Systems Analysis of hiRX for Improved Reprocessing Safeguards

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Abstract

The goal of this work was to examine the potential safeguards improvement of the High Resolution X-ray (hiRX) technology for aqueous reprocessing plants. Existing plants utilize K-Edge and Hybrid K-Edge Densitometry for routine accountancy measurements for the key input and output accountability tanks. The IAEA also use these technologies for verification measurements. The hiRX technology has the potential to significantly reduce the measurement uncertainty with an equally simple but less costly instrument that uses a much smaller sample size. Such a technology could be highly desirable for both the operator and inspector. This work utilizes a Separations and Safeguards Performance Model of a PUREX reprocessing plant to determine the safeguards improvement offered by the reduced measurement uncertainty of hiRX. Results show that a significant improvement in the ability to detect a protracted diversion may be possible.
Acknowledgement

This work was funded through the Next Generation Safeguards Initiative’s Safeguards Technology Development subprogram under the Office of Nonproliferation and International Security of the National Nuclear Security Administration. The authors would like to acknowledge Shirley Johnson for her review and insights into this work.
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<th>Description</th>
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<tbody>
<tr>
<td>C/S</td>
<td>Containment/Surveillance</td>
</tr>
<tr>
<td>CoK</td>
<td>Continuity of Knowledge</td>
</tr>
<tr>
<td>DA</td>
<td>Destructive Analysis</td>
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<tr>
<td>hiRX</td>
<td>High Resolution X-ray</td>
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<td>HKED</td>
<td>Hybrid K-Edge Densitometry</td>
</tr>
<tr>
<td>HRGS</td>
<td>High-Resolution Gamma-Ray Spectrometry</td>
</tr>
<tr>
<td>IAEA</td>
<td>International Atomic Energy Agency</td>
</tr>
<tr>
<td>ID</td>
<td>Inventory Difference</td>
</tr>
<tr>
<td>IDMS</td>
<td>Isotope Dilution Mass Spectrometry</td>
</tr>
<tr>
<td>IIV</td>
<td>Interim Inventory Verification</td>
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<tr>
<td>ITV</td>
<td>International Target Values</td>
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<tr>
<td>KED</td>
<td>K-Edge Densitometry</td>
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<tr>
<td>KMP</td>
<td>Key Measurement Point</td>
</tr>
<tr>
<td>LANL</td>
<td>Los Alamos National Laboratory</td>
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<tr>
<td>MBA</td>
<td>Material Balance Area</td>
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<td>MTHM</td>
<td>Metric Tons of Heavy Metal</td>
</tr>
<tr>
<td>MOX</td>
<td>Mixed Oxide</td>
</tr>
<tr>
<td>MUF</td>
<td>Material Unaccounted For</td>
</tr>
<tr>
<td>NCC</td>
<td>Neutron Coincidence Counter</td>
</tr>
<tr>
<td>NDA</td>
<td>Non-destructive Analysis</td>
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<tr>
<td>NMA</td>
<td>Nuclear Material Accountancy</td>
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<tr>
<td>NRTA</td>
<td>Near Real Time Accountancy</td>
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<tr>
<td>OSL</td>
<td>On Site Laboratory</td>
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<tr>
<td>PIT</td>
<td>Physical Inventory Taking</td>
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<td>PIV</td>
<td>Physical Inventory Verification</td>
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<td>PUREX</td>
<td>Plutonium and Uranium Extraction</td>
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<tr>
<td>QA/QC</td>
<td>Quality Assurance/Quality Control</td>
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<tr>
<td>RE</td>
<td>Random Error</td>
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<tr>
<td>RRP</td>
<td>Rokkasho Reprocessing Plant</td>
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<tr>
<td>SE</td>
<td>Systematic Error</td>
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<tr>
<td>SMMS</td>
<td>Solution Measurement and Monitoring System</td>
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<td>SMS</td>
<td>Solution Monitoring System</td>
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<tr>
<td>SRD</td>
<td>Shipper-Receiver Difference</td>
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<tr>
<td>SSPM</td>
<td>Separation &amp; Safeguards Performance Model</td>
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<tr>
<td>THORP</td>
<td>Thermal Oxide Reprocessing Plant</td>
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<tr>
<td>UREX</td>
<td>Uranium Extraction</td>
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<td>XRF</td>
<td>X-ray Fluorescence</td>
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1.0 Introduction

Nuclear Material Accountancy (NMA) measurements and inspector verification of those measurements can be very challenging in a reprocessing facility due to the large number of chemical and physical forms, and complex matrices in the process. Analytical measurements can require significant resources both in equipment costs and inspector/analyst time. Any new measurement technology being considered should reduce the uncertainty of the measurement and the cost, utilize easy-to-operate equipment, and limit the burden of maintenance and calibration.

