 1976) so a total of 200 assemblies are input into the facility. The assumed assembly for the reference facility is shown in Figure 9.

⁴ SimEvents terminology for values passes with an entity.

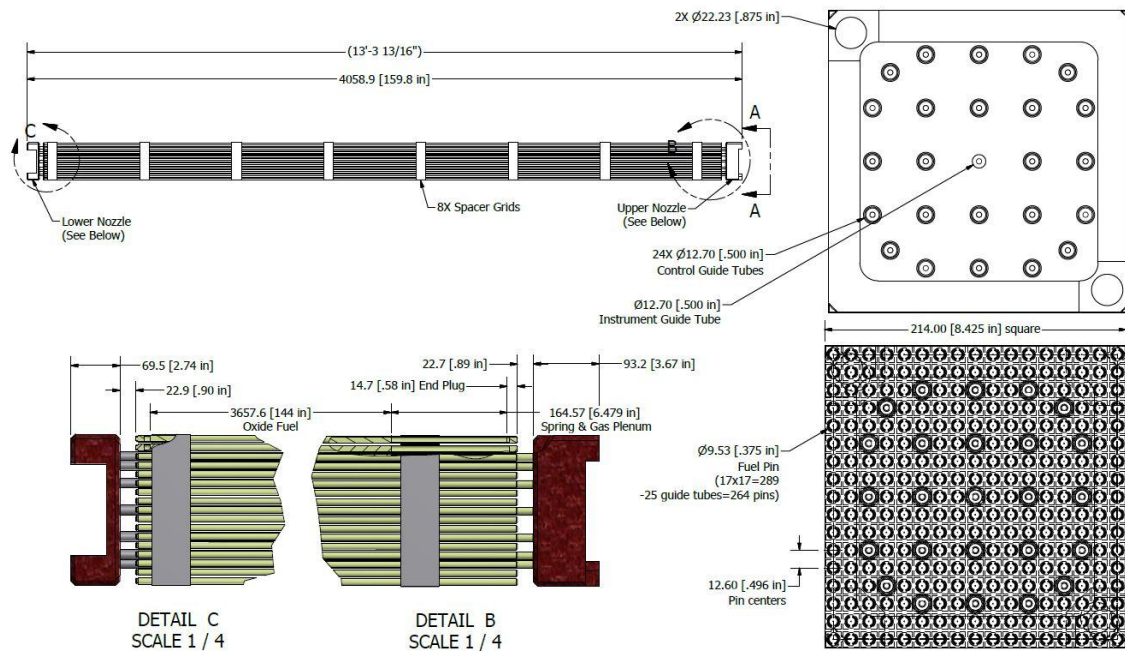


Figure 9 Westinghouse 17 X 17 PWR assembly

The process has bottlenecks due to varying processing times for each unit, thus storage must be accommodated for items until processing is available. The 17 entities created within a 30-day balance period create a constant input for each balance period over a one-year campaign. However, in a real facility, assemblies enter the facility as long as storage within the cell is available so as to not shutdown the entire facility if maintenance is required on equipment related to transfer into the cell.

Once the entity is created, it is passed to an NDA delay block, which represents the time delay for the input accountancy measurement and associated movements to storage before processing. The measurement is given an input uncertainty, which represents the combination of both random and systematic errors, and the mean value for each of the nine groups. A random number is generated from the normal distribution using the built in *randn* function in MATLAB with a mean of zero and standard deviation

is variable from the initialization, with this random value then multiplied by the mean and the result added to the mean value. This process is then repeated for each of the nine input variables. The resulting measurement is representative of a random variation based upon the device's uncertainty, which can then be varied for sensitivity analysis in *SNF_source.m*. The simulated measurement signal is passed to the plant monitoring subsystem to be used in the ID calculation, while the mean value is passed to SNF storage.

The storage subsystems contain a first in first out queue, which orders entities as they enter the block and releases the oldest entity when the next block is available.

Within the fuel disassembly step, the fuel assembly hardware is removed, followed by fuel pin cladding, while the fuel pellets are loaded into oxide transfer cans (OTCs). The process is estimated to take 4.8 hours. Each OTC, containing now four groups (after cladding and hardware are removed), has a capacity of 123 fuel pins or half of the initial input mass of the assembly. The OTC is then moved to the transfer lock between the air and processing cell. A mock up of the transfer lock is shown in Figure 10, showing an OTC transfer to the processing cell (right) (Benedict).

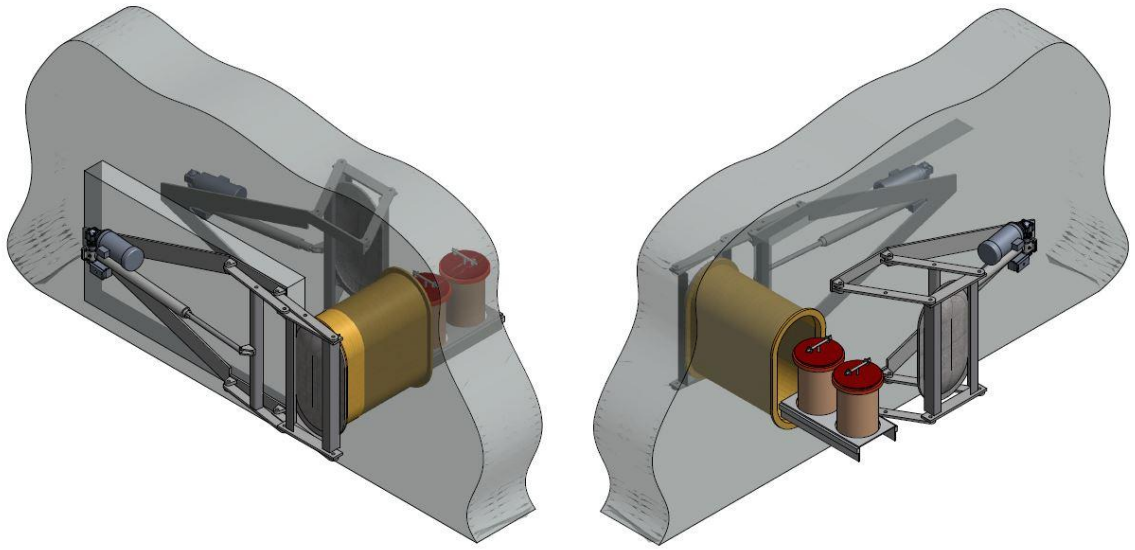


Figure 10 Transfer lock mock-up

Once two OTC are available at the lock they are transferred to the basket-loading unit. The cladding hulls are separated with the assumption that 99.9% of the fuel is removed in the decladding process (KAERI, 2013); these are compacted and transferred to the waste treatment cell for further processing. The pin-slitting device is shown in Figure 11, with the collection chute to the left of and the OTC underneath the slitting device (yellow box) (KAERI, 2013).

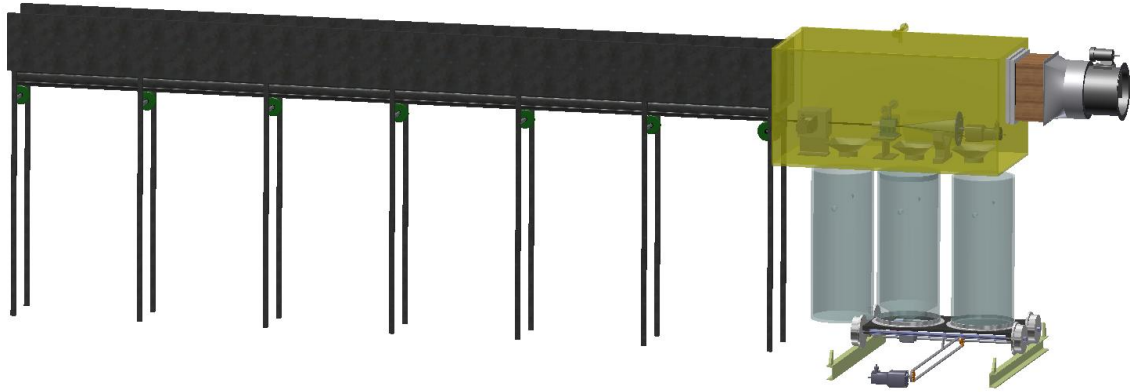


Figure 11 Pin Slitter Design by KAERI

A separation ratio of 0.999 is assumed and streams are split with the residual in the cladding. The PSPM representation of the pin slitting operation is shown in Figure 12.

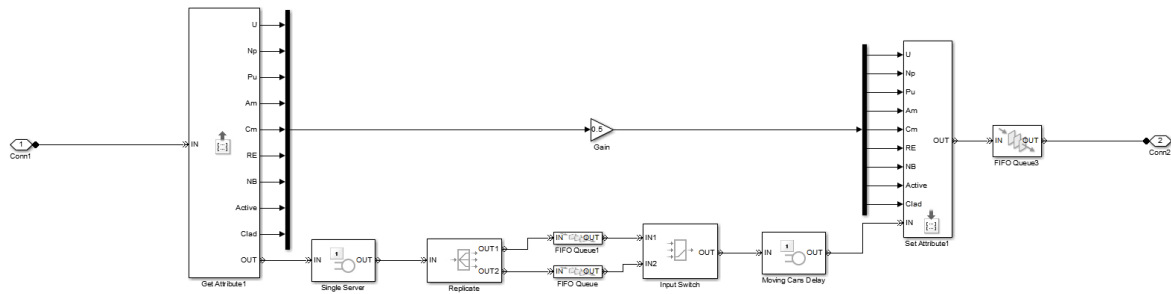


Figure 12 Pin Splitter subsystem of PSPM

The PSPM assumes a four-hour time delay between transfers from the decladding unit to the basket loading and uses an N-server with two servers. The N-server block allows N entities to be in the time delay and forces all other entities to wait until it has a space for new entities. This allows for two entities, in this case, to be passed to the next unit after the four hour processing time. Once the OTCs arrive at the basket module loader, two OTCs are emptied into the empty basket module (BM). The processing units further along in the process (electroreducer, ER, and metal processor) are designed to accept a

BM until it is emptied after the metal processing unit finishes and refilled with more oxide fuel. Fresh salt is also added to replenish the electroreducer at this point. This process is modeled with the batching subsystem, which combines two sequential entities by summing the masses of each of the groups associated with the entity and then outputting a new entity once two OTCs have arrived; see Figure 13.

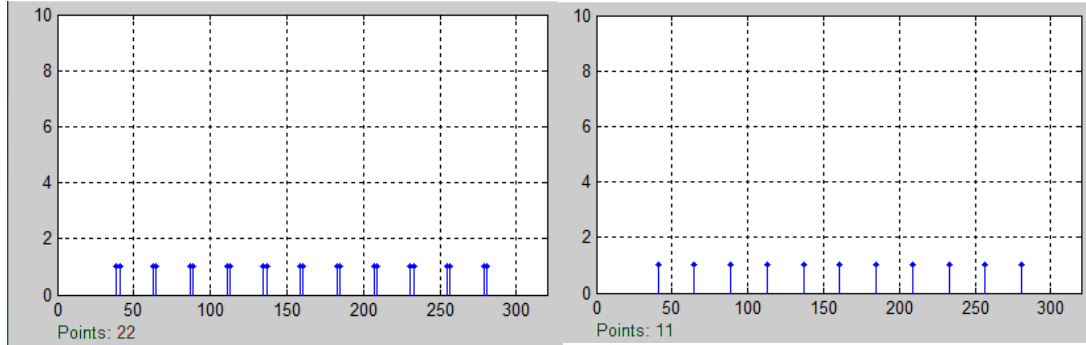


Figure 13 Entities before (left) and after (right) the batching block. The number of entities is plotted vs. time.

Once the BM has been loaded, it is then sent to the accumulate subsystem which groups two BMs to be sent to the electroreducer. The accumulate subsystem is similar to the batching, though a new entity is not created; instead, two entities are passed simultaneously, each with all the attributes of a single BM, once two entities enter the subsystem; see Figure 14.

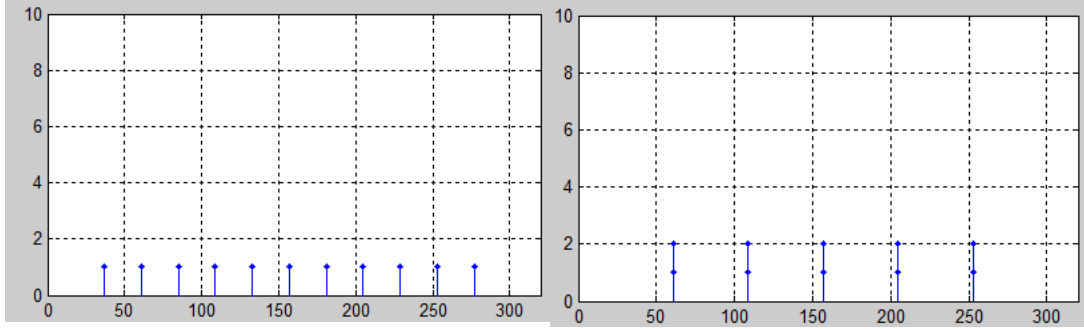


Figure 14 Entities before (left) and after (right) the accumulate block. The number of entities is plotted vs. time.

The set of BMs are then stored and processed once the electroreducer is available. The slitter has much greater throughput capacity than the BMs processed through the electroreducer and electrorefiner. Although other ways could be used to address this issue to minimize storage requirements, the PSPM allowed for up to seven BMs in storage but realistically the inventory could be minimized by having only inventory needed for expected maintenance; which may be only two to four BMs. This storage does not affect the safeguards approach but should be set by operational considerations.

The electrolytic reduction unit accepts two full BMs, which are processed for 72 hours and transferred to the ER. The BM processing, however, is offset about 36 hours so that the average output from the electroreducer is one BM every 36 hours. The offset allows for the unit to be continuously processing at any time. The PSPM also models this behavior using an output switch with two identical processing paths; however, the lower path has an offset delay of 36 hours so the resulting output from this subsystem is a reduced BM every 36 hours. The *Ereducer* subsystem is shown in Figure 15 with the lower path circled in red.

