Safeguards modeling for studies of pyroprocessing input accountancy and fuel impacts on ID and SEID

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Introduction

Fast reactors capable of consuming actinides recovered from used nuclear fuel (UNF) are among some of the many advanced reactors under development. Pyroprocessing is a technology which lends itself well to actinide recovery to harvest fuel for fast reactors, as the end products are metallic and plutonium is never isolated, leading to a less desirable target for diversion and proliferation. However, pyroprocessing has never been commercially performed.

Input accountancy is a challenge in pyroprocessing. While destructive assay (DA) accountancy methods for safeguarding special nuclear material (SNM) are used in aqueous reprocessing with dissolved fuel assemblies (FAs), these are incompatible with the solid FAs entering pyroprocessing. Established nondestructive assay (NDA) techniques have not been able to deliver both rapid and accurate assessments of SNM.

A team of researchers at Idaho State University (ISU) on the Resonance Absorption Densitometry for Materials Assay Security Safeguards (RADMASS) project awarded to General Electric Vernova Advanced Research Center (ARC) and funded through ARPA-E ONWARDS is modeling a new method for input accountancy called dual isotope notch observer (DINO) detection. DINO detection may improve upon the time needed for low error measurement in NDA methods, rendering pyroprocessing a viable commercial enterprise. The ability of the new technology to meet safeguards requirements will be examined through the development and utilization of two pyroprocessing safeguards models: one for metallic fuels from sodium fast reactors (SFRs) and other for light water reactor (LWR) fuels.

Reprocessing is subject to safeguards for SNM as it separates the valuable actinides from fission products. Aqueous reprocessing achieves separation through chemical extraction while pyroprocessing uses electrochemical separation methods. There are presently no commercially operating reprocessing facilities in the United States (US), though there are efforts by a handful of companies to resume commercial reprocessing. All reprocessing facilities need to meet safeguards to prevent and detect diversion of SNM, but a clear regulatory framework for licensing a reprocessing facility under the US Nuclear Regulatory Commission (NRC) does not yet exist. The gap in regulations for reprocessing facilities in the US has been noted in SECY-09-0082 by the NRC in 2009 [1].

Safeguards pertinent to commercial reprocessing in the United States are assumed to include the guidelines for material control and accounting (MC&A) established in 10 CFR part 74, in addition to reprocessing regulations in 10 CFR parts 50 and 70 [3, 4, 5, 6]. These include the limits on inventory difference (ID), also known as material unaccounted for (MUF) internationally, and the standard error of inventory difference (SEID) [5]. ID and MUF are calculated from the difference in measured input and outputs while also accounting for internal gains and losses, and SEID is the standard deviation of all inventory measurements weighted by measurement errors [4].

NRC guidelines specify that Category 1 facilities must operate with the SEID under 0.1% of active inventory (AI) and the ID be no more than three times the value of the SEID with a 200 g limit for the plutonium ID [5]. Internationally, the IAEA specifies that the SEID, called σ_{MUF} in IAEA terms, not exceed 1 SQ of SNM which is 2.42 kg of plutonium and 25 kg of low-enriched uranium-235 [7]. If the error margins in input accountancy are too high, it is unlikely that pyroprocessing can meet these safeguards requirements on ID and SEID.

The IAEA has well-defined safeguards for reprocessing facilities, though all existing facilities perform aqueous reprocessing. It is projected that the NRC will adopt similar measures in clarifying the licensing of commercial reprocessing facilities in the US. There are ten traditional safeguards used at all reprocessing facilities in accordance with the IAEA safeguards agreement [8]. These ten approaches are: (1) “Material Balance Areas (MBAs) for nuclear material accounting”, (2) “Defined Key Measurement Points (KMPs) for measuring the flow and inventory of nuclear material”, (3) “Defined Strategic Points for containment and surveillance (C/S) and other verification measures”, (4) “Nuclear Material Accountancy, supported by review of operating records and state reports”, (5) “Annual Physical Inventory Verification (PIV) – typically a ‘shutdown cleanout’ inventory taking”, (6) “Verification of domestic and international transfers of nuclear material”, (7) “Statistical evaluation of the nuclear material balance to determine “Material Unaccounted for” (MUF)”, (8) “Routine, (monthly) interim inventory verifications (IIVs) for the timely detection of possible diversion of nuclear material”, (9) “Verification of facility design information”, and (10) “Verification of the operator’s measurement system” [8].