The high resolution X-ray (hiRX) technology may provide significantly improved measurements of nuclear material accountancy and verification samples at reprocessing plants [1]. This technology uses a low energy X-ray generator with doubly curved optics to create a monochromatic X-ray source for excitation along with a doubly curved optic for collection of an X-ray emission unique to the element(s) of interest. The result is a spectrum with a single peak and near zero background. The measurement does not require moving parts, other than those needed to change out samples. Microliter sized samples are used which reduces the waste produced by the analytical lab. The smaller X-ray source leads to lower instrument costs as compared to Hybrid K-Edge Densitometry (HKED), which is the leading state of the art.

The purpose of the work being reported here is to determine an improvement in diversion detection thresholds in a safeguards system that uses the hiRX technology as compared to current practices. Because a facility’s NMA system and the IAEA’s safeguards approach for reprocessing plants may require a large and varied number of measurements and monitoring systems, a systems-level approach is required to analyze the overall improvement of a new technology. The Separation and Safeguards Performance Model (SSPM) is used to perform this analysis.

The SSPM tracks the material flows through a PUREX reprocessing plant and simulates the NMA systems. Diversion scenarios are added to the model to determine detectability using current measurement systems as compared to using the hiRX technology. The detection probability as a function of the diversion fraction is examined. The results are presented along with various implementation options.
2.0 Background

The goal of an operator’s NMA system and its verification by inspectors/regulators is to provide assurance that all nuclear material is accounted for and that no material has been lost or diverted. For domestic safeguards, the plant operator and national regulatory agency are concerned about potential process upsets or diversion of material by an individual or sub-national group. For international safeguards, the verification of the operator’s NMA system provides assurance that the host country is not diverting material for non-peaceful purposes and that the facility is not being misused. Regardless of the user, accountancy and verification add cost and burden to both the plant operator and regulatory agencies. The following provides an overview of current NMA and verification practices in existing commercial reprocessing plants.

2.1 Current Reprocessing Safeguards

Reprocessing plants are divided into Material Balance Areas (MBAs) over which a mass balance can be performed. Although uranium (U) and plutonium (Pu), are the primary nuclear materials to be measured and verified, there may also be other elements of interest, such as curium and neptunium. A large reprocessing facility may have from 3 to 5 MBAs. The first MBA normally contains the spent fuel receipt and storage areas, the head-end mechanical cells, and dissolution and clarification processes. Item accountancy is used in the front end for assemblies from receipt to dissolution, with their U and Pu quantities based on the reactor’s calculated declarations. Some neutron and gamma-ray spectrometry measurements can be used to monitor the assemblies and may provide an estimation of quantities of nuclear material present. The material balance differences over MBA 1, referred to as the Shipper-Receiver Difference (SRD), are usually due to the large uncertainties associated with the reactor burn-up calculations. After dissolution of the sheared fuel, samples are taken from the input accountability tank where the first accurate determination of the nuclear material is made. Volume and density are determined by in-tank dip-tube systems with electromanometers.

MBA 2 may contain the bulk of the separation and purification processes. All currently operating reprocessing plants utilize the PUREX separation process. After separation and purification, the U and Pu are measured in accountability tanks for transfer to the conversion process in MBA 3. These tanks are sampled for U/Pu analyses, and in-tank dip tube systems with electromanometers are used to determine volume and density. If the oxide conversion process for either or both U and Pu are also in MBA 2 then there would be no accountability tank and the accountancy measurements and verification for the inventory change would be done on the product containers. Prior to transfer to storage, U and Pu oxides may be measured and verified by either sample taking and destructive analyses (DA), or by non-destructive assay (NDA) methods. The containers and content are also weighed. Inspector samples may be analyzed in an On Site Laboratory (OSL), if available, or shipped to a central laboratory. Waste treatment and storage areas may be in MBA 2 or in a separate MBA, depending on the agreed accountancy structure.

The U, Pu, and mixed oxide (MOX) product storage areas make up the last MBA. Surveillance systems may monitor all activity in the storage areas and/or the containers may be placed under seal to maintain continuity of knowledge (CoK). If containment/surveillance (C/S) fails, or on a
random basis, inspectors may take samples from the product containers in storage. However, access is often difficult for sample taking, and NDA is preferred for inventory verification.