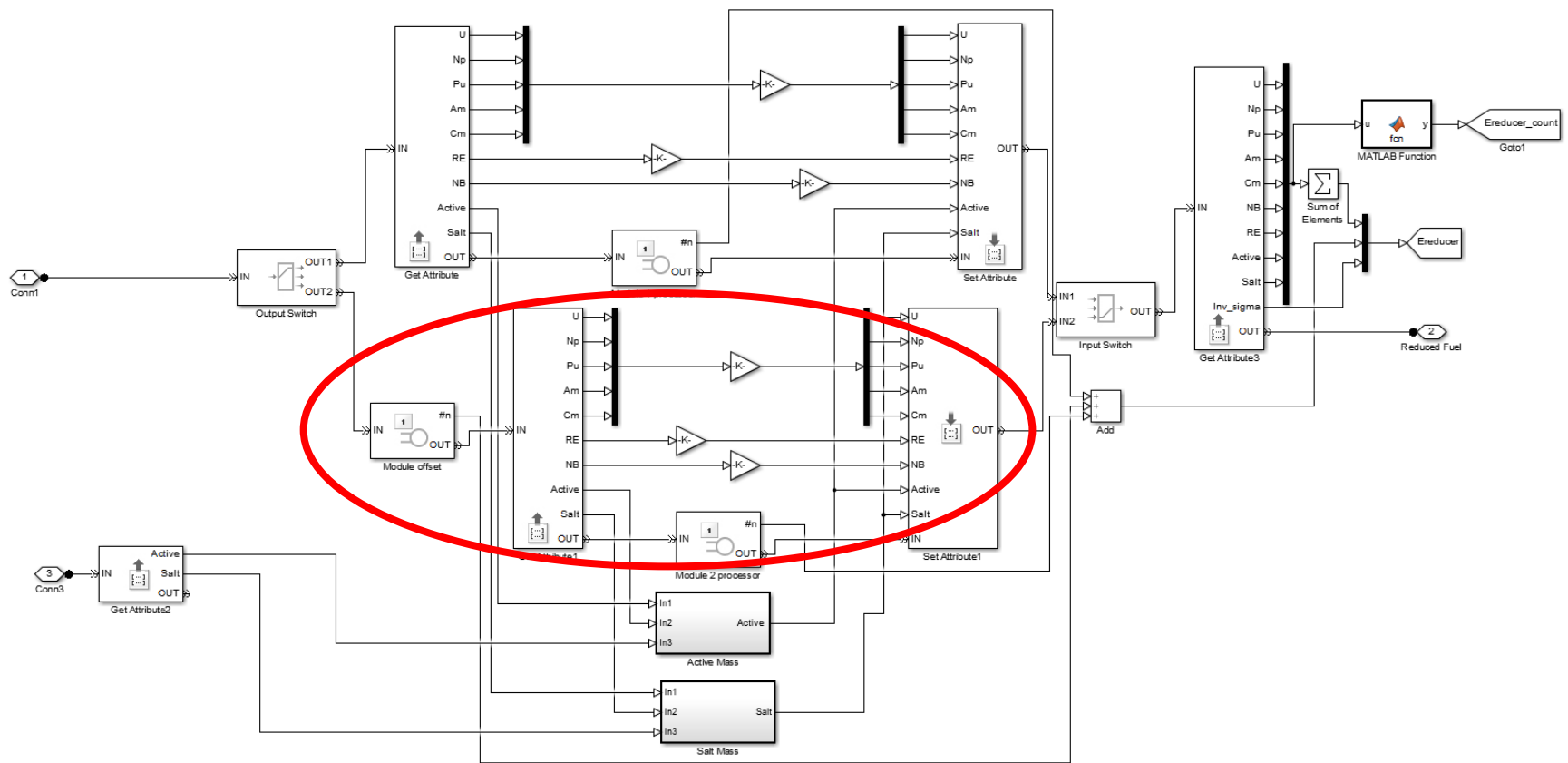


Figure 15 Ereducer subsystem of PSPM

The PSPM calculates the change in mass from oxide to metal in this process, using ^{235}U for example,

$$\text{mass of } U_{\text{metal}}^{235} = \frac{A_{U235}}{A_{U235} + (2 * A_O)} \text{mass of } U_{\text{oxide}}^{235} \quad (8)$$

Where A_{U235} and A_O are the atomic masses of ^{235}U and oxygen respectively. After each third BM, some recycled salt containing active metals is added to the vessel.

Once reduced to metallic form, the BM is now sent to the heart of the pyroprocessing, the ER, where the actinides, rare earths, and active metals will dissolve into the salt; the noble metals will remain within the BM. The ER, just as the electroreducer, accepts two BM and processes for 36 hours; however, only one BM is inserted each 36 hours. The ER has four different products it creates throughout processing. The majority of material is output in the uranium product (U product), followed by the uranium and transuranic product (U/TRU product), metal waste, and a portion of the salt is removed for treatment of fission product buildup. The Uranium within the BM when added to the salt forms the least stable chloride compound as compared to the transuranics, rare earths, and active metals; however, if a voltage is applied across a cathode rod and anode (the BM), uranium and salt can be electro-transported and deposits onto a cathode rod. The uranium salt dendrite produced is ~20 weight percent salt. This is a semi-continuous process so cathode rods are periodically withdrawn from the ER and sent to the U processor. The PSPM models this processing as six discrete additions to the U processor every six hours, using an output switch with

six outputs and six individual delay blocks with delays increasing by six hours. The U/TRU dendrite contains the transuranic elements, some uranium, ~0.1% rare earth metal, and salt collects on a solid cathode. The U to TRU ratio deposits as 1:2.4 (Benedict). A U/TRU cathode is produced after 36 hours with the same weight percent salt as the U product; this is in turn sent to the TRU processor. The noble metals do not dissolve so the BM after processing is transferred to the metal waste-processing unit. A continuous salt recycle process is needed independent to batch processing to maintain a desired oxidant (salt high in UCl_3) concentration and salt height within the vessel. To approximate this process, salt containing fission products and actinides is withdrawn with each BM and stored in a salt collection tank, while oxidant salt is added after every third BM is processed. A key feature of the ER is once the fuel in a BM is dissolved, it can no longer be associated with a specific input assembly and is mixed in with the other constituents of the ER processing vessel; up until this point the fuel pellets could be traced back to a specific input assembly. The PSPM uniquely captures this feature of the process as well in the ER mass separation subsystem. Each group's mass is treated separately within this subsystem, in essence as a large pot with the additions from the recycle loop and from the beginning processing of the BM; removals are only triggered by the insertion of a BM. The working inventory of the ER is not zero. A large actinide inventory (on the order of a 100 kg at equilibrium) is needed before extraction of U and U/TRU cathodes occurs. The PSPM, however, is modeled assuming this equilibrium has been reached; therefore changes in this working inventory occur due to intentional diversion (decreases in inventory) or residual buildup due to incomplete processing (gains to inventory). The PSPM's increase in working inventory after one year of simulated

operation is 4.34 kg of Pu.

The U product from the ER is emptied into the U processing vessel as six separate additions per BM; the vessel has a capacity of one BM of material. Once full, the U product is heated in an induction furnace to boil off the adhering salt and produce a homogenous uranium product in 25 hours. The adhering salt, as it is boiled off, is piped via heated salt lines to the salt collection tank. While the majority of the U product is transferred out of the cell to storage, ~15 kg per BM is collected to be used in the production of the oxidant recycled salt returned to the ER. The PSPM also passes the number of entities in the U processor as well as the total mass within the vessel to the plant monitoring system.

The U/TRU product must also have the adhering salts removed before a final product is created. This is accomplished in a smaller device similar to the U processor. One U/TRU ingot is produced from each BM processed in the ER, with a weight of 8 kg. A sample for destructive analysis is taken from each ingot to determine isotopic composition. The U/TRU ingot is then moved through its own dedicated, one-way transfer lock to the storage vault (200 transfers per year). The U/TRU ingot is the material most attractive to a potential adversary and needs to be the focus of the facility security and safeguard systems. Additional information about the mass of material transfers and the number of entities in processing is passed to the monitor system.

The noble metals remaining in the BM after electrorefining are added to the metal waste processor vessel. The vessel is full after ten BMs are processed in the ER and processed for 25 hours. After processing is completed, the metal waste is transferred to the waste treatment cell while the distilled salt is added to the salt collection tank. A full

cycle from when the first noble metals are added to the formation of the metal waste form takes 390 hours (~16 days). This process is modeled similar to the TRU processing system with the batching subsystem to collect 10 BM then a delay is represent the processing time and the resulting composition of the metal waste is then passed to the monitoring system.

The salt collection tank accepts the salt from the ER, U processor, TRU processor and the Metal processor, and once the total salt mass reaches 327 kg (after three BM are processed) the tank empties into the drawdown vessel for further treatment of the salt.

This is represented in the PSPM with the salt mass of each of the inputs summed together and fed to an if-else system to test if the mass is greater than 327 kg. If true, then the salt mass is decreased by 327 kg and fed back as residual; if false, the mass is fed back as residual. A second subsystem is used to sum the mass of the other groups (other than the salt) and after three batches passes the values to the new salt entity representing the salt moving to the drawdown vessel. No explicit delay blocks are associated with this system; however, the batching block acts as a delay as it requires three BMs to collect before it releases a new entity. Figure 16 shows the implementation of the *Salt Collection Tank* block.

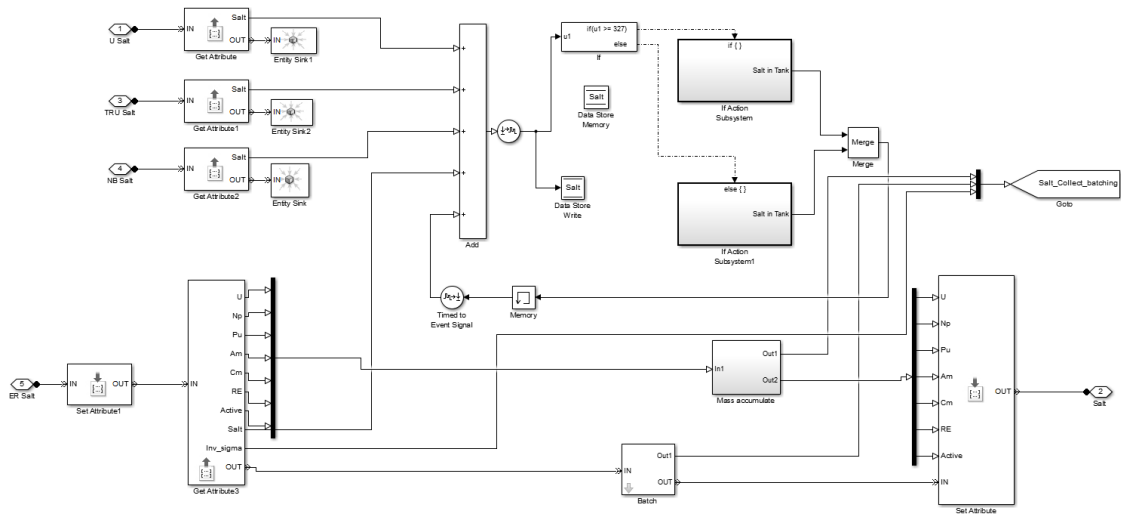


Figure 16 Salt Collection Tank block

The first process performed on the salt is actinide drawdown, which draws out the actinides and a small fraction of rare earths (as chlorides) and collects them on cathodes in a metal form via electrolysis for 20 hours. While most actinides present in the ER recycle salt will collect on the cathode, a small fraction of the curium and americium will not and will stay in the salt. The actinide free salt is further processed via rare earth drawdown, another electrolysis process to reduce the rare earths, curium, and americium to metal waste form with some salt and active metal also. The majority of the rare earth free salt is transferred to the final recycle operation, while ~23% remains in the drawdown vessel for production of the UCl_3 needed in electrorefining, known as oxidant production. The oxidant production process uses the U metal set aside after U processing as well as the actinide cathodes created during actinide drawdown. Oxidant production bubbles chlorine gas across the metal surfaces and after 15 hours produces the oxidant product to be reinserted into the ER. This entire three-process drawdown takes on average 49 hours to accomplish. The PSPM models each process as a separate subsystem.

Figure 17 shows the recycle operations modeled in the PSPM.

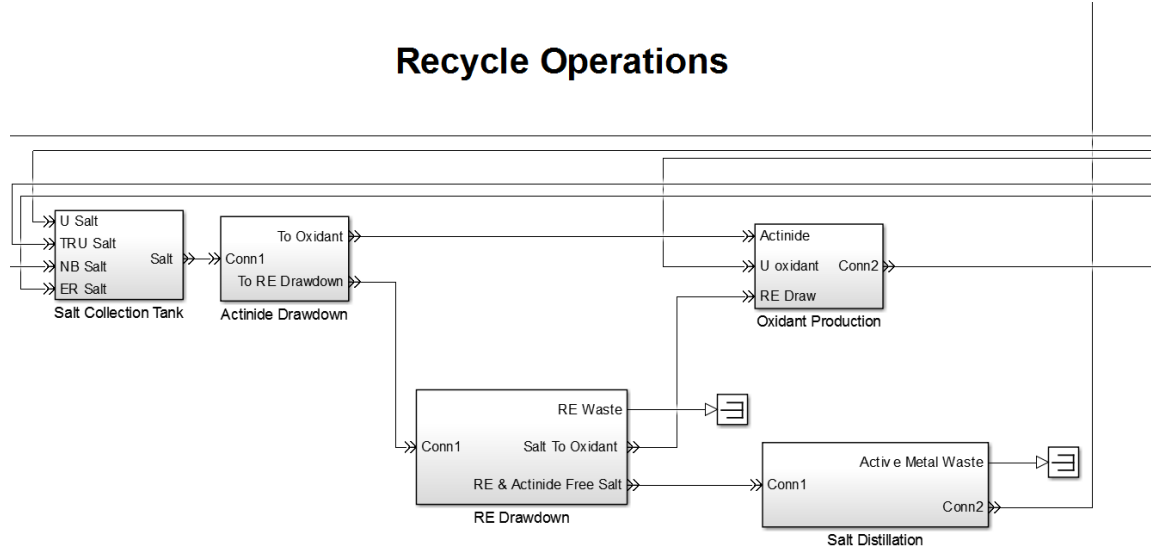


Figure 17 Recycle Operations blocks

Each subsystems has the same layout: entities are passed into the system where information on mass of the salt and constituents transferred, as well as the number of entities being processed to the monitoring system; once sufficiently delayed the groups are separated using ratios described in the process flow (Benedict).

The final recycle process removes a concentration of active metals from the rare earth free salt sent to the salt storage tank. The salt storage tank holds two rare earth drawdown cycles worth of salt before salt crystallization is performed. The salt crystallization process creates a concentrated active metal waste form containing about one third of the active metals contained in the rare earth free salt over the course of 49 hours. Once processing is completed, the remaining salt is transferred back into the electroreducer. The active metal waste is combined with the rare earth waste form as a ceramic waste before being transferred to the waste treatment cell. A complete salt

treatment cycle, including the three drawdown processes and salt crystallization takes about eight days. The PSPM again uses the batching subsystem to accumulate the two salt batches from rare earth drawdown before processing can occur; however, in process inventory is not assumed to be needed, as no SNM (U or Pu) is present in this process. If SNM is detected in the active metal waste form, it would warrant an alarm to trigger for off normal process operation. The final separations are again based upon the process flow described by ANL, creating an active metal waste form as well as salt re-entrant to the electroreducer.