Approaches numbered 1, 2, 4, 7, and 8 are most relevant to safeguards modeling for the RADMASS project. Modeling pyroprocessing is an important step for investigating the capability of a facility to meet safeguards requirements under various fuel conditions such as variations burnup, reactor power, enrichment, and composition. Other parameters are being investigated including uncertainty values for input accountancy in DINO detection, facility throughput, actinide recovery rates, process times, and other measurement error values. The discovery of the most impactful parameters in safeguards compliance is useful in preparation for commercialization.

DINO Detection

DINO detection begins with a high intensity beam of photons of tunable energy and narrow bandwidth from a laser Compton scattering (LCS) source. When tuned to the energy of nuclear resonance fluorescence (NRF) for a particular isotope [9], the source becomes ideal for use in the NDA of nuclear material [9]. In pyroprocessing, there are two primary isotopes of interest: U-235 and Pu-239 with NRF energies of 1.73 MeV and 2.14 MeV respectively [10, 11].

Figure 1 depicts the DINO detection method geometry. The beam first encounters an interrogation object such as a fuel pin, which results in absorption of the NRF energy followed by fluorescence. UNF produces a high radiation field, which renders direct detection of the NRF of the interrogation target difficult. Instead, the diminished beam with a decrease in NRF energy continues through collimators and next impinges upon a witness foil made of the same isotope, which absorbs the same NRF energy and fluoresces. The amount of fluorescence from the witness pin directly correlates with that of the interrogation object. The semi-circular detector depicted in yellow in Figure 1 collects both the scattered and the NRF emissions from the witness foil. The total energy deposited is recorded. The method is then repeated with a non-resonant witness foil to provide a comparison and quantify the energy deposited without fluorescence in the witness foil [9]. The two measurements are used to produce a metric with a known relationship to the areal density of the resonant isotope. The relationship is produced by a calibration procedure using targets of known composition [12, 13]. The DINO detection method has been modeled in MCNP 6.2 with excellent agreement to the literature [9, 13].

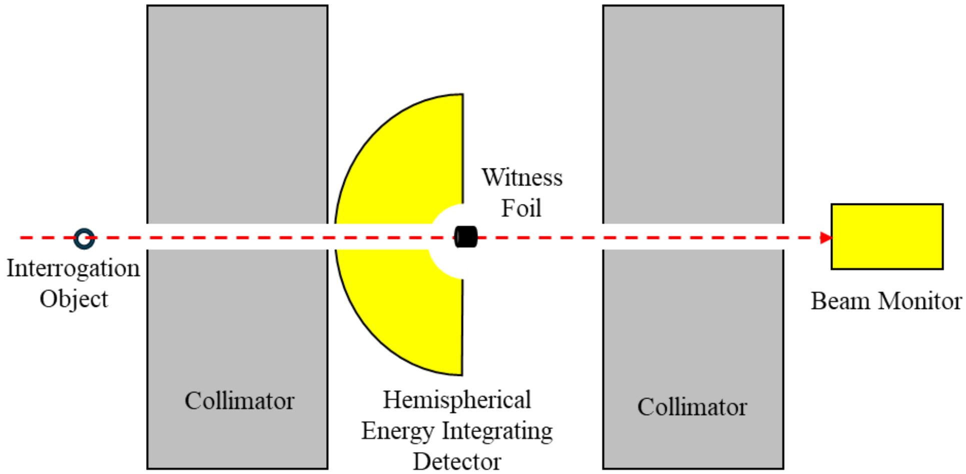

Figure 1 DINO Detection

A calibration curve is established with a function of areal density α, which is directly related to enrichment of the isotope. The calibration is performed separately for uranium and plutonium with their individual NRF energies. The error values for each isotopic interrogation are calculated, called fractional standard deviation (FSD).

It may be possible to interrogate an entire fuel assembly (FA) rather than single pins, depending on the strength of the LCS source [14]. When only representative fuel pins are interrogated from each FA, the used FA will first be subject to computed tomography (CT) to select representative pins for interrogation. CT is also capable of determining if any pins have been diverted [15]. After a CT scan, the FA will be disassembled so that the selected fuel pins may be used in the DINO detection system for mass measurements of U-235 and Pu-239. Masses of other isotopes will be inferred.