**Euratom Reprocessing Facilities**

Reprocessing facilities in the European Union are under Euratom safeguards. Although the IAEA and Euratom safeguards approaches for reprocessing facilities may differ, there are similar technological methods being implemented by Euratom for accountancy and safeguards procedures in the La Hague (France) and Sellafield (UK) plants [2]. The two processing facilities at La Hague, UP2 and UP3, are both able to process 800-1000 metric tons of heavy metal (MTHM) of spent fuel per year. The Thermal Oxide Reprocessing Plant (THORP) facility at Sellafield has a capacity of 900 MTHM/yr. The OSL analysts at these plants use radiometric and Hybrid K-edge (HKED) methods to perform elemental and isotopic measurements. Other measurements include the determination of weight of powders, and density and volume of solutions. Some samples are sent to the European laboratory, TUI in Karlsruhe, Germany, for analyses.

The operator measures material flows and inventories at Key Measurement Points (KMPs) at MBA boundaries and within the process. The flow measurement points are the inputs and outputs to a MBA. The measurements are recorded and periodically declared and verified by inspectors. Once per year, a physical inventory taking is used to calculate an overall material balance, and the material unaccounted for (MUF) value is calculated. During normal operation the MUF value should fall within the limits of the combined measurement errors. In addition, the operator has implemented Near Real-Time Accountancy (NRTA) to determine plant balances more frequently using input and output measurements as well as estimates, models, and measurements of in-process inventory.

Samples from the input accountability tank are measured using Hybrid K-Edge Densitometry (HKED). This method is a combination of K-Edge Densitometry (KED) and X-ray Fluorescence (XRF). KED is used to measure the highest concentration of U or Pu at values from 25 g/L up to 400 g/L. Since the Pu concentration is much lower than the U concentration (on the order of 1:100) XRF technology is used to provide more accurate Pu measurements. HKED is used for every batch accountability measurement. Measurement uncertainties achieved using HKED on the dissolver solution are about 0.2\% for U and 0.6\% for Pu. The International Target Values (ITV) 2010 uncertainties for HKED are $\sigma_r=0.2\%$, $\sigma_s=0.2\%$ for U and $\sigma_r=0.8\%$, $\sigma_s=0.5\%$ for Pu in the dissolver solution [3]. The U and Pu nitrate solutions that are produced after separation are measured at the output accountability tanks using KED. ITV uncertainties for KED are $\sigma_r=0.2\%$, $\sigma_s=0.2\%$ for U and $\sigma_r=0.3\%$, $\sigma_s=0.3\%$ for Pu in the product tanks [3].

Gamma and mass spectrometry are used to measure the U and Pu isotopic composition. The results taken from these methods are used to characterize the calibration solutions for the HKED tests. Although only used for approximately 10\% of the samples, analytical techniques such as Isotope Dilution Mass Spectrometry (IDMS) are used to provide a quality assurance check on the concentration of U and Pu within a sample. Using mass spectrometry, the OSLs have shown capabilities of attaining concentration measurement uncertainties below 0.1\%; however, the
process is labor intensive and therefore cannot be used for all batches. It is mainly used for calibration, to resolve operator-inspector differences, and to determine uranium enrichment.

Computer systems are used to trigger scheduled samplings of the fuel which are tracked and recorded from entry to storage [2]. Automatic sampling of the fuel is scheduled throughout the chemical separation process but additional samples can be requested randomly by plant operators and/or inspectors. Automatically generated accountancy reports which provide details on the dissolver solution samples and their movement through the plant are used to help derive the U and Pu concentrations in the system. From 2005 to 2009, the OSL analyzed between 500 and 800 samples at La Hague annually using various techniques while the OSL analyzed between 140 and 220 samples at Sellafield annually.

**Rokkasho Reprocessing Facility**

With a throughput of 800 MTHM/yr, the Rokkasho Reprocessing Plant (RRP) is of similar size to the European plants, and a design duplicate of UP3 at La Hague [4-6]. Figure 1 depicts the RRP accountancy structure. To enhance IAEA accountancy verification activities at KMPs, additional assurance measures, such as process monitoring, are applied at Other Strategic Points (OSPs). The OSP measures also provide confirmation of the operational status of the facility and that it is as declared. The IAEA has implemented a number of advanced technical methods and systems, many specifically designed for RRP. These include installed, unattended measurement and monitoring systems, and inspection procedures, such as short notice inspector visits to agreed in-process areas. Monitoring systems are installed throughout the facility from receipt of spent fuel to the transfer of product material to storage.