The last elements of the process model subsystem are the various material detection and diversion blocks. Each divert block consists of an embedded MATLAB function block with a function script like that in Table 1.

```
function y = fcn(u)
global simtime
if simtime< 7600 %Start of diversion scenario
y = u;
elseif simtime>7650 %End of diversion scenario
y=u;
else
y=u*0.6; %Amount of material diverted
end
```

Table 1 Divert material MATLAB code

The user is able to modify the times for starting (the *if* statement) and ending (the *elseif* statement) of the diversion scenario and the amount of material to divert is set in the *else* statement depending on the scenario of interest (i.e. an abrupt diversion might assume all the material is gone so $u*0$ or a protracted diversion over a longer period of time could be $u*0.7$). If no diversion is desired, then the subsystem can be commented through so signals pass through as though no block was there.

The sensor subsystems also contain an embedded MATLAB function to produce

the randomized signal expected from any detection device used to provide safeguard information. The script written for each sensor block performs the same function; however, each sensor has a different initial signal composition so the array locations differ between the sensors. Each of the material signals has the same basic structure as seen in equation 9.

$$[\text{Mass of group}_i (i=1 \dots n), \sigma \text{ of detection device}, \sigma \text{ of inventory device}] \quad (9)$$

The detection device is assumed to provide normally distributed values so the built in MATLAB *randn* generator is used. The *randn* generator uses the MATLAB Mersenne Twister algorithm to produce normally distributed random numbers. The mass contained in the *u* input array is assumed to be the mean value; therefore the random normally distributed value can be calculated for each element within the array as shown in Table 2.

```
for i=1:length(u)
    mass(i)=u(i)+ sigma(1)*randn;
end
```

Table 2 MATLAB code for random signal generation.

Within each iteration of the *for* loop the *randn* function returns a new normally distributed random number; *sigma (1)*, refers to the device measurement uncertainty. A caveat of the Simulink environment requires the seed of random number generator to be manually seeded. This occurs because each embedded MATLAB function operates in its own “function space” which means that each call of the *randn* function in independent MATLAB functions (those not located within the same subsystem) will begin with the same seed, consequently the same random number sequence for each sensor introduces a correlation amongst measurements and randomness is no longer guaranteed. However, this is easily solved by seeding each generator differently. The PSPM adds a specific

number to the seed provided from the initialization file to ensure each generator is seeded differently. Seed choice does not need to be random itself as this only picks the starting place of the generator in the sequence. As an example, two different seeds of one and two produce two completely random sequences with no correlation between each other.

The second major system is the monitoring subsystem, which calculates the state of the plant at the end of each inventory period chosen in the initialization file.

Figure 18 shows the monitoring system implemented in the PSPM. The monitor system receives 17 signals from the various processes and input and output streams within the process subsystem. Samples are taken at the end of each inventory period and then the data is passed to an embedded MATLAB function to calculate the ID, SEID, and perform Page's test, described in Section 5.4 for diversion testing.

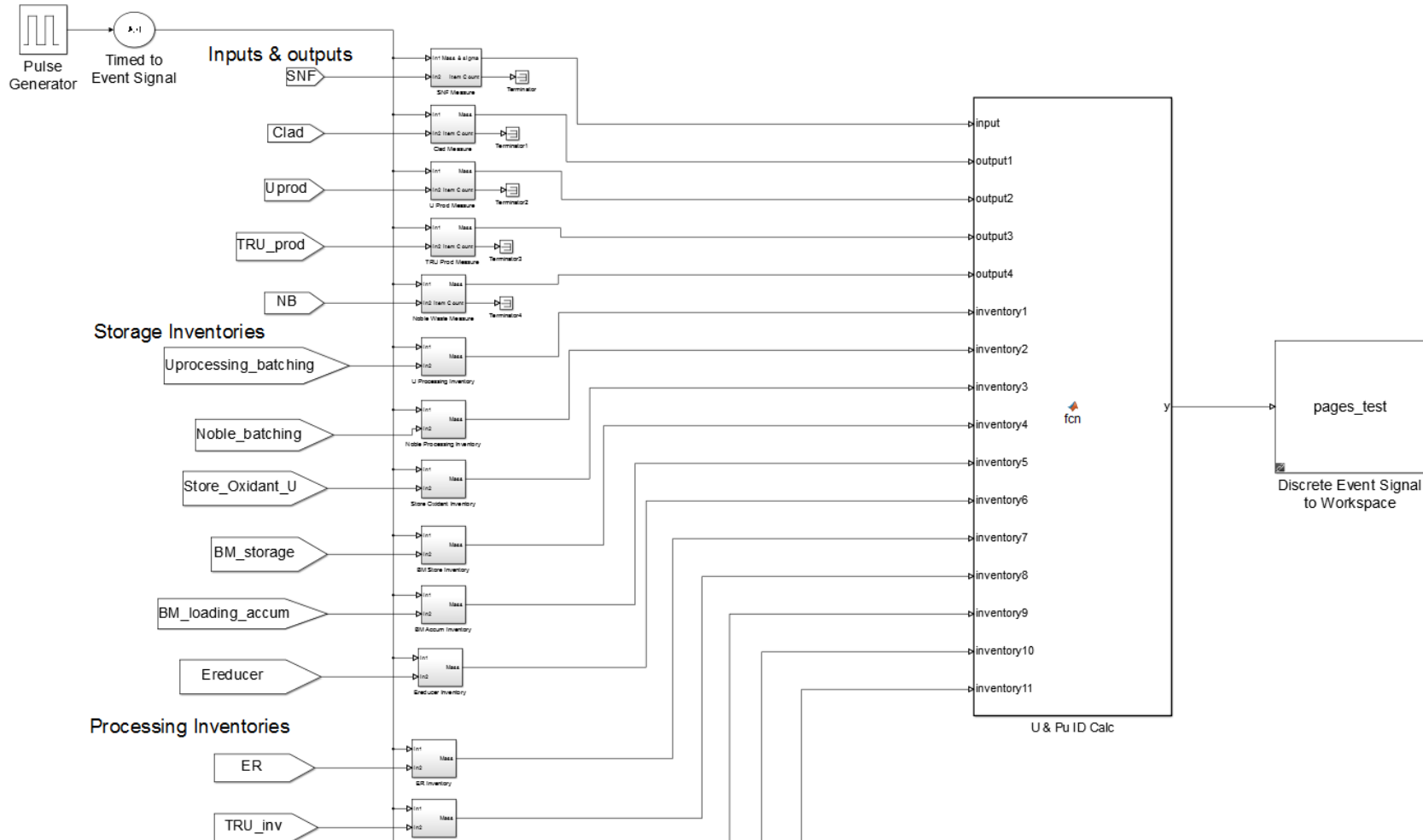


Figure 18 Monitoring system in Simulink

For the SNF input and each of the four products containing SNM a signal must be summed for each addition to or subtraction from the MBA, this is accomplished using an embedded MATLAB function in Table 3.

```
function [y,count] = fcn(signal,sigma)
global simtime InvPeriod
persistent cumsum c n
if isempty(cumsum) % Intialize variables
    cumsum=zeros(9,1);
    c=0;
    n=1;
end
t=(simtime+312)/(n*InvPeriod); % Determine if the next entity occurs
if t>=1 % after the next balance period
    cumsum=cumsum+signal; % If yes then output the total vector
    c=c+1; % cumsum
    y=[cumsum;sigma];
    count=c;
    n=n+1;
    c=0;
    cumsum=zeros(9,1);
else % If no then continue to accumulate
    c=c+1; % and output a vector of zeros
    cumsum=cumsum+signal;
    y=zeros(10,1);
    count=0;
end
```

Table 3 MATLAB code to output the total SNF transfers at the end of a balance period

The *sigma* vector contains both the device uncertainty as well as the inventory device uncertainty, though some strategies will not require knowledge of both of the inventory device uncertainty. The timing of each of the different measurements differs significantly as only one or two metal waste forms will be created in a 30-day period, while 34 cladding measurements will be taken in the same time span. The time from the last measurement in a balance period to the actual end of the balance period must be estimated (in the code provided in Table 3 this is 312 in the expression for *t*) so the final sum can be output before the next balance period begins. The same approach is used for

the SNF input, cladding waste, U product, U/TRU product, and metal waste form. Though the input vectors for each will vary depending on the number of materials contained within. An item count is also output to verify the number of processed items matches the number of measured items, though this is not used by the PSPM at this time.

The remaining 12 signals are storage or processing vessel inventories. These do not require summation over the balance period, instead the value of the signal at the end of balance period must be the only value passed and zero otherwise. To ensure all of these measurements are simultaneously passed to the ID calculation block a signal latch and pulse generator with a period set by the inventory period in the initialization file is used to write out the values only. An example of the SNF signal before the latch (top) and after (bottom) is shown in Figure 19. The mass (kg) is plotted against the simulation time (hr). Comparing the two graphs the random nature of the SNF mass is seen in the top graph while the integrated single mass passed at the end of an inventory period is observed in the bottom graph.

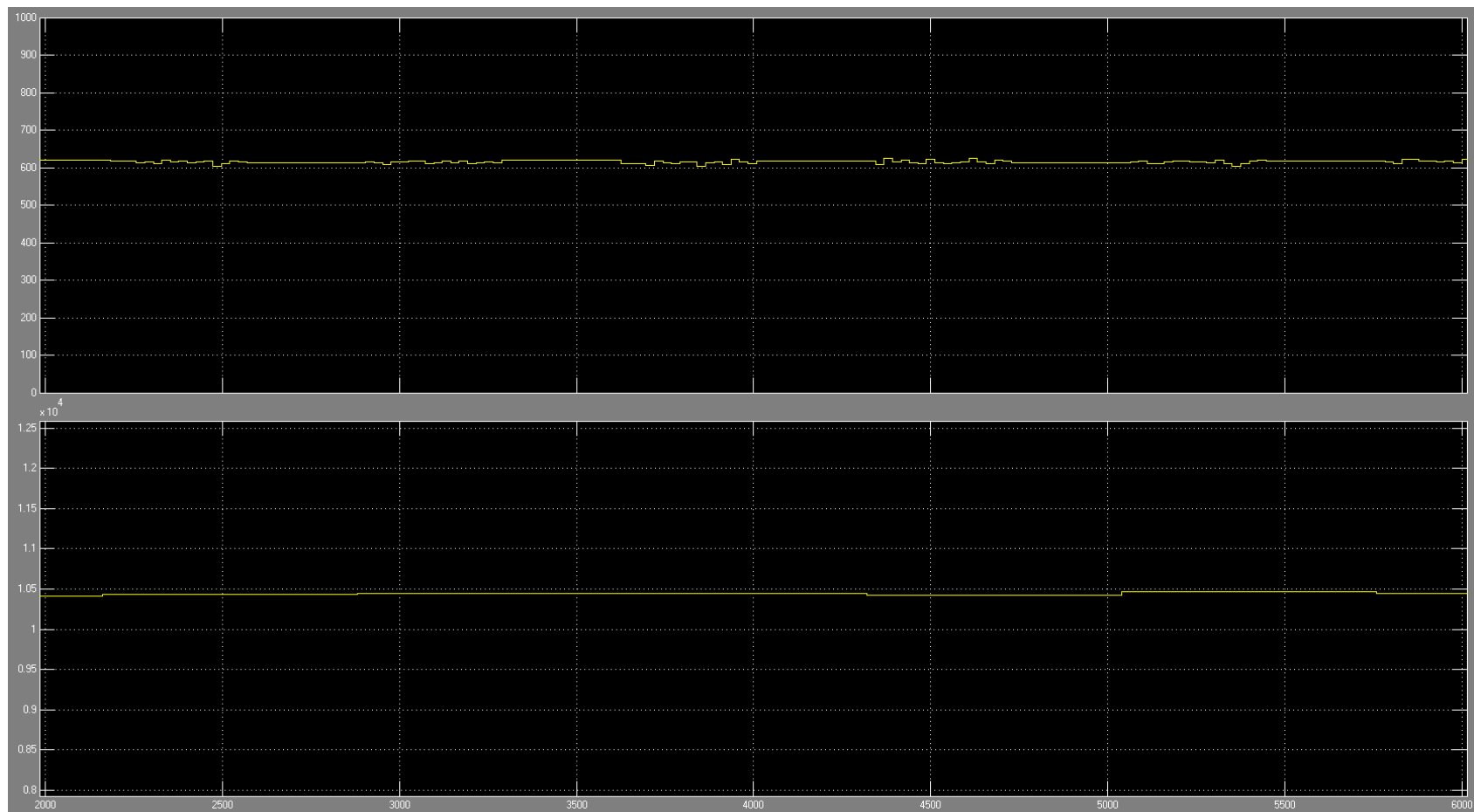


Figure 19 Signal before signal latch (top) and after (bottom). Mass (kg) is plotted vs time (hr)

Both the storage and vessel inventory signals are structured as a mean mass of the item (no elemental breakdown for each group), the number of entities contained in storage or being processed, and the inventory device's uncertainty. Even though each strategy tested did not use all 12 inventory signals in the ID determination, the data are still passed for each balance period to facilitate easy testing of various strategies with minimal model changes.

An embedded MATLAB function calculates the ID for both U and Pu. The full code can be viewed in Appendix A.3. A vector *id* is created to store the ID, inventory, σ^2_{inv} and SEID for both U and Pu. Within the process subsystem the chemical compositions of the fuel change from oxides to chlorides and metals during the various processes. The mass data passed to the monitor system is based on chemical composition of these processes therefore all signals are converted to represent the amount of metal present to provide a common comparison point. Four different strategies are tested in this work, known as levels, with each using different signals. These strategies are described in detail in Section 7.2. Once the *id* vector has been calculated then the SITMUF series is created and Page's test is performed as described in Section 5.4. Finally the output back to the model is defined.

5.2 Model Assumptions

Previous research by Cipiti has shown the utility of a Simulink model for testing a safeguard strategy's effectiveness (Cipiti B. e., 2012). A key assumption of this work was that the processing could be done within a common 24-hour period. However, under guidance of ANL pyroprocessing experts, the 24-hour balance period was not used for

this work; instead a 30-day balance period was chosen. Table 4 summarizes the processing times assumed for each process and number of units processed each year.

Process	Time (hr)	# of processed units/yr
Fuel Disassembly	4.8	200 Assemblies
Electrolytic Reduction	72	200 BM
Electrorefining	72	200 BM
U Processing	25	200 BM
TRU Processing	29	200 BM
Noble Metal Processing	25	20 Metal Waste
Actinide Drawdown	20	67 Drawdown Cycles
Rare Earth Drawdown	14	67 Drawdown Cycles
Oxidant Production	15	67 Drawdown Cycles
Salt Distillation	49	34 Active Metal Waste

Table 4 PSPM processing times and units processed in a year

An additional four-hour delay to allow for material transfers from one unit to the next is included along with the processing times listed above.