The literature suggests that 1% FSD in 300 seconds latency is feasible, where latency is the length of interrogation time. It is determined from the MCNP DINO model that to achieve the RADMASS target of 1% FSD in 300 seconds latency, the necessary LCS flux is on the order of $10^{11}\ \gamma/s$ for U-235 and $10^{12}\ \gamma/s$ for Pu-239 in a single SFR fuel pin. There is no developed source capable of these fluxes at the present time [16].

The interrogated and inferred contents of a FA, the latency and FSD values from DINO detection are used in the pyroprocessing safeguards modeling which is the core subject of this paper. The safeguards model can also be used to inform DINO modeling about the FSD uncertainties capable of meeting safeguards requirements, and the latency determined for those error values. The latency also has potential to limit facility throughput.

Pyroprocessing and Modeling

A pyroprocessing facility follows a defined process as exemplified by the process used in the Fuel Cycle Facility (FCF) at Idaho National Laboratory (INL), regulated by the Department of

Energy (DOE) rather than the NRC. The process is accompanied by process monitoring to protect and track SNM through use of input accountancy, mass balance measurements, and DA for actinide products produced. The mass information is used in determining ID and SEID for safeguards.

Pyroprocessing begins with the input of UNF to a hot cell. The fuel is examined, interrogated to determine isotopic content, and chopped in preparation for processing in an electrorefiner (ER). The ER is a high temperature bath of molten salts in which actinides are separated from fission products by the collection of actinides on cathodes. Preparation for electrorefining differs for oxide and metallic fuels. Metallic fuels from an SFR may be chopped with their cladding and placed directly in anode baskets for electrochemical separation. Oxide fuels from an LWR necessitate the removal of cladding and reduction of the chopped oxide fuels to metal, as electrorefining operation is dependent on the metallic state of the materials separated.

The remaining steps of pyroprocessing are independent of the fuel type. Uranium (U) and mixed uranium and transuranic (U/TRU) products may be recovered on cathodes in the ER with the contents of the cathodes then collected and processed in cathode processors to remove accompanying salts. Some uranium may also be reserved from uranium cathodes for oxidant production to maintain the correct balance of actinides to maintain actinide recovery in the ER. The noble metals remain undissolved in the anode baskets and are collected as waste. Lanthanides and active metal wastes in salt treatment processes are removed to purify the salt for reuse in the ER.

There are multiple safeguards measures implemented in a reprocessing facility but not all are needed for the safeguards modeling. Safeguards relevant to the models include the monthly interim balance verifications, also referred to as balance periods or inventory periods. These are set to 30 days, the longest permitted by IAEA regulations, in the absence of clarity from the NRC. Existing fuel facilities regulated by the NRC have inventory assessments up to every two months or as infrequently as annually [2]. Material balance areas (MBAs), the zones within a plant where the nuclear material leaving a zone is compared against the inputs to the zone for tracking, are implied within the model. MBAs are established with key measurement points using mass balance measurements. These measurements are also used in calculating ID and SEID for the monthly inventory checks.

The safeguards models are built in SimEvents, a subset of MATLAB/Simulink capable of modeling bulk movement of materials, as is appropriate for pyroprocessing. Pyroprocessing is simulated with a process model and data collected from a simulated monitoring system. There are two dominant versions of the model: one for metallic fuels from SFRs and the other for oxide fuels from traditional LWRs. Subsets of these versions are adjusted for specific throughputs.

In lieu of physical DINO detection, the contents of UNF are derived from depletion analysis performed in SCALE 6.3.1 using evaluated nuclear data file version 8 (ENDF 8) for SFR fuel modeled on driver fuel fabricated for the Fast Flux Test Facility (FFTF), followed by a year of decay. For testing LWR fuels, the depletion analysis is performed for standard Westinghouse pressurized water reactor (PWR) and General Electric boiling water reactor (BWR) fuels. The FSD error values from DINO modeling and the material content from depletion work becomes the inputs for the safeguards models. The models can also be used to inform DINO system FSD and latency values compatible with pyroprocessing safeguards compliance for various throughputs.