![IAEA accountancy structure for RRP](image-url)
The joint-use IAEA-State OSL primarily employs HKED, XRF, and IDMS to determine the concentrations of the U, Pu, and Cm in independently taken inspector samples. HKED is the primary measurement system, with 256 samples being analyzed in 2008. IDMS is used on a random selection of samples in order to complement the HKED measurements. Due to sample preparation requirements IDMS results may take up to 10 working days, whereas HKED results can be available in 1 day, although with higher uncertainties [7].

The Solution Measurement and Monitoring System (SMMS) provides highly accurate solution level measurements for the determination/verification of solution volumes and densities in all major process vessels. The SMMS also provides continuous monitoring of solution flows and added assurance to the verification of inventory changes and inventory. The SMMS-1 that is installed on the 12 most important process vessels are inspector owned and controlled, and use high-accuracy electromanometers which can attain a measurement uncertainty of ±0.05%. The joint-use SMMS-2 provides solution measurements on 80 less-important vessels. The Solution Monitoring System (SMS) is a specifically developed piece of software that is used to collect, process, and analyze the data from the SMMS.

2.2 hiRX Technology

The hiRX technology is based on monochromatic wavelength dispersive X-ray fluorescence which uses an X-ray optic technology to generate monochromatic excitation of the specimen and another optic to selectively collect the emitted X-rays of the target elements and direct them to the detector [1]. The result is a spectrum with a peak specific to one element with negligible background. Multiple optics can be used on the same instrument to detect up to four different elements.

Initial goals with hiRX technology were to attain a sensitivity of 10 ppm for Pu detection, improve safety and accuracy of Pu measurements, and develop new sampling methods for reducing sample size. Experiments performed with hiRX technology at Los Alamos National Laboratory (LANL) in recent years have exceeded expectations as hiRX capabilities have consistently attained better than 10 ppm detection limits and with smaller sample sizes than anticipated (200 microliters or less). From these tests, it is expected that hiRX can improve both the input and output accountability measurements to within an error of 0.1% with a rapid turn-around-time. This would be a 6-7 fold improvement in measurement uncertainty for Pu accountability in the dissolver solution and a 3 fold improvement in measurement uncertainty for the Pu nitrate product. Although hiRX has not yet been deployed to currently operating reprocessing plants, development of laboratory-based instruments for hiRX is in its initial phase and a field prototype unit is under development.

Recent testing has examined the performance of hiRX using surrogate spent fuel dissolver solutions [1]. The surrogate solution contained U at 10 g/L, Pu at 0.03 g/L, and 50 other elements at 0.1 g/L. The PuLa line at 14.279 keV was clearly seen with very low background.
3.0 SSPM Modeling Overview

The Separation and Safeguards Performance Model (SSPM) [8,9,10,11] is a transient reprocessing plant model developed at Sandia National Laboratories for safeguards analysis. Various versions of the model exist to model UREX+, PUREX, and Electrochemical reprocessing plant designs. The SSPM is constructed in MATLAB Simulink and tracks cold chemicals, bulk fluid flow, solids, and mass flow rates of elements 1-99 on the periodic table. The number of tracked species can be expanded to include chemicals or specific isotopes as needed.

The SSPM contains a number of capabilities including:

- A spent fuel source term library for user-defined runs with varying initial enrichment, burnup, and cooling time.
- Heat load and radioactivity tracking at any location in the plant.
- Customizable measurement points with user-defined errors.
- Automated calculation of inventory difference (ID) or material unaccounted for (MUF) and error propagation in real-time, including statistical modules for determining alarm conditions.
- User-defined diversion scenarios.
- Integration of process monitoring data, material measurements, material accountancy procedures, and physical security data for complete plant monitoring.

For this work, the SSPM was modified to create a PUREX plant model. This model includes the front end processes that make up MBA1 and separation through pulsed columns that make up MBA2. Oxide product storage is not currently included in the model. Figure 2 shows the overall plant model in Simulink. The front end of the plant is very similar to past work on the UREX+ model [10]. The focus of this work was on the materials accountability over MBA2, so MBA1 was not examined other than to ensure that the model was operating as expected.