To create a more representative model of plant operations, a discrete event model was adapted. A discrete event simulation passes entities from one block to another, once the block has performed its function the information is passed to the next in discrete packets. The model only solves when entities are passed between blocks and not for elapsed time between events in between. This is ideal for modeling batch operations, such as pyroprocessing.

While the majority of operations within the pyroprocess are discrete operations, two are not. These two continuous operations are the uranium extraction from the electrorefiner (ER) and the salt withdrawal from the ER. The uranium is continuously

deposited on cathode rods during the processing of a basket module in the electrorefiner, with this dendrite material collected at the Uranium processor until enough material for a processor batch is collected. The PSPM assumes this continuous process can be approximated as six discrete deposits of uranium six hours apart at the Uranium processor. The second continuous operation is the salt treatment from the ER. Salt from the ER must be cleaned to remove fission products, maintain a salt height within the vessel, and replenish the UCl_3 concentration within the ER to maintain the electro-transport process. Detailed analysis to understand the timing of these removals and insertions from the ER were not available at the time of this work so it was assumed the salt recycle operations would occur after three basket modules were processed.

5.3 Model Verification

A simple example is used to verify model results are consistent with the process flow sheet. The example to be analyzed is the single Pu input flow into the MBA (SNF assemblies) and two output flows from the MBA (Cladding & Hardware waste and TRU product) for an inventory balance period. A single assembly contains 5.71 kg of Pu oxide (calculated from ORIGEN file). The Cladding & Hardware waste contains 0.01% of the initial amount of Pu in the assembly. The TRU ingot consists of 5.025 kg of Pu, as calculated using the process flow sheet. Over a 30-day inventory period, 17 assemblies will be input as well as Cladding & Hardware waste transfers. However, the number of TRU products processed per inventory period ranges from 15 to 16. The inventory period from 2160 to 2880 hours was chosen as the first period where the model represents equilibrium; 16 TRU products are transferred during this period. All materials must be

evaluated in the same chemical form so the oxide fuel is multiplied by 0.8814 (the theoretical conversion from oxide to pure metal for TRU; see equation 8. A summary of the calculated results for this inventory period is shown in Table 5. The results of the PSPM for this inventory period are presented along with the calculated results in Table 6.

Transfers	Pu Metal Mass (kg) per Assembly	Total Pu Metal Mass (kg) for Inventory Period
SNF input assembly	5.033	85.561
Cladding & Hardware Waste	0.0050	0.085
TRU Product	5.025	80.4

Table 5 Calculated Pu Metal mass for a Single Assembly and Inventory period

Transfers	Calculated Total Pu Mass (kg)	PSPM Total Pu Mass (kg)	C/E
SNF input assembly	85.561	85.5575	1.00004
Cladding & Hardware Waste	0.085	0.0856	.9930
TRU Product	80.4	80.4	1.0000

Table 6 Calculated Pu mass compared with PSPM Pu mass

Table 6 shows excellent agreement between model and calculated total Pu mass for the inventory period; the differences are between calculated and model values arise from rounding errors. This provides verification that the PSPM does perform as expected based upon the process flow sheet.

5.4 Page's Test

An accepted statistical test for material diversion is the Page's test (Cipiti B. e., 2012; Burr, 2013). The Page's test can be tuned to be sensitive to both abrupt and protracted diversion attempts, as well as processing anomalies. For these reasons, the test is included in the PSPM. An ID series is created such as:

$$\overrightarrow{ID} = [ID_1, ID_2, \dots ID_n] \quad n = 1, 2, 3 \dots n \quad (10)$$

Each ID_i in the ID series represents the inventory differences calculated every 30 days. An assumption of Page's test is that each ID_i is independent of previous terms within the series; this assumption is not valid for our application, however. ID as discussed in Section 2.1, uses the ending inventory of one balance period as the starting inventory of the next period. Another source of dependence is introduced as the same equipment is used to calculate ID each balance period. To account for these dependencies the ID series can be transformed into a standardized, independently transformed material unaccounted for (SITMUF) series. A variance/covariance matrix can be constructed:

$$\begin{bmatrix} V_{11} & C_{12} & C_{13} \\ C_{21} & V_{22} & C_{23} \\ C_{31} & C_{32} & V_{33} \end{bmatrix} \dots$$

Where V_{ii} and C_{ij} can be calculated by,

$$\begin{aligned} V_{ii} = & inv_i^2 (\sigma_{in,R}^2 + \sigma_{in,S}^2) + \sum_{k=1}^4 out_{i,k}^2 (\sigma_{out,R,k}^2 + \sigma_{out,S,k}^2) \\ & + \sum_{k=1}^{11} \{ inv_{i,k}^2 (\sigma_{inv,R,k}^2 + \sigma_{inv,S,k}^2) \\ & - inv_{i-1,k}^2 (\sigma_{inv,R,k}^2 + \sigma_{inv,S,k}^2) \} - 2 \sum_{k=1}^{11} inv_i inv_{i-1} \sigma_{inv,S,k}^2 \end{aligned} \quad (11)$$

$$\begin{aligned}
C_{ij} = & in_i in_j (\sigma_{in,R}^2 + \sigma_{in,S}^2) + \sum_{k=1}^4 out_{i,k} out_{j,k} (\sigma_{out,R,k}^2 + \sigma_{out,S,k}^2) \\
& + \sum_{k=1}^{11} \{ inv_{i,k} inv_{j,k} (\sigma_{inv,R,k}^2 + \sigma_{inv,S,k}^2) \\
& - inv_{i-1,k} inv_{j-1,k} (\sigma_{inv,R,k}^2 + \sigma_{inv,S,k}^2) \} \\
& - \sum_{k=1}^{11} inv_{i,k} inv_{j-1,k} (\sigma_{inv,S,k}^2 + \sigma_{inv,R,k}^2 [if\ j - i = 1]) \\
& - \sum_{k=1}^{11} inv_{i-1,k} inv_{j,k} (\sigma_{inv,S,k}^2 + \sigma_{inv,R,k}^2 [if\ i - j = 1])
\end{aligned} \tag{12}$$

In equations 11 and 12, *in* refers to the SNF input, *out* is one of the six outputs (U product, TRU product, Cladding waste, hardware waste, ceramic waste or metal waste), and *inv* refers to one of the 11 different inventory measurements taken throughout the process. The *i-1* or *j-1* subscripts refer to prior balance periods while the *R* and *S* subscripts are the random and systemic errors respectively. The random error is only applied in the last two terms of the covariance if the condition is true. The variance/covariance matrix is symmetric about the main diagonal. A lower triangular matrix, [T], and diagonal matrix, [U], can be constructed from the variance/covariance matrix, V.

$$[T][V][T]^T = [U]$$

$[T]^T$ is the transpose of the matrix [T]. An independent transformed ID series, I, can be expressed as:

$$[T] * \overrightarrow{ID} = \vec{I}$$

Finally SITMUF can be calculated as:

$$\overrightarrow{SITMUF} = \frac{\vec{I}^r}{[U]} \quad (13)$$

Once the SITMUF series has been constructed, Page's test can be implemented.

$$S_i^+ = \max (S_{i-1}^+ + SITMUF_i - k, 0) \quad (14)$$

An alarm is triggered when,

$$S_i^+ > h$$

Page's test uses pairs of h and k values to set alarm thresholds. The k variable determines the sensitivity of the test while h is the alarm threshold, with an infinite set of possible (h, k) values. Previous work has shown the utility in using a large k such as $k=2.8$ and $h=0$ to test for abrupt diversion (Cipiti B. e., 2012; Jones, 1988); a threshold can then be determined for protracted diversion based on the abrupt test. Similar results as those reported by Cipiti (Cipiti B. e., 2012) were shown to be effective for the PSPM, so $h=5$ and $k=0.3$ are used in the PSPM.

The calculation of the covariance matrix requires knowledge of both systemic and random variation in each measurement instrument, however many of the techniques suggested for NDA are yet to be implemented in the field, consequently data on these two quantities are unknown. Instead an overall variance for the measurements are assumed,

$$\sigma^2 = \sigma_R^2 + \sigma_S^2$$

The PSPM can be updated when these unknowns are better understood.

Implementing the matrix operations within the PSPM is taxing on model run time, so a simplification can be made. If all measurements are assumed to be independent within a balance period (i.e. different pieces of equipment), then the following modifications can be made to the above equations (Cipiti B. , 2011):

$$X_1 = ID_1 \quad (15)$$

$$X_i = a_i ID_{i-1} + ID_i, i = 2, 3, 4 \dots$$

The coefficient a depends on both the inventory and total ID variance as:

$$a_1 = 0$$

$$a_i = \frac{\sigma_{inv}^2}{\sigma_{ID}^2 - a_i \sigma_{inv}^2}, i = 1, 2, 3 \dots \quad (16)$$

The Page's test now becomes:

$$S_1^+ = X_1$$

$$S_i^+ = \max\{S_{i-1} + X_i a_i - k \sigma_{X,i-1}\}, i = 2, 3, 4 \dots \quad (17)$$

With,

$$\sigma_{X,i}^2 = \sigma_{ID,i}^2 - \frac{\sigma_{inv}^4}{\sigma_{X,i-1}^2}$$

The alarm condition is now:

$$S_i^+ > h \sigma_{X,i}$$

The simplified Page's test has been implemented in the PSPM for both diversion and non-diversion scenarios. Material diversion by convention is represented as a positive alarm signal.

6. Challenges to Conventional Safeguards Posed by Pyroprocessing

A pyroprocessing facility has many features within its process that pose a challenge to applying safeguards regulations as they stand today. These challenges are twofold; the regulations have been tailored for the only reprocessing technology deployed to date (aqueous) and the environment in which the processing occurs degrades most detection equipment quickly, requiring the need for instrumentation modification. The following section outlines the challenges posed by pyroprocessing along with its unique benefits.

6.1 Hot Cell Environment

Both the air and argon processing hot cells will have very high radiation backgrounds, which will degrade material quickly and disrupt electronic signals. Simulations performed by Pope calculated the dose rate a worker at FCF would receive without the window shielding intact as 23 Gy/hr (Pope C. e., 2009). This analysis can be used to estimate the radiation field within the cell assuming a $1/R^2$ relationship. Figure 20 is an extrapolated plot from the radiation source (the ER). Labels on the graph provide reference distances of the cell wall and operator corridor from the ER, however the analysis does not account for any shielding in place.

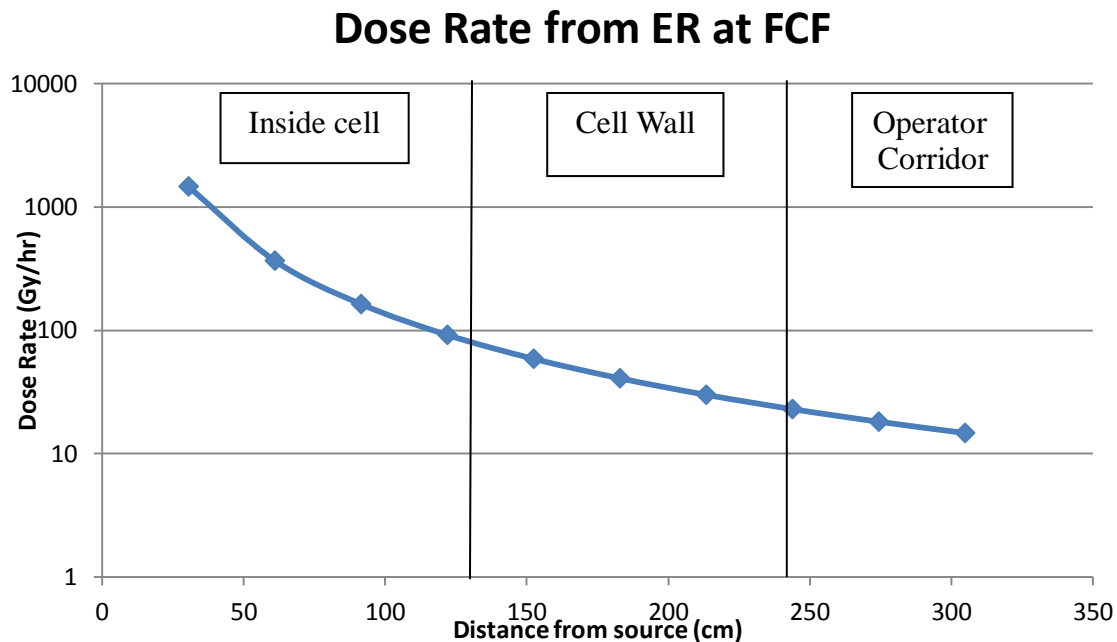


Figure 20 Dose Rate from ER as a function of distance.

It is reasonable to assume that the reference facility will experience in cell fields greater than those of FCF due to its factor of ten greater throughput. Electronic signals in radiation fields this large experience increase in background noise as well as material embrittlement, which can cause cabling, or other vital equipment to fail. Significant shielding of the electronics will be required to lengthen their operational time.

6.2 Lack of an Accountability Tank

An important aspect of the safeguard strategy for aqueous reprocessing facilities is the input and output accountability tanks. These tanks allow the operators to have low uncertainty measurement (~0.1-0.5%) of material before processing begins and after processing occurs, allowing for closing material balances within regulations. Though with reliance on dissolving the fuel the input shipper/receiver difference cannot be calculated until the samples are analyzed; an experience at THORP demonstrated this

challenge. In April 2005, a leak in an accountability inlet nozzle was discovered after approximately 83,000 liters of dissolver product solution (about 22,000 kg of dissolved fuel of which 160 kg was plutonium) leaked onto the facility floor. It was estimated the leak had occurred for eight months before detection (Health and Safety Executive, 2007).

Pyroprocessing, however, does not have a point in the initial processing stages, which would allow for an accountability tank type measurement. The desirable aspect of the measurement is the homogenous dissolution of the entire SNF assembly, so that the isotopic distribution of the solution can be determined. The fuel pellets in pyroprocessing are in an inhomogeneous form until they are dissolved within the electrorefiner salt. The fuel pin and corresponding fuel pellets will have wildly varying isotopic distributions based upon irradiation history, positioning within the reactor core (i.e. near control rods), and initial enrichment.

Various attempts have been made to create a homogenous measurement point after the assembly is disassembled; the ROK's proposed reference facility grinds the fuel pellets into a fine powder, which can be destructively analyzed (Kim, 2012). While this may help safeguard implementation it also increases the burden on safety systems. The maximum consequence accident in a pyroprocessing facility is a fire within the argon hot cell and if spent fuel is present in powder form during this accident, this powder could be released through the ventilation system. This possible scenario means the ventilation system will need to be of the highest safety grade construction, increasing maintenance and construction costs of the facility.