The model in Figure 2 follows a flowchart amended for metallic fuels for the Fast Reactor Safeguards Pyroprocessing Performance Model (FRPSPM), built with the assumption that the SFR fuel could be processed similarly to the pyroprocessing facility designed by Argonne National Laboratory (ANL) for processing LWR fuels [17]. Figure 2 shows the process model for the FRPSPM with boxes representing process subsystems. The UNF input is in the upper left-hand corner, while outputs are on the right-hand side of the diagram. The ER is the largest block near the center. The salt recycling subsystems are lower and to the left of the ER.

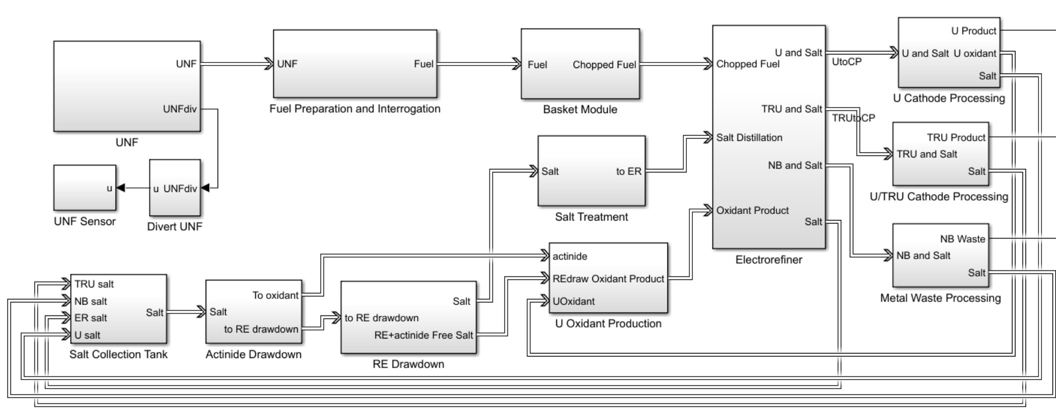

Figure 2 Fast Reactor Pyroprocessing Safeguards Performance Model

The Light Water Reactor Pyroprocessing Safeguards Performance Model (LWRPSPM) seen in Figure 3 follows an ANL flowchart for oxide pyroprocessing [17, 18, 19]. It contains the additional steps mentioned previously, including the removal of cladding during fuel preparation and the use of an electro-reducer to convert oxide fuel to a metallic state. There are other significant differences in processing the two fuel types including size of fuel assemblies, the mass allowed in the anode baskets due to criticality considerations, and lower enrichments of LWR fuels. The LWRPSPM is not yet optimized for use in parametric studies.

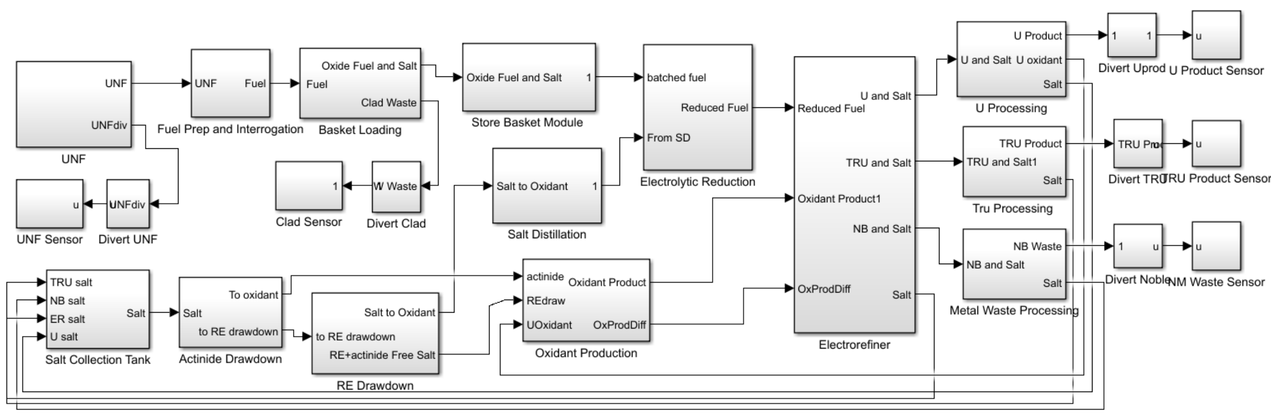

Figure 3 Light Water Reactor Pyroprocessing Safeguards Performance Model

Monitoring systems accompany the process models shown in Figures 2 and 3, simulating measurements, gathering data in MATLAB from the process model, and outputting data by balance period with uncertainties. The monitoring system compiles data from inputs, outputs, wastes, and ER inventory, and returns the amount of material in a particular process at the close of each balance period. It then calculates process inventory, ID, and SEID for both U-235 and plutonium.