The model is based on the assumption of a 2000 MTHM/yr plant; however, future work will reduce the plant size to 1000 MTHM/yr in order to be more consistent with existing reprocessing plants. Since the diversion scenarios examined for this report were based on diverting certain percentages of the flow streams, the results should not change in moving to a smaller plant model—the diversion times would just be twice as long, but detectability should be the same. This will be discussed further in the results section.

The Simulink model uses signals to represent the material flows between processing units. The processing units are subsystems that perform math functions on the signals to represent the operation. The subsystems simulate tanks filling and emptying, and determine how the material separates into the output streams. Separations are hard assumptions that do not currently have chemistry modeling included. The inventory of material in each processing unit is tracked.

The blue blocks shown in Figure 2 are measurement blocks that simulate what is being measured, the uncertainties, and the frequency of the measurement. Additional measurement points, not visible in the high-level view, are included in the model. The simulated
measurements are used within the model to perform periodic inventory balances. A Page’s Test is used to set alarm conditions. The Page’s Test is a well-accepted statistical test for detecting both abrupt and protected diversions for reprocessing safeguards [12,13]. The test can be tuned to reach an appropriate balance between sensitivity and false alarm probability. Earlier work was reported in 2011 on the use of Page’s Test in the Simulink model to do material balance evaluations [8]. This past work examined a simplified Page’s Test that produces acceptable results without significantly slowing down the model run time. A Pu balance has been the focus of the work to date for simplicity, but future work can examine the U balance as well.

The red blocks shown in Figure 2 are diversion points that can be turned on or off to represent material loss. The user can define when the diversion starts and stops, and the fraction of material removed. In this manner, the model is used to determine how the measurement system will respond to a material loss.
3.1 MBA2 Model Details

This section describes how the model and material balance is setup in Matlab Simulink. This initial work includes a number of assumptions and simplifications, but future work will review the model and add detail where needed.

The material balance over MBA2 starts with the measured dissolver solution from the accountability tank. The accountability tank is fed in batches until the max level is reached (once every eight hours), at which point the model simulates tank sampling. Accountancy is achieved using a concentration measurement from the sample multiplied by a level measurement of the tank—these measurements are all simulated with errors in the model. The accountability tank block feeds material into the PUREX feed tank block in pulses.

The PUREX feed tank may be used for chemistry adjustment for the separations process, but it also serves as buffer to maintain a continuous flow of material into the pulsed columns. Sampling of the feed tank is currently not practiced routinely, so this is not included in the model. However, solution monitoring techniques can estimate the amount of actinides present based on the accountability tank measurements and process monitoring measurements (flow rates and level measurements). The model simulates an actinide estimate with larger uncertainty values.

The pulsed columns use liquid-liquid extraction to separate the U and Pu from one another. The model tracks both the U and Pu concentrations sent to the waste tank, the Pu decontamination area, the U decontamination area, and the solvent recycle mixer-settler. The PUREX Separation block therefore yields four outputs. On existing plants, measurements of the pulsed columns are not feasible due to the separations that are occurring, but models have been developed that can estimate the U and Pu content to within 10%. These estimates are simulated in the Simulink model.

A very small amount of residual fission products remain in the U and Pu streams after the initial U/Pu partitioning process is complete. Plants therefore utilize secondary extraction cycles for the purpose of further decontaminating the U and Pu streams. The U Decontamination and Pu Decontamination blocks within the model correspond to those processes. Again, only estimates of U and Pu content are possible for these processing steps.

Following all of the extraction cycles the solvent and acid are transferred to clean-up cycles so that they can be recycled for use in the main process. The recycled solvent and acid contain minute amounts of U and/or Pu, which must be accounted for by the operator and verified on a random basis by the inspector. Confirmatory measurements are simulated in the model.

The U and Pu Surge/Buffer Tanks provide surge/buffer capacity and regulate flow into the output accountability tanks in batches. U and Pu content can be estimated using solution monitoring techniques as described above. However, the solution monitoring techniques used in the purification cycles use down-stream measurements from the output accountability tanks. In the Simulink model, the actinide quantities are simulated as an estimate with higher uncertainties.
The U and Pu product tanks and the waste tank collect the nitrate products and waste for accountancy purposes. Routine sampling on each batch is used for low uncertainty measurements of the U and Pu product. The waste tank or final waste form may be measured to confirm low quantities of actinides. Additional denitrators or calciners are used to convert the liquid into a solid product, but these are outside of MBA 2. Future work will need to develop the back end of the plant in more detail. For now, the focus of the modeling analysis is only on MBA 2.