Another possible strategy is using gross neutron counting to track Pu bearing materials throughout the processing cells. Use of this method would require knowledge

of the contribution of Cm to the total count, as Cm in LWR fuel is a significant contributor. A DA sample of homogenous powder of input fuel is used to determine the Cm/Pu ratio; providing a correction to the gross neutron tracking signals for Cm contribution. This method assumes Cm and Pu will transport the same throughout the process; with any gross neutron count in a waste form equating to some Pu present. This assumption has not been able to be experimentally verified by work conducted at INL (Bean, 2010). The ANL process flow assumes some Cm as well as Am will be present in the ceramic waste form, so this strategy is not valid for this work.

6.3 In Process Measurements

Conventional safeguards strategies separate process knowledge from safeguards knowledge. For aqueous reprocessing this can be a challenge because of the number of tank samples needed throughout the process, but the solution is fundamentally simple with cylindrical geometry and material spread throughout the facility. However, the ER does not have a simple geometry and contains two assemblies worth of fuel plus holdup at any time. A few other conditions complicating the issue are the 500°C temperature of the operating salt within the ER as well as the hot cell environment, discussed in Section 6.1, any detection system operate in consistently. Much effort has been spent looking into this issue at ANL and INL as well as universities. Most techniques developed have not been capable of providing precise inventory measurements but could be used for process monitoring to provide gross estimates of the in vessel contents. Since process data is not used in safeguards, a dedicated safeguards in-process device would be needed as well; taking up valuable floor space by possibly having duplicate detectors

within the hot cell increasing the cost of the facility overall.

6.4 Ability to Detect Diversion

The most important feature of any safeguard system is how alarms are raised when diversion is detected. As discussed in Section 2, regulations are based upon SEID, which is a measure of the effectiveness of the integrated measurement devices. SEID provides a means to determine if the measurement system can detect a protracted diversion over many balance periods; however, it is insensitive to abrupt diversion attempts. The more sensitive Page's test, discussed in Section 5.4, can be used to supplement the SEID measurements to strengthen a system. Page's test is, however, a sequential test so diversion attempts early in a campaign are easier to detect than those later. A point will occur where the signal will no longer exceed the test threshold and no alarm will trigger. The effect of sequential testing is illustrated in Figure 21, showing two Page's test signals for diversion of one SQ of material during two separate inventory balances. The left graph shows the Page's test signal (red line) exceeding the threshold $h=5$ (magenta line) at $t=2160$; the diversion event occurs from $t=1500$ to 1600 hours. In this case, the alarm is tripped within 30 days of diversion resulting in successful detection. The right graph displays the same threshold, $h=5$, however diversion occurs from $t=8000$ to 8100 hours. An initial increase is seen in the resulting Page's test signal following the diversion however this does not exceed the threshold and no alarm is raised. The threshold of $h=5$ was chosen based upon its performance in testing in abrupt and protracted diversion scenarios as well as its use in other implementations of the Page's test (Cipiti B. e., 2012).

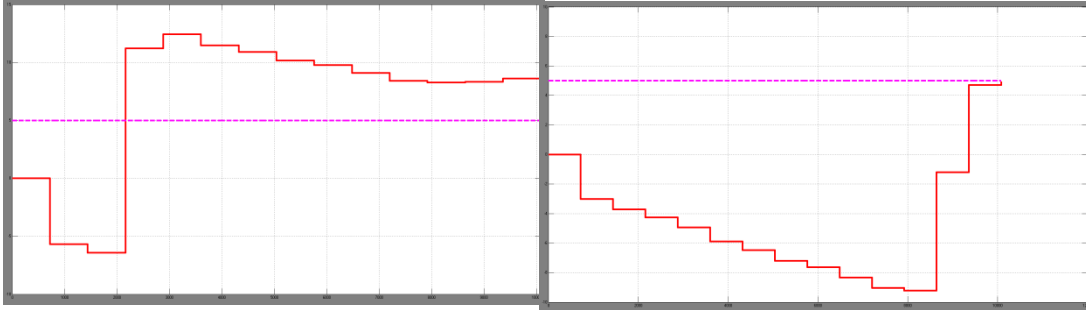


Figure 21 Page's test signal (red) and threshold $h=5$ (magenta) for two cases of a SQ diversion

Using the PSPM a comparison between the SEID and Page's Test alarms can be seen in Table 7.

Last Month in Which 1 SQ Diversion is Detected		
σ	SEID (month)	Page's test (month)
0.10%	>24	22
0.50%	>24	8
1%	16	3
2%	3	0
5%	0	0

Table 7 SEID vs. Page's test for 1 SQ diversion

The results in Table 7 were obtained assuming each input, output, and inventory measurement was made with uncertainty, σ . The results also used the inventories from each of the processing vessels including the holdup to provide a best-case detection scenario. As is seen from the results, if the uncertainty is 0.1% for all measurements, then the system would be capable of detecting diversion for two years before recalibration

would be needed; however, this is not achievable with current detection techniques for any measurements. A more realistic, but still difficult scenario is the 1% case. As shown in Table 7, any abrupt diversion after three months would go undetected by the Page's test and SEID is insensitive to abrupt diversion so this diversion would go undetected, an unacceptable scenario. However, if a recalibration of the measurement devices (test with known standards and obtain new calibration curves for each device) is performed after the third balance period, the Page's test could be reset and be valid for another three months. The SEID limit would then require an estimation of the in cell inventory to reset for another processing campaign. A plant operator would ideally desire to have a campaign length of a year with monthly balances to confirm diversion is not occurring.

7. Risk Informed Safeguards Approach

7.1 Basis from Nuclear Safety

The current safeguard approach has been effective for current facilities with current technology but it will not be sufficient for the next generation reactors requiring reprocessing. To illustrate this point a comparison of MC&A detection goals for a large LWR reprocessing facility and a moderate throughput fast reactor reprocessing facility is shown in Table 8 (Lineberry, 2011).

Aqueous Reprocessing Technology Results		
	LWR Application	Fast Reactor Application
Heavy Metal Capacity	800 MTiHM/yr	250 MTiHM/yr
Pu Content (% HM)	1.33%	6-20%
Pu mass flow (max)	36 kg/day	50-170 kg/day
Inventory period	30 days	30 days
Input Pu mass uncertainty	0.70%	0.70%
SEID	7.86 kg	10.92-37.13 kg
Altered inventory period to achieve SEID goal	9.2 days	6.6-2.0 days
Altered input Pu uncertainty to achieve SEID goal	0.10%	Not Possible

Table 8 Aqueous Reprocessing Technology Results

If only the inventory period is modified to meet IAEA detection goals then about a nine day inventory period is needed for the LWR facility and two to six days at a fast reactor facility; the LWR period is feasible but would not be possible for a fast reactor facility. If the input Pu mass uncertainty was improved similar results are achieved. This example highlights the need for a new strategy to safeguard these facilities; a risk informed approach utilizing the Safeguard Hazard Matrix (SHM).

The SHM is adapted from safety analysis in nuclear facilities. The Department of Energy regulates fuel cycle facilities based on a hazards analysis where accidents are analyzed based on frequency of occurrence and consequence of the accident. An example of the risk matrix used in nuclear safety analysis is provided in Figure 22. An unmitigated case, one where no safety systems are taken into account, is analyzed to determine its consequence. This is used to address the importance of a safety system to the facilities operation and what quality standards for that system are necessary.

Likelihood	Anticipated	7	11	14	16
	Unlikely	4	8	12	15
	Extremely Unlikely	2	5	9	13
	Beyond Extremely Unlikely	1	3	6	10
		Negligible	Low	Moderate	High
Consequence					

Figure 22 Nuclear Safety Risk Matrix

A similar approach could be applied to facility safeguards as well. Other proposed approaches plot the amount of material diverted against the diversion success probability (P_s) then an overall risk for the scenario can be determined, shown in Figure 23 (Cavalieri d'Oro, 2012).

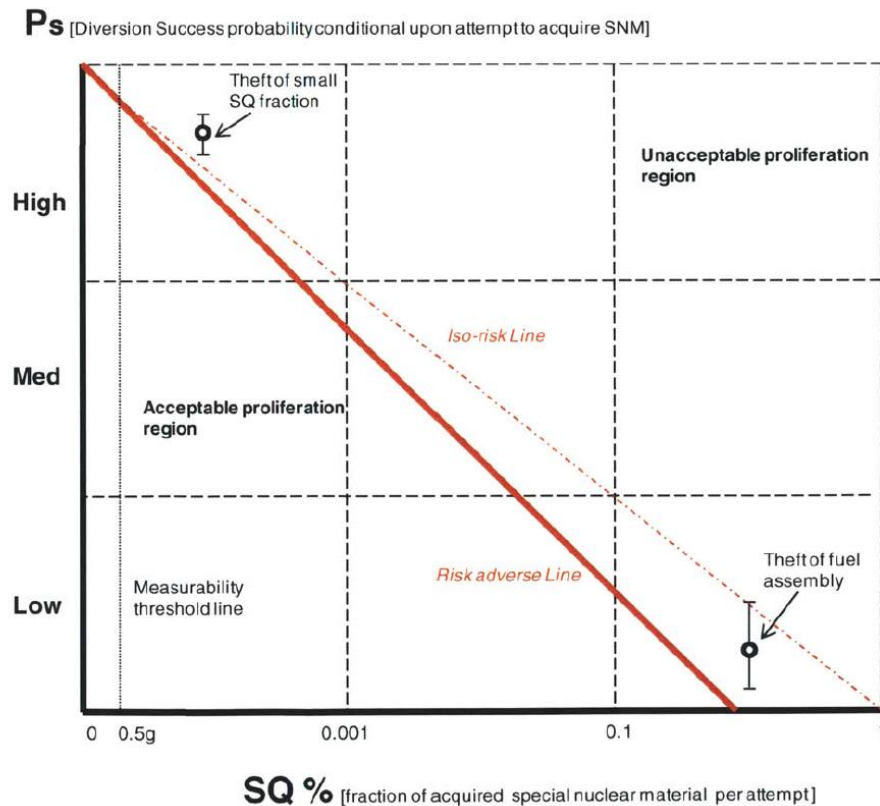


Figure 23 P_s vs. $SQ\%$ diverted

P_s is based upon the number of attempts needed to divert one SQ as well as the accessibility of the material within the facility. Based upon some regulator guidance an acceptable risk range would need to be developed, similar to how the NRC has set the safety risk of core damage of a power plant to be 1×10^{-5} per year for new reactor designs.

7.2 Safeguard Strategy Descriptions

The PSPM was used to investigate if different safeguard approaches, with varying measurement techniques, would be able to detect the loss of a SQ of SNM. Common to each of the strategy is the assumption that the one MBA is used to encompass both the processing and fuel preparation cells. Figure 24 shows a possible facility layout with arrows to denote transfer pathways across the proposed MBA boundary (blue, dashed line). The arrows represent entrance/exit pathways of the MBA: Yellow-SNF input assemblies, Red-U/TRU, Green-U product, Metal waste, and Ceramic waste, Orange-Cladding & Hardware waste, Purple-Large equipment transfer lock.

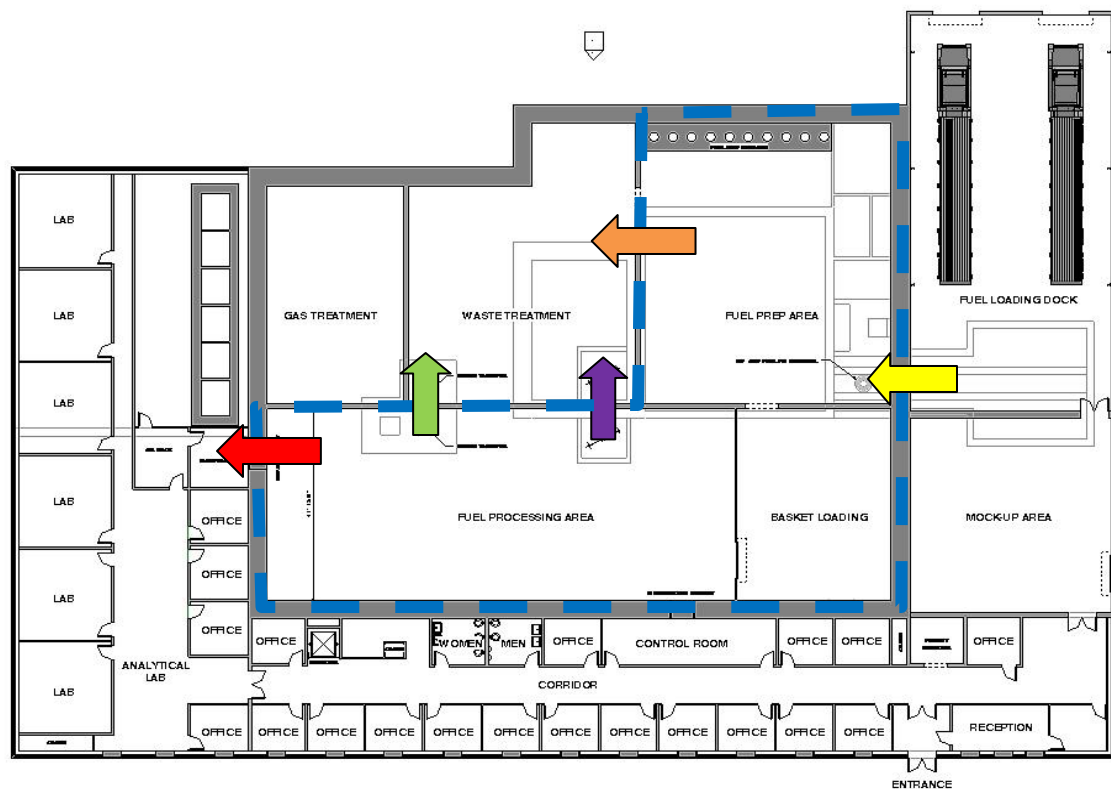


Figure 24 MBA outline for reference facility.

The equipment transfer lock is not included in this analysis further, as use of this pathway

would constitute an off normal processing event, which is not considered by this work. It is, however, acknowledged that this transfer lock will need C/S measures to guarantee SQs of material are not removed while components are transferred to/from the processing cell.

Four safeguard strategies, known as levels, were used to demonstrate the effectiveness of a risk matrix as a tool in safeguard evaluation. Each level builds upon elements of the previous level to enhance the strategy.

The level 1 strategy consists of treating the facility as a black box with only the inputs and outputs considered. ID can be calculated as the transfers into the MBA minus the transfers out of the MBA, with no measure of the inventory within the facility. Figure 25 shows key measurement points (KMPs) and the material measured at each point for this strategy in blue.