Table 1 displays example values calculated across twelve 30-day balance periods of continual operation in the FRPSPM. The annual throughput modeled is 25 MT annually, with fuel depletion for a reactor power of 120 MW/MTU, and burnup of 20 at.% which is analogous to 188 GWd/MTHM in terms of LWR burnup. Note that the process inventory increases with each month due to the accumulation of unrecovered actinides in the ER.

Table 1 Example Calculations from the FRPSPM (kg)

Inv Period	U235 Inv	U235 ID	U235 SEID	Pu Inv	Pu ID	Pu SEID
1	21.68	-2.701	1.608	24.51	-0.223	2.113
2	22.18	-2.758	1.607	25.12	0.147	2.114
3	22.88	-2.429	1.610	25.83	-0.233	2.114
4	23.44	-2.827	1.610	26.35	0.245	2.110
5	24.12	-2.959	1.608	26.98	-0.324	2.109
6	24.73	-3.015	1.609	27.79	-0.223	2.113
7	25.27	-2.740	1.607	28.23	-0.093	2.109
8	25.84	-2.559	1.607	28.97	-0.204	2.109
9	26.46	-2.050	1.608	29.59	-0.167	2.109
10	27.10	-2.729	1.607	30.29	-0.334	2.111
11	27.81	-3.245	1.605	30.94	0.015	2.109
12	28.41	-2.647	1.606	31.54	-0.325	2.109

The inventory values reported in Table 1 represent the materials in processing at the close of a balance period and exclude the materials in various storage locations within the hot cell. Active inventory is taken to be the sum of both process and hot cell storage inventories. The plutonium ID is typical of expectations, averaging near to 0 with both positive and negative values appearing. A negative ID indicates that more material exited the facility than entered and when consistently negative as it is in the U-235 ID column of Table 1, reflects an area for modeling accountancy improvement. However, the model is used for comparative values in parametric studies, so the current state of the model meets the purposes of this work. The magnitude of the plutonium ID values shown in Table 1 exceed 200 g in many of the balance periods. If the 200 g limit indeed applies in domestic reprocessing, such could precipitate a plant shutdown for recovery of plutonium for accountancy. This could necessitate a time-consuming flushout of the ER.

The throughput of a facility is dependent on the duration of uninterrupted operations, so the results in Table 1 assuming continual operations represent an upper bound for ID and SEID for any inventory period. It becomes an upper limit for a plant such that ID and SEID never exceed the values calculated for continual throughput if plant conditions are modeled accurately. Continuous operation is not practical due to safeguards compliance and plant maintenance. Monthly inventory checks, annual physical inventory checks, and maintenance and equipment failures all contribute to downtime. Past pyroprocessing safeguards modeling and planning from ANL assumed 200 days of annual operation [18, 17]. The FCF also operates on this scale [20]. The Korean Atomic Energy Research Institute (KAERI) in South Korea assumes 250 days of operation [20]. The throughputs listed in Table 2 are calculated for 200 days of operation despite the continual modeling performed.

There are many other assumptions incorporated into the FRPSPM results presented. It is assumed that one SFR assembly per anode basket is adequate for criticality safety [21]. Up to 8 anode baskets may be loaded per ER according to ANL [17]. Transfers of material were allotted 30 minutes for removal and replacement elsewhere though the actual time may be lower in some cases. The salt treatment steps were assumed to be equivalent for LWR and SFR fuels [17].

The FRPSPM has multiple operational throughput versions, in recognition that different throughputs in pyroprocessing are supported by different numbers of internal elements such as ERs, cathode processors, and salt processing equipment. The process times for the different FRPSPM throughputs were adjusted accordingly to reflect the number of pieces of equipment in use. The calculations of ID, SEID, and active inventory are affected by the number of process elements and the errors accompanying multiple replications of process elements.