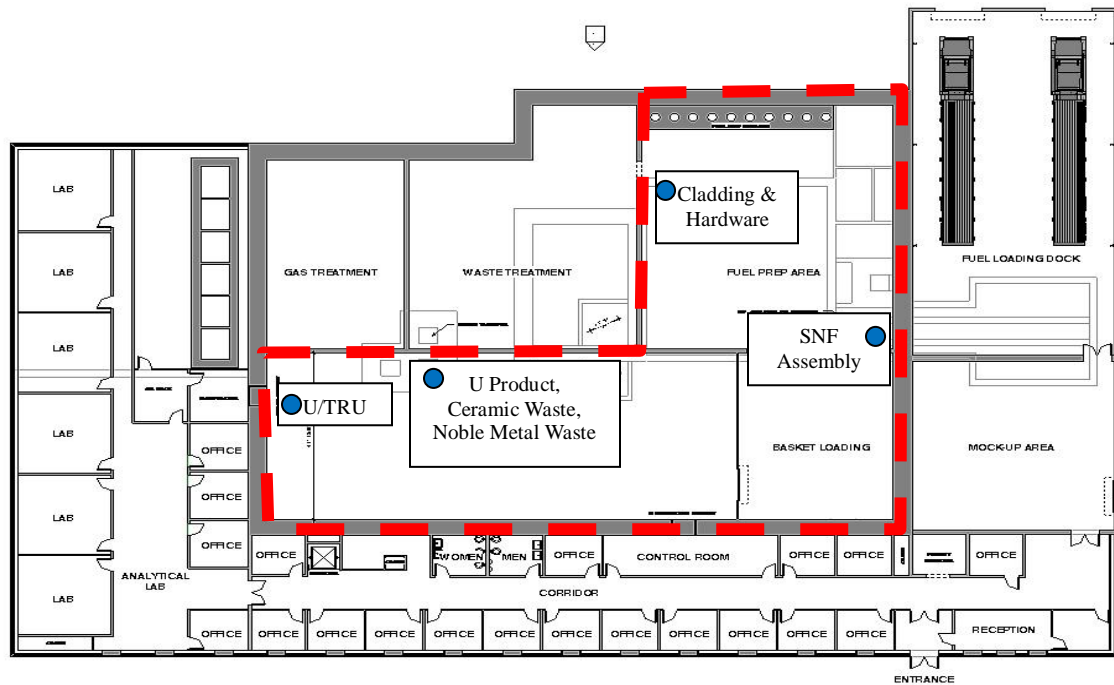


Figure 25 Level 1 Key measurement points

This is considered as the reference or unmitigated case and additional safeguards systems are needed to decrease the risk to an acceptable level (to be determined by a regulator). Any material differences are assumed to be contained within the MBA.

Level 2 expands upon the level 1 strategy by including measurements from storage locations of material. Using the PSPM storage locations containing SNM at the end of each balance period were determined. Four storage locations were identified and incorporated into inventory measurement; storage of Uranium to be sent to oxidant production, storage of BMs awaiting electroreduction, a BM awaiting a second before it is stored before electroreduction, and the salt collection tank. Figure 26 shows the KMPs of level 1 in blue and new level 2 KMPs in green with associated locations within the cell.

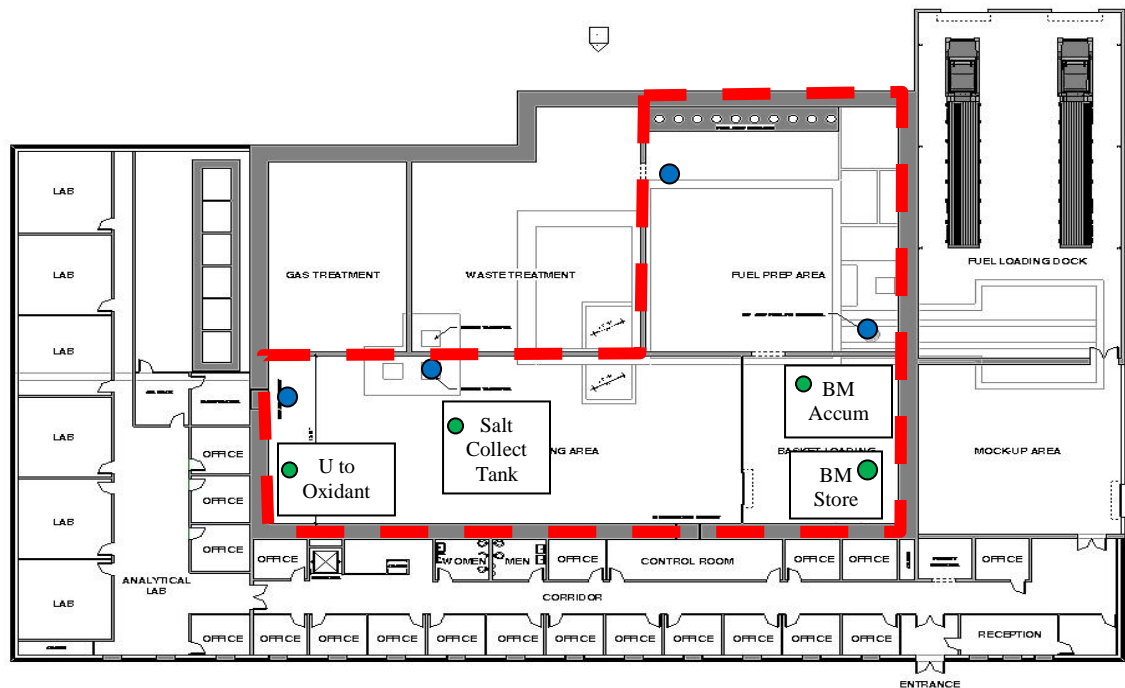


Figure 26 Level 2 Key measurement points

For each location, the overall mass is reported along with the corresponding measurement uncertainty from the process model to the monitoring system. Within the *U & Pu ID Calc* function block a percentage of the total mass corresponding to either U or Pu is applied. For example: the BM storage signal is multiplied by 91.37% and 1.112% for U and Pu respectively. These ratios are based upon the process flow sheet (Benedict). This strategy represents the conventional safeguards approach and the current inability to accurately sample processing vessels with demonstrated techniques today.

Level 3 integrates process data into level 2, a process informed safeguards strategy. The process monitor system, which can have the most impact on a safeguard system, is a detailed mass tracking system. With such a system, mass balances are used to weigh every item before and after it is processed by a unit or moved from one zone to another within the MBA; the MBA should be divided up into zones for each processing unit. Including process information in the safeguards strategy will require the mass tracking system to be available for both the operator and international inspectors. Traditionally this stipulation has required two separate devices, one for the IAEA and second identical system for the operator, however joint use systems have been implemented at RPP in areas where two detectors are not possible. These systems provide independent and identical signals to each party, which can be analyzed by each organization with separate conclusion drawn (Johnson, 2010). Figure 27 shows the KMPs for level 1 and 2 in blue and green respectively; level 3 & 4 KMPs are shown in yellow.

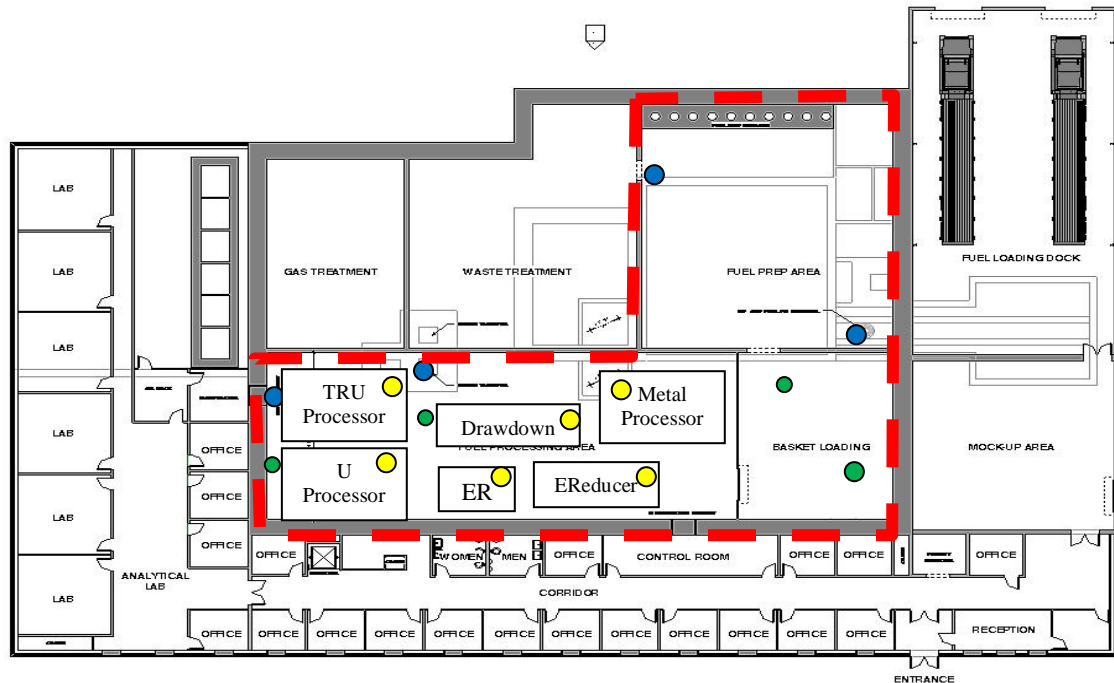


Figure 27 Levels 3 & 4 Key measurement points

The balances are connected to a network, which can store all vital information about a specific item such as starting weight when it entered a zone, the item's serial number, and the balance to be used so an appropriate calibration factor can be applied. Verifying an item is the correct weight and serial number by two independent observers adds an additional layer of security. Also the tracking system gives great assurance that no diversion has occurred with that item. This is the system implemented at FCF. This system has been used for almost 20 years (with some upgrades from the original system installed in 1995) to handle over 35,000 fissile material transfers (as of 2006) resulting in zero mass violations (Pope C. , 2007).

A system similar to that of FCF would be capable of providing information on the number of items in processing throughout the facility. For example: the number of BMs

in the electroreducer at the end of a balance period can be added to the inventory using an assumed average composition for a BM or that specific BM if it is an off normal variant because it has been tracked since entering as a fuel assembly. The mass passed to the *U & Pu Calc* function is found by multiplying the average mass of the item (randomly sampled as described in Section 5.1 for the SNF sensor), for example a BM of oxide fuel, the number of entities present at the end of the balance period, the weight percentage of U or Pu within the item, and the mass conversion to a metal form (if not already a metal). This strategy adds safeguard information while not forcing the facility to shut down for a balance closure each 30 days or less as occurs in aqueous facilities. This is a necessary attribute of any pyroprocessing strategy because the ER cannot be shut down once it is brought to operating temperature. The ER in FCF has been at temperature since 1995. As implemented in the PSPM only the holdup within a processing vessel is not measured so the ID represents the holdup and random fluctuations from measurement uncertainties.

Level 4 applies rejection sampling to the input and output measurements determined in level 3; the same KMPs as level 3 are used. The use of an integrated mass tracking system implies every item entering and exiting the MBA will be weighed and only items within an accepted tolerance will be allowed to leave or enter the cell; anomalies will be rejected and not be allowed into the cell or an alarm is tripped. The PSPM determines a random mass from a normal distribution with a mean of the mass from the process model and a standard deviation of the measurement device for each group. However, in reality if the sum of all these random measurements exceeds the tolerance of the mass balance, then the item would be rejected. To illustrate this, assume an item is composed of three materials a, b, and c with corresponding average masses of

one, two, and three. Now assume the measurement device has a standard deviation of 10% for each individual material while the balance has a tolerance of 0.1% for the total mass of the item. The average item mass is then six; however, using random sampling for each material could result in an estimation of the item mass greater than the mass balance's tolerance. A MATLAB script was created to show the effect of rejection sampling for this case, the results included in Table 9 were generated with one million simulated masses for each material.

	No Rejection Sampling		Rejection Sampling	
Material	Mean	STD	Mean	STD
a	0.9998	0.1000	1.000	0.0819
b	1.9994	0.1002	2.000	0.0818
c	2.9996	0.0999	3.0001	0.0818
Total	5.9988	0.1000	6.0001	0.0170

Table 9 Rejection sampling effect

The rejection sampling is invoked if the sum of a, b and c is greater than or less than the mean $\pm 3*\sigma_{\text{balance}}$ (6.018 and 5.982 respectively). If these conditions are true, the loop continues to generate random normally distributed pairs until the condition is false. Rejection sampling is only a benefit if the device distribution has a greater uncertainty than the mass balance uncertainty, and if both are similar in magnitude less benefit is gained. This technique results in an order of magnitude decrease in the standard deviation of the total mass of the item. A great benefit to the safeguard system, it helps to

reduce the spread of ID measurements and gain increased assurance from less precise MC&A equipment.

8. The Safeguard Hazards Matrix

Each of the levels discussed in Section 7.2 were implemented in the PSPM with varied measurement uncertainties for each level with the exception of level 1 (only one case was run to establish the reference case), to be evaluated using the Safeguard Hazards Matrix.

8.1 Methodology

The PSPM was set to run for 10,080 hours (14 months) allowing two months for units to reach equilibrium. A MATLAB script, *multiruns.m*, was created to repeatedly run the PSPM and collect output data for post processing. The repeated execution code is provided in Table 10.

```
for i=1:batch
    seed=ceil(1000.*rand(1,1));
    simout=sim('simevent_processflow_rs.slx','ReturnWorkspaceOutputs',
    'on','SrcWorkspace','current');
    test=simout.get('pages_test');
    if i==1
        data=[test.signals.values];
    else
        data=[data;test.signals.values];
    end
    disp(i);
end
```

Table 10 MATLAB code for multiple execution of model

The model was run using a Dell Optiplex 9020 desktop computer with eight Intel Core™ i7-4770 CPUs and 32 GB of RAM. To produce all results the PSPM was run 1,000 times (referred to as a batch) each with different random seed values to not produce identical runs; the average run time was two hours for seven batches run concurrently⁵. Change in

⁵ Seven instances of MATLAB were run each on a single CPU while the 8th CPU was left to run the operating system and other background applications.

batch size followed a roughly linear relationship so 100 runs would take 750 s (12.5 min) and 10,000 runs would take 20.8 hours. This was deemed too resource intensive so a batch size of 1,000 was used. As a reference point, the Separations and Safeguards Performance Model-EChem (Cipiti B. e., 2012) was run for only 1,660 hours and batches were set at 100 with run times on average of 4.5 hours on an eight core GPU computer (McDaniel, 2014). This comparison demonstrates the advantage of using a discrete event model as opposed to a continuous model.

The PSPM outputs a vector, *pages_test*, containing the time, U and Pu ID calculation results as well as the Page's test results upon completion of a single run. These are then stored in a vector, *data*, for each run of a batch. After a batch is complete the resulting data vector is broken up into two separate elements related to U and Pu. The ID results for U and Pu are each averaged over all runs as well the SEID, and a standard deviation is calculated for each averaged value. The detection probability is calculated using the average SEID and reversing the methodology used in Section 2.1 to determine the σ_{MUF} threshold for 95% detection, the detection probability for loss of one SQ can be shown to be:

$$DP = \frac{\text{erf}\left(\frac{Q}{2 * SEID * \sqrt{2}}\right) + 1}{2} \quad (18)$$

In equation 18, Q is the amount in kg of material representative of one SQ, SEID is the calculated SEID from the PSPM, and DP is the detection probability. A factor of two in the error function arises from the assumption that the false alarm probability is also the same; see Section 2.1.

8.2 Results

As discussed in Section 7.1, using the SHM to evaluate safeguard strategies would be of great benefit for any facility. The risk is determined as proportional to the amount of material unaccounted for (ID) and the probability to detect a loss of a SQ. Using the PSPM, the limiting factor for a SQ of material is the Pu, not the U, was observed. Several cases were simulated using the PSPM and plotted in Figure 28; the corresponding measurement uncertainties for each data point are included in Table 11.

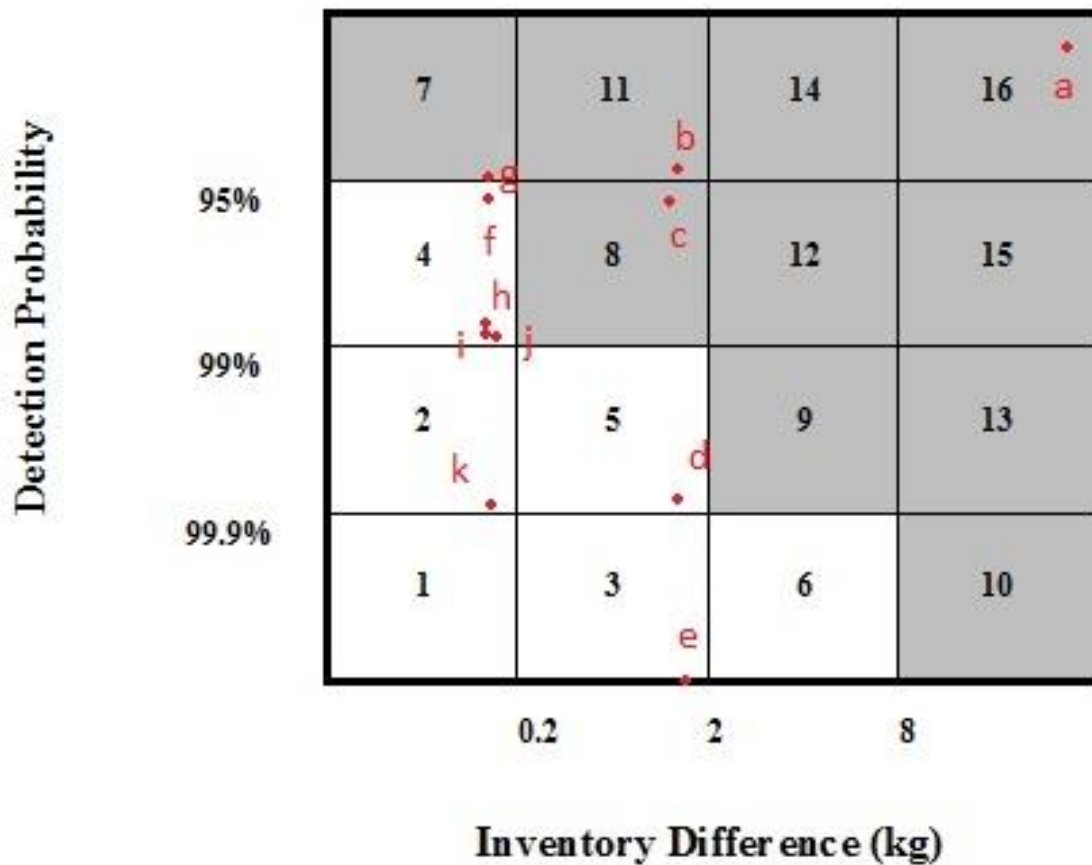


Figure 28 Detection Probability vs. Inventory Difference.

Case	Strategy	σ SNF	σ TRU prod	σ U prod	σ NB	σ Clad	σ Inv
a	Level 1	1%	1%	1%	1%	1%	-
b	Level 2	5%	5%	10%	10%	10%	0.5%
c	Level 2	2%	2%	10%	10%	10%	1%
d	Level 2	1%	1%	1%	1%	1%	1%
e	Level 2	0.5%	0.5%	0.5%	0.5%	0.5%	0.5%
f	Level 3	2%	2%	10%	10%	10%	1%
g	Level 3	1%	3%	10%	10%	10%	1%
h	Level 3	1%	2%	10%	10%	10%	1%
i	Level 4	2%	0.5%	10%	10%	10%	0.5%
j	Level 4	2%	1%	10%	10%	10%	0.5%
k	Level 4	1%	1%	1%	1%	1%	1%

Table 11 Case Legend for Figure 28 and Figure 29

The limits for the x-axis bins were based upon the requirement by DOE for SEID < 2 kg, while the others were the author's intuition. Risk bins 7-16 (grayed boxes) are unacceptable strategies while bins 1-6 are acceptable. A general feature of the strategy, shows that as measurement uncertainty is decreased within a strategy the point moves down the matrix (ex. b to e) and as each additional strategy decreases the ID the points moves to the left (ex. d to k).

Upon inspection of the standard deviation in the simulated ID calculations of a batch, it is observed that while SEID is a measure of the detection system it is not a measure of the variation in ID over several balance periods.

For level 2, even though its ID average was 1.8 kg the standard deviation varied

substantially throughout the simulation, while levels 3 and 4 did not have as substantial variations, as shown in Table 12.

Strategy	STD
Level 2	9.42
Level 3	0.85
Level 4	0.75

Table 12 PSPM standard deviation average for each strategy

A positive conclusion that no diversion occurred can be made confidently for levels 3 and 4, though the same cannot be made for level 2. To remedy this observation a quality factor, QF, is proposed as an empirically derived factor to be applied to the calculated ID.

$$QF = \frac{3 * STD \text{ of } ID}{SQ \text{ of material}}$$

The QF could be determined from operational data or simulated with the PSPM in the absence of operational data. A compensated ID, ID_c , can now be calculated as:

$$ID_c = QF * ID$$

This effectively rewards strategies for low variation in measurements while disadvantaging those with largely varying IDs. Figure 29 reflects the inclusion of the QF with the results shown in Figure 28; Table 11 provides a description of each case.

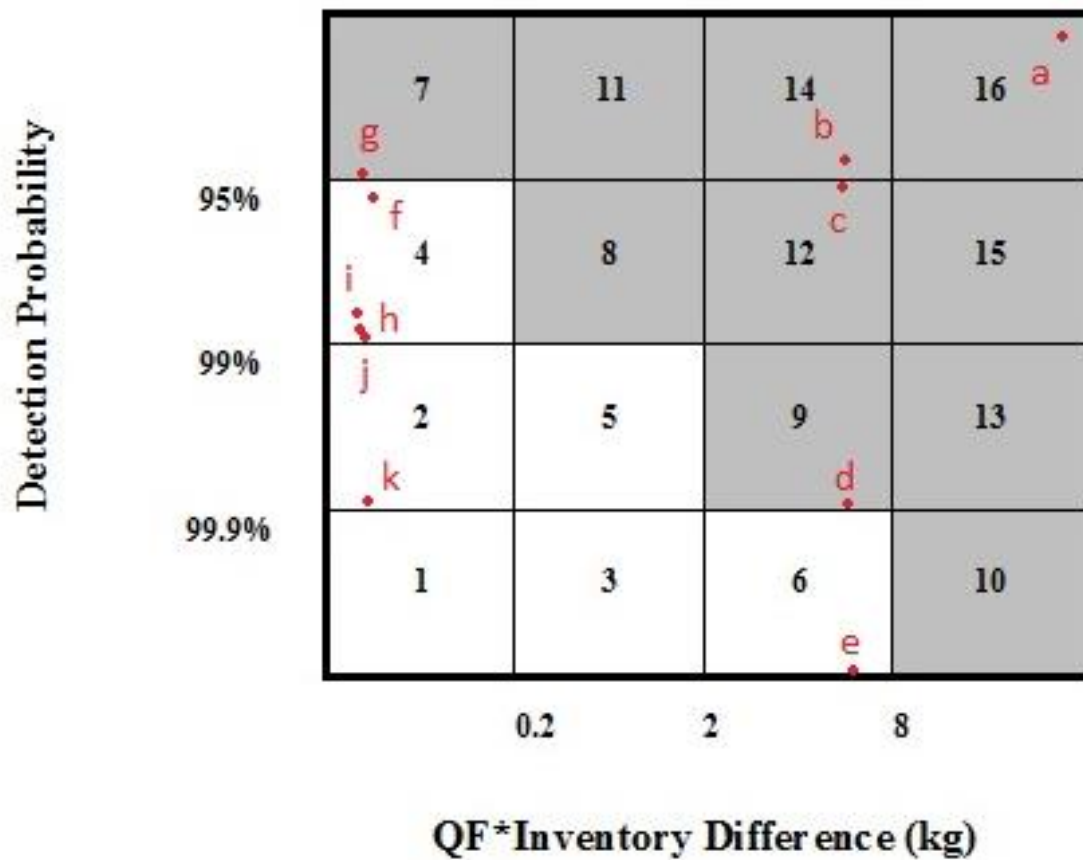


Figure 29 Detection Probability vs. ID risk matrix

As can be seen in Figure 29, cases b through e are shifted right an entire risk bin while cases f through k shift to the left to be closer to the center of the bin; point d actually crosses from acceptable to unacceptable.

Some important observations to be made from Figure 29 are 1) the conventional safeguards approach will not work for this facility unless detection is capable of uncertainty levels <1% (closer to 0.5% will be needed) and 2) process knowledge is of great benefit to obtaining a material balance closer to zero and allows for possible scenarios with reasonably achievable measurement uncertainties today.

The sensitivity of each measurement contributing to the ID calculation can be

demonstrated as well using the PSPM. The SEID calculated for each case (a-k) is shown in Table 13.

Cases	σ SNF	σ TRU prod	σ Inv	σ Clad	SEID (kg)
b	5%	5%	0.5%	10%	5.7089
c	2%	2%	1%	10%	2.3899
d	1%	1%	1%	1%	1.3415
e	0.5%	0.5%	0.5%	0.5%	0.6708
f	2%	2%	1%	10%	2.3949
g	1%	3%	1%	10%	2.5186
h	1%	2%	1%	10%	1.8771
i	2%	0.5%	0.5%	10%	1.7912
j	2%	1%	0.5%	10%	1.9062
k	1%	1%	1%	1%	1.3505

Table 13 Top Three Most Sensitive parameters and SEID

From Table 13, it can be seen that both the input spent fuel measurement and the TRU product measurements both greatly dominate the SEID while the other measurements do not have as great an effect. The detection probability will be less than 95% for an ID if σ_{SNF} or σ_{TRU} are greater than 2%. The cladding measure is less sensitive in the Pu balance since it has such a low Pu concentration (~5 g per BM and total of ~85 g per balance period) while ~85 kg of SNF and TRU product enter/exit the MBA within a 30 day period (any observed difference represent the amount of Pu inventory within processing vessels or storage at the end of a balance period). Mass tolerances of <1% for a 0-120 kg WE400C electronic balance have been documented at FCF (Orechwa, 1997). New balances will need to be constructed with mass upper bounds of one metric ton, capable of withstanding the high radiation environment in-cell. The TRU product can be assayed destructively within the cell by drilling a sample from the ingot or when the ingot is formed a break off mold is also filled with the same homogenous molten TRU product.

The sample can then be analyzed at an analytical lab located onsite using isotopic dilution mass spectroscopy (IDMS), which in a hot cell environment has a total σ of 0.28% to 0.42% (IAEA, Nov 2010). Current NDA waste assay techniques can be applied to the cladding waste to achieve a 10% uncertainty. The most challenging measurement is the SNF fuel. As outlined in Section 4.1, a coupled PNAR/SINRD detector and neutron tomography suite has the best chance of success; however, further investigation will be needed to determine the precision capable of such a system. Currently NDA techniques are accurate to ~10 % though the NGSF project is aiming for ~1%. If only 3% is obtained, very low (~0.5%) mass balance, and TRU product measurements (<0.5%), then this MC&A system, case i in the SHM, could only be possible using a process informed safeguard strategy. The SHM shows cases e, f, and h through k are acceptable safeguard strategies, with f, h, i, and j most reasonably achievable within the foreseeable future. Case e is not thought to be feasible, SNF uncertainty measurement of 0.5%, without a breakthrough in the fundamental underlying physics of detection and case k is thought to be achievable more long term (10-15 years) for the same reason.

9. Conclusions

An overview of the current state of reprocessing regulations was conducted for three potential regulators: IAEA, NRC, and DOE. A conclusion from this review showed inconsistencies among each regulator (the NRC having the most). If a commercial pyroprocessing facility is to be built, further work must be done to develop an effective safeguard strategy. A brief overview of the current developments in NDA technology was also conducted with the aim of finding technologies suited for pyroprocessing's unique challenges. Several candidate technologies were discussed as well as possible use locations within the facility. In an effort to aid in testing the effect of proposed safeguards strategies, the pyroprocessing safeguards performance model (PSPM) was created using MATLAB/Simulink and the SimEvents toolbox. The PSPM was used to test four strategies: the black box, conventional, process informed, and process informed with rejection sampling. An empirically derived quality factor was developed to provide a measure of the spread of the calculated ID over the course of operation. This assigned less risk to ID measurements with narrow varying measurements while assigning more risk to those with large variations. The Safeguard Hazards Matrix was created, Figure 29, representing probability to detect one SQ of diverted material against the compensated ID. Six possible detection scenarios were shown to be of acceptable risk of which four are achievable within the next five years; all of which use process informed strategies. It has been shown that including process information, specifically mass balance data from an extensive mass tracking system, reduces the risk in a pyroprocessing facility significantly allowing IAEA safeguards to be applied.

10. Future Work

- The PNAR/SINRD with tomography suite shows much promise but requires more investigation to determine the number of projection profiles needed and investigation into the integration of these two system together.
- While the PSPM was developed under the assumption of process equilibrium further work can be conducted looking into the startup conditions and the safeguards implications.
- Another safeguards strategy of interest consists of ER designed for processing a single assembly. This would minimize the 100 kg working inventory within the ER assumed in this work. The PSPM is flexible enough to model this with some minor modifications.
- After a Page's test alarms for diversion, further investigation is needed to understand how to reset this alarm from the facility operation perspective.