The numbers of pieces of equipment required to keep an uninterrupted pace in pyroprocessing by throughput are identified in Table 2. The pace is set by the number of FAs per day in the second column. The abbreviations are as follows: ER means electrorefiner, CP means cathode processors, and DD represents the drawdown vessels used in salt purification.

Table 2 Number of process stations for varying throughputs

Throughput	FA/day	ERs	U CPs	U/TRU CPs	DD
25 MT	2	1	2	2	2
50 MT	4	2	3	3	3
75 MT	6	3	4	5	5
100 MT	8	3	5	6	6

In different terms, if future SFR plants have the generating capacity of the FFTF, three ERs are capable of processing UNF from up to 32 SFRs for 100 MT throughput. It is therefore unlikely an SFR pyroprocessing capacity beyond 100 MT will be needed. Larger throughputs for modeled facilities have higher ID and SEID values than smaller throughputs.

Results and Discussion

Studies of fuel parameters have been performed for various conditions. The first parameters investigated with the FRPSPM for safeguards compatibility are the fuel burnup, power, enrichment, input accountancy error values, output and ER inventory DA error values, and throughput. Burnups were 5 at.%, 10 at.%, 15 at.%, 20 at.%, and 25 at.%. Note that 1 at.% corresponds to about 9.4 GWd/MTHM. Modeled reactor powers were 100, 110, 120, and 130 MW/MTU. Uranium-235 weight enrichments tested for SFR fuels were 10%, 17.775%, and 20%. Throughputs of 25 MT, 50 MT, and 100 MT have been studied. Input accountancy and output error values were adjusted to find the values needed for safeguards compliance for the various fuel inputs and throughputs.

Results have been inspected for safeguards compliance. The RADMASS program goal is that the SEID $< 0.1\%$ of AI from the rule from the NRC for Category 1 facilities, and that the ER is not flushed out for accountancy more than once annually. Related NRC safeguards are that the plutonium ID remains under 200 g and the ID does not exceed 3 times the SEID [5]. AI has been defined by the NRC for licensed facilities as the sum of SNM in beginning inventory, ending inventory, additions to inventory, removals from inventory after all common terms are excluded [22]. Excluding all common terms in pyroprocessing leaves the active inventory as the sum of all material still in process at a given point in time. The IAEA includes all radioactive material present at a nuclear facility in their inventory assessments, so for pyroprocessing this would include the SNM outside the hot cell in both FAs on site and actinide ingots removed to a storage vault on site [23]. It is unclear whether the inclusion of either internal hot cell storage or SNM outside the hot cell is permissible in AI calculations by the NRC [22].

For this analysis, the facility AI included materials stored within the hot cell awaiting the next step in processing. This includes stored anode baskets awaiting the ER, cathodes with uranium and mixed U/TRU awaiting processing, and wastes containing actinides. The sum of all U-235 in the hot cell varies between 1000-7800 kg, while plutonium AI content ranges from 500-1900 kg depending predominantly on the burnup of the fuel.

The plutonium ID limit is explained in 10 CFR § 74.53 for research activities as "investigate any difference greater than 200 grams of plutonium or U-233 or 300 grams of U-235 that exceeds three times the estimated standard error of the inventory difference estimator" [24]. The plutonium ID limit elsewhere in 10 CFR part 74 indicates that either condition could trigger an investigation, as "U^{235} inventory difference ≥ 3 SEID and 200 grams of Pu/U^{233} or 300 grams of U^{235} (HEU)" [5]. For this work both interpretations are considered. Exceeding the ID limit for plutonium will prompt flushouts in the ER for accountancy so regulatory clarification will be needed.

Table 3 contains data from the study of enrichment effects on safeguards. It displays the maximum values calculated for process inventory, ID, and SEID across a year of simulated data for SFR fuels with a 20 at.% burnup at 120 MW/MTU, in the 100 MT throughput model, using 1% error values for all measurements of input accountancy, outputs, and process monitoring. These data show that if there is more U-235 there is less plutonium, and vice versa because lower U-235 enrichment means more U-238 is available for transmutation into plutonium with the effect more pronounced for high burnups.