11. Bibliography

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Appendix A. MATLAB Program Files

A.1 SNF_source.m Input File

```
%% Import the data
[~, ~, raw] = xlsread('C:\Users\riletony\Dropbox\safeguards\Papers to
myself\SNF source.xls', 'Sheet1');
raw1 = raw(2:33,2);
raw2 = raw(2:7,4);
raw3= raw(2:4,6);
%% Create output variable
FPMass = reshape([raw1{:}],size(raw1));
ActMass = reshape([raw2{:}],size(raw2));
APMass = reshape([raw3{:}],size(raw3));
%% Clear temporary variables
clearvars raw raw1 raw2 raw3;
%% Initiate mass values from SNF for simulink model
% Origen source file is based on 1 MT while model should be on a per
% assembly basis so mass from file will be halved
U= ActMass(2)/2;
Np= ActMass(3)/2;
Pu= ActMass(4)/2;
Am= ActMass(5)/2;
Cm= ActMass(6)/2;
% Cladding and Hardware
% Contains Activation Products (Zr, Nb, Mo, & Se)
Clad_HW=[APMass(1:3)./2;FPMass(1)/2];
% Active Metal Elements
% Contains: Rb, Cs, Sr, Ba, Sm, Eu
Active=FPMass(18:23)./2;
% Rare Earth Elements
% Contains: Y, La,Ce, Pr, Nd, Pm, Gd, Tb, Dy
RareEarth=FPMass(24:32)./2;
% Noble Metals
% Contains: Zr, Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te
Noble= FPMass(6:17)./2;
clearvars FPMass APMass ActMass
%% Each waste stream is collapsed to a constant
Clad_HW=sum(Clad_HW);
Active= sum(Active);
RareEarth=sum(RareEarth);
Noble=sum(Noble);
%% Convert flows from grams to kg
U=U/1000;
Np=Np/1000;
Pu=Pu/1000;
Am=Am/1000;
Cm=Cm/1000;
Clad_HW=Clad_HW/1000;
Active=Active/1000;
RareEarth=RareEarth/1000;
Noble=Noble/1000;
```

```

%% Setup Measurement Uncertainties for the Different Detectors
throughout the model
sigSNF=0.01; %SNF Assembly detector
sigTRUprod=0.01; %TRU product detector
sigUprod=.01; %U Product detector
sigNB=0.01; %Noble Metal Waste detector
sigClad=0.01; %Cladding and HW waste detector
sigMassXL=0.01; %Mass balance tolerance for items >50 kg
sigMassSm=0.01; %Mass balance tolerance for items <50 kg
sigRE=0.01; %Rare Earth Waste detector
sigAM=0.01; %Active Metal Waste detector
%% Set Inventory Period for Plant Balance Calculation
InvPeriod=30; %Enter the period in days here.
InvPeriod=InvPeriod*24; %Do not change this line
seed=0;

```

A.2 Multiruns.m Input File

```

%% Variable declaration
tic
batch=1000;
Ur= zeros(15*batch,3);
P=zeros(15*batch,3);
Ucount=0; Ualarm=0;
Pucount=0; Pualarm=0;
t=zeros(15,1);
etest=zeros(15,1);
%% Execute Simulation batch
for i=1:batch
    seed=ceil(1000.*rand(1,1));
    simout=sim('simevent_processflow_rs.slx','ReturnWorkspaceOutputs',
'on','SrcWorkspace','current');
    test=simout.get('pages_test');
    if i==1
        data=[test.signals.values];
    else
        data=[data;test.signals.values];
    end
    disp(i);
end
%% Processing of alarm signals from the simulation runs
for j=1:9
    if j<5
        for i=1:length(data)
            Ur(i,j)=data(i,j);
        end

    else
        for i=1:length(data)
            P(i,j-4)=data(i,j);
        end
    end
end

```

```

        end
    end
    P=[Ur(:,1).*720,P];
    clear j i
    toc
    %%
    %determine detection probability and the STD of ID measurements
    IDave=sum(P(:,2))/(length(P)-batch);
    SEIDave=sum(P(:,3))/(length(P)-batch);
    detect=(erf((8/(2*SEIDave))/sqrt(2))+1)/2;
    IDstd=std(P(:,2));
    f=[abs(IDave),SEIDave, 1-detect, IDstd];

```

A.3 U & Pu Calc Function Block Script

```

function y = fcn(input,output1, output2,output3,output4,inventory1,
inventory2, inventory3, inventory4, inventory5,
inventory6,inventory7,inventory8,inventory9,inventory10,inventory11)
global simtime
persistent id time a b sigv2 sigx2 Vsit Xsit
size=14;
if simtime>0
if isempty(time)
    time=1;
    id=zeros(size,8);% 1-4 U balance, 5-8 Pu balance:
                    % ID, inv, sigma inv^2, sigma ID^2
    a=0;
    b=0;
    sigv2=0;
    sigx2=0;
    Vsit=0;
    Xsit=0;
else
    time=time+1;
end
in=[input(1)*.8814,input(3)*.8814];
ins=[in(1)^2*(input(10)^2),in(2)^2*(input(10)^2)];
out1=[output1(1)*.8814,output1(3)*.8814];
o1=[out1(1)^2*(output1(10)^2),out1(2)^2*(output1(10)^2)];
o2=output2(1)^2*(output2(2)^2);
o3=[output3(1)^2*(output3(7)^2),output3(3)^2*(output3(7)^2)];
out4=output4(1)*.8814;
o4=out4^2*(output4(3)^2);
inv1=inventory1(1)^2*(inventory1(3)^2);
inv2=inventory2(2)^2*(inventory2(3)^2);
inv3=inventory3(2)^2*(inventory3(3)^2);
invent4=[inventory4(1)*.9137*.8814,inventory4(1)*.0112*.8814];
inv4=[(invent4(1)^2)*(inventory4(3)^2),invent4(2)^2*(inventory4(3)^2)];
invent5=[inventory5(1)*.9137*.8814,inventory5(1)*.0112*.8814];
inv5=[invent5(1)^2*(inventory5(2)^2),invent5(2)^2*(inventory5(2)^2)];
invent6=[inventory6(1)*inventory6(2)*0.78263,inventory6(1)*inventory6(2)
)*.009561];

```

```

inv6=[invent6(1)^2*(inventory6(3)^2),invent6(2)^2*(inventory6(3)^2)];
invent7=[(inventory7(3)*(419.1924/6)),(3-inventory10(2)-
3*inventory9(2))*4.2457];
inv7=[invent7(1)^2*(inventory7(5)^2),invent7(2)^2*(inventory7(5)^2)];
invent8=[inventory8(1)*inventory8(2)*.1598,inventory8(1)*inventory8(2)*
.30696];
inv8=[invent8(1)^2*(inventory8(3)^2),invent8(2)^2*(inventory8(3)^2)];
invent9=[inventory9(1)*inventory9(2)*.02253*.691,inventory9(1)*inventor
y9(2)*.04035*.691];
inv9=[invent9(1)^2*(inventory9(3)^2),invent9(2)^2*(inventory9(3)^2)];
invent10=[inventory10(1)*.07976*.691,inventory10(1)*.14224*.691];
inv10=[invent10(1)^2*(inventory10(3)^2),invent10(2)^2*(inventory10(3)^2
)];
invent11=inventory11(1)*inventory11(2)*.81823;
inv11=invent11^2*(inventory11(3)^2);
%%
%Level 1
% if time==1
%     %Uranium
% id(time,1)=in(1)-(out1(1)+output2(1)+output3(1)+out4);
% id(time,2)=0;
% id(time,3)=0;
% id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4;
%     %Plutonium
% id(time,5)=in(2)-out1(2)-output3(3);
% id(time,6)=0;
% id(time,7)=0;
% id(time,8)=ins(2)+o1(2)+o3(2);
% else
%     %Uranium
% id(time,1)=in(1)-(out1(1)+output2(1)+output3(1)+out4);%+id(time-1,1);
% id(time,2)=0;
% id(time,3)=0;
% id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4;%+id(time-1,4);
%     %Plutonium
% id(time,5)=in(2)-out1(2)-output3(3);%+id(time-1,5);
% id(time,6)=0;
% id(time,7)=0;
% id(time,8)=ins(2)+o1(2)+o3(2);%+id(time-1,8);
% end
%%
%Level 2
% if time==1
%     %Uranium
% id(time,2)=inventory3(2)+invent4(1)+invent5(1)+invent10(1);
% id(time,1)=in(1)-(out1(1)+output2(1)+output3(1)+out4)-id(time,2);
% id(time,3)=inv3+inv4(1)+inv5(1)+inv10(1);
% id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4+id(time,3);
%     %Plutonium
% id(time,6)=invent4(2)+invent5(2)+invent10(2);
% id(time,5)=in(2)-out1(2)-output3(3)-id(time,6);
% id(time,7)=inv4(2)+inv5(2)+inv10(2);
% id(time,8)=ins(2)+o1(2)+o3(2)+id(time,7);
% else
%     %Uranium

```

```

% id(time,2)=inventory3(2)+invent4(1)+invent5(1)+invent10(1);
% id(time,1)=in(1)+id(time-1,2)-id(time,2)-out1(1)-output2(1)-
output3(1)-out4;
% id(time,3)=inv3+inv4(1)+inv5(1)+inv10(1);
% id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4+id(time,3);%+id(time-1,3);
%      %Plutonium
% id(time,6)=invent4(2)+invent5(2)+invent10(2);
% id(time,5)=in(2)-id(time,6)+id(time-1,6)-out1(2)-output3(3);
% id(time,7)=inv4(2)+inv5(2)+inv10(2);
% id(time,8)=ins(2)+o1(2)+o3(2)+id(time,7);%+id(time-1,7);
% end
%%
% Level 3
if time==1
    %Uranium
    id(time,2)=inventory1(1)+inventory2(2)+inventory3(2)+invent4(1)+invent5
    (1)+invent6(1)+invent7(1)+invent8(1)+invent9(1)+invent10(1)+invent11;
    id(time,1)=in(1)-(out1(1)+output2(1)+output3(1)+out4)-id(time,2);
    id(time,3)=inv1+inv2+inv3+inv4(1)+inv5(1)+inv6(1)+inv7(1)+inv8(1)+inv9(
    1)+inv10(1)+inv11;
    id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4+id(time,3);
    % Plutonium
    id(time,6)=invent4(2)+invent5(2)+invent6(2)+invent7(2)+invent8(2)+inven
    t9(2)+invent10(2);
    id(time,5)=in(2)-out1(2)-output3(3)-id(time,6);
    id(time,7)=inv4(2)+inv5(2)+inv6(2)+inv7(2)+inv8(2)+inv9(2)+inv10(2);
    id(time,8)=ins(2)+o1(2)+o3(2)+id(time,7);
else
    % Uranium
    id(time,2)=inventory1(1)+inventory2(2)+inventory3(2)+invent4(1)+invent5
    (1)+invent6(1)+invent7(1)+invent8(1)+invent9(1)+invent10(1)+invent11;
    id(time,1)=in(1)+id(time-1,2)-id(time,2)-out1(1)-output2(1)-output3(1)-
    out4;
    id(time,3)=inv1+inv2+inv3+inv4(1)+inv5(1)+inv6(1)+inv7(1)+inv8(1)+inv9(
    1)+inv10(1)+inv11;
    id(time,4)=ins(1)+o1(1)+o2+o3(1)+o4+id(time-1,3)+id(time,3);
    %Plutonium
    id(time,6)=invent4(2)+invent5(2)+invent6(2)+invent7(2)+invent8(2)+inven
    t9(2)+invent10(2);
    id(time,5)=in(2)-id(time,6)+id(time-1,6)-out1(2)-output3(3);
    id(time,7)=inv4(2)+inv5(2)+inv6(2)+inv7(2)+inv8(2)+inv9(2)+inv10(2);
    id(time,8)=ins(2)+o1(2)+o3(2)+id(time,7);%;
end
%%
% Conversion to SITMUF series
%Uranium
if time==1
    a=0;
    sigv2=id(time,4);
    V=id(time,1);
else
    a=id(time,3)/(id(time,4)-a*id(time,3));
    sigv2=id(time,4)-((id(time,3)^2)/sigv2);
    if sigv2<0
        sigv2=id(time,4);
    end
end

```



```

        end
        V=a*id(time-1,1)+id(time,1);
    end
    %Plutonium
    if time==1
        b=0;
        sigx2=id(time,8);
        X=id(time,5);
    else
        b=id(time,7)/(id(time,8)-b*id(time,7));
        sigx2=id(time,8)-((id(time,7)^2)/sigx2);
        if sigx2<0
            sigx2=id(time,8);
        end
        X=b*id(time-1,5)+id(time,5);
    end
    %%
    %Simplified Page's Test on SITMUF & Define model outputs
    k=0.5;
    % Uranium

    Vsit=Vsit+(V/sqrt(sigv2))-k;
    %Plutonium

    Xsit=Xsit+(X/sqrt(sigx2))-k;
    if id(time,5)>=8
        t=[id(time,5);Xsit];
    else
        t=zeros(2,1);
    end
    y=[time;id(time,1);sqrt(id(time,4)).01;Vsit;id(time,5);sqrt(id(time,8)
);Xsit;t];
    else
        y=zeros(9,1);
    end
end

```

Appendix B. DOE Graded Safeguards

	Attractiveness Level	Pu/U-233 Category (kg)				Contained U-235/Separated Np-237/Separated Am-241 and Am-243				All E Materials Category IV
		I	II	III	IV	I	II	III	IV ¹	
WEAPONS Assembled weapons and test devices	A	All	N/A	N/A	N/A	All	N/A	N/A	N/A	N/A
PURE PRODUCTS Pits, major components, button ingots, recast able metal, directly	B	≥2	≥0.4<2	≥0.2<0.4	<0.2	≥5	≥1<5	≥0.4<1	<0.4	N/A
HIGH-GRADE MATERIALS Carbides, oxides, nitrates, solutions (≥25 g/L) etc.; fuel elements and assemblies; alloys and mixtures; UF ₄ or UF ₆	C	≥6	≥2<6	≥0.4<2	<0.4	≥20	≥6<20	≥2<6	<2	N/A
LOW-GRADE MATERIALS Solutions (1 to 25 g/L), process residues requiring extensive reprocessing; Pu-238 (except waste); UF ₄ or UF ₆ (≥ 20% < 50% enriched)	D	N/A	≥16	≥3<16	<3	N/A	≥50	≥8<50	<8	N/A

ALL OTHER MATERIALS Highly irradiated ³ forms, solutions (<1 g/L), compounds; uranium containing <20% U-235 or <10% U-233 ² (any form, any quantity)	E	N/A	N/A	N/A	Reportable Quantities	N/A	N/A	N/A	Reportable Quantities	Reportable Quantities
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