Table 3 Maximum values from enrichment impact study (kg)

Enrichment	U235 Inv	U235 ID	U235SEID	Pu Inv	Pu ID	Pu SEID
10 %	19.247	-3.3981	2.084	87.351	2.724	10.850
17.775 %	59.322	-10.483	6.428	67.916	2.163	8.439
20 %	76.274	-13.482	8.261	61.883	1.957	7.692

The plutonium input accountancy is the single greatest limiting factor in safeguards compliance, and other measurement errors were found to have negligible effect. The target input accountancy error, or FSD, has been determined to be 0.38% for U-235 and 0.19% for plutonium across all powers, burnups, and enrichments for 100 MTHM throughput for the primary recovery rate and chosen AI capacity. Lower burnups have more U-235 and less plutonium content, while higher burnups have less U-235 and more plutonium. The power in a reactor affects material content only slightly: higher power means slightly more plutonium and less U-235 and vice versa. Varying enrichments for SFR fuels results in the same pattern. Even changing the throughput of a facility does not alter the needed DINO error values if hot cell storage areas are scaled with the throughput. However, because the AI value increases proportionally alongside the SEID, the determined error value for meeting input accountancy does not change.

To achieve NRC safeguards compliance with SEID <0.1% AI, inclusion of storage within the hot cell is essential in defining AI, though the determination of storage capacity for this purpose seems disingenuous. The required input accountancy error values could be addressed in multiple ways. One option is to extend the dwell time for DINO detection to lower the FSD to the desired values. The DINO detection time for an FSD of 0.38% for U-235 is about 14 hours using current LCS technology. Over 55 hours are needed to obtain 0.19% for plutonium accountancy. This slows input accountancy down too far for commercial viability. Another option is to quadruple the storage within the hot cell, thereby increasing the needed DINO detection FSD value for plutonium to 0.76% and uranium up to 1.5%. These measures may not be necessary if facility inventory outside the hot cell is included in active inventory.

At present, there is no LCS source capable of producing the targeted intensity. If there were an LCS source capable of allowing 1% FSD with 300 second latency as desired by the RADMASS program, the FSD of 0.38% for U-235 would need 35 minutes latency and the 0.19% FSD for plutonium accountancy would require about 2.3 hours latency. These time requirements would limit the throughput for a metallic fuels pyroprocessing facility to approximately 25 MT annually unless multiple DINO detection devices are used. However, these input accountancy error values were found for a hypothetical AI size and the regulatory requirements regarding AI are unclear. If the SNM both inside and outside the hot cell is considered AI as is the case for IAEA inventory, then meeting the SEID < 0.1% AI requirement is far less problematic for any throughput.

Study results show that the UNF FA plutonium composition and accountancy matter more for IAEA safeguards compliance with an SEID ceiling of 2.42 kg Pu than it does for the NRC's SEID < 0.1% AI regulation. However, if the NRC confirms that an ID over 200 g of Pu prompts a shutdown for accounting and plutonium recovery in reprocessing, the throughput and plutonium ID become relevant in the NRC regulations. One possibility to lower the plutonium ID is shortening the balance periods. Improved plutonium accountancy may be needed for any throughput to keep the plutonium ID below 200 g consistently, although the 25 MT and 50 MT throughputs achieve this limit more often for low burnups containing less plutonium. Since high burnups are the target in running SFRs, this greatly affects the feasibility of pyroprocessing with metallic fuels. Pyroprocessing with oxide fuels from LWRs may meet this safeguards requirement more easily.

Ongoing Work

FRPSPM studies continue with recovery rates and process times with the understanding that by increasing the recovery rate the process time similarly increases [25]. Increasing recovery rates may also yield more attainable input accountancy error values. A more rigorous assessment of recent results for enrichment, power, burnup, and throughputs is underway. This work will support continued DINO modeling work and the development of a digital twin by the ARC.

The LWRPSPM, once optimized, will be used to investigate effects on SEID from varying PWR and BWR fuels of multiple enrichments and the other parameters studied with the FRPSPM. It is unclear at this time whether LWR fuels would present the same regulatory difficulties given the different fuel enrichment, plutonium content, and FA size.

The effects of the parameters studied will be quantified to identify those most impactful on meeting safeguards requirements. These parameters will be used towards the development of a figure of merit (FOM) indicative of commercial pyroprocessing viability.

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