Figure 3 shows the inventories in each of the key processing units in MBA2 that are used for the material balance. The left graphs show the total liquid inventory in liters, and the right graphs show just the total Pu content in kg. These graphs give an idea of the transient nature of the plant operations.
Figure 3: MBA2 inventories during normal operation: total liquid volume (left), total Pu content (right); x-axis scale is in units of hours from startup.
3.2 Safeguards System Modeling

NMA in a PUREX plant is dominated by the uncertainties associated with the inventory change measurements, primarily the input and output measurements, which need to be kept as low as possible. Achieving low uncertainties during the periodic Interim Inventory Verification (IIV) for timeliness is more difficult due to the continuous operation of the process. Although there may be opportunities for better inventory measurements between customer campaigns when there is a flushout, the material balance is only closed after the yearly ‘clean-out’ prior to a Physical Inventory Taking (PIT) and the Physical Inventory Verification (PIV) at which time low uncertainty measurements can be done. Therefore, more frequent IIVs are carried out with random sampling and using estimates of the in-process inventory where sampling is not possible.

For the SSPM, it was assumed that plant balances were calculated every 8 hours (once per accountability tank batch) using the input and output measurements along with higher uncertainty estimates of the in-process inventory. The input and output accountability tanks are modeled using low uncertainty measurements that are achieved with best practices at existing plants using HKED or KED. See Table 1 for the assumed uncertainties at various points. The surge vessels are assumed to be estimated using solution monitoring techniques at slightly higher uncertainties than the accountability tanks. The U/Pu content in the pulsed columns is assumed to be estimated to ± 10%. The balance period can easily be changed in future work, and should not affect the results presented here significantly.

All of the measurements or estimates that are used for the material balance include both random and systematic errors, which are set by the user. The random error is truly random, and Simulink applies a random number generator to each measurement that will be different for each consecutive measurement. The error applied will follow a normal distribution with a mean of zero and standard deviation equal to the relative random error chosen by the user. This small error, either positive or negative, is added to the true value to give a simulated measurement. The systematic error is applied differently—a random number generator is used to apply an error only once at the beginning of the run, and then this error is held constant (for each individual measurement) for the remainder of the run. This simulates the bias generated by the systematic errors.

The material balance calculates the inventory difference (ID) once every 8 hours and also tracks the cumulative sum of the ID and error propagation. For simplicity, only Pu balances are currently modeled and shown in the results. A Page’s Test is used to set alarm conditions to detect material loss. The test has been chosen to keep the false alarm probability below 5% while still containing enough sensitivity to detect abrupt and protracted diversions.

Because of the random nature of the measurements, every run will lead to slightly different results. When the diversion is near the detection limits, some runs may signal an alarm using the Page’s Test, and some runs will not. In order to determine the detection probability, multiple runs are required. For each test case, 100 runs were completed to determine an average detection probability.
4.0 Modeling Results

The goal of the exercise using the model was to compare the traditional use of HKED for accountancy measurements under protracted diversion events with the use of hiRX. HKED is more than adequate for detecting abrupt diversions, but the advantage of hiRX is seen in the ability to detect longer protracted diversions. Table 1 shows the measurement uncertainties (including both random and systematic relative errors) that were assumed for the different cases. The first set of columns labeled as “Traditional” show the errors assuming the base case, or what is typically done in existing reprocessing plants. The uncertainties shown are for Pu measurements using a variety of measurement (both NDA and DA) as shown in reference 3. The second set of columns labeled as “hiRX” assume an improved measurement at input and output accountability using the hiRX technology. The third set of columns labeled as “Expanded hiRX” assume additional use of hiRX at the PUREX Feed Tank and Pu Surge Tank.

Only the locations highlighted in yellow changed for the different cases. The inventory measurements in the pulsed columns were all assumed to be 10% measurements, and the other product and waste tanks were assumed with measurement uncertainties from best practices shown in reference 3.

Table 1: Pu measurement uncertainties for the traditional, hiRX, and expanded hiRX runs

<table>
<thead>
<tr>
<th>Location</th>
<th>Traditional</th>
<th>hiRX</th>
<th>Expanded hiRX</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>RE</td>
<td>SE</td>
<td>RE</td>
</tr>
<tr>
<td>Accountability Tank</td>
<td>0.8%</td>
<td>0.5%</td>
<td>0.1%</td>
</tr>
<tr>
<td>PUREX Feed Tank</td>
<td>2%</td>
<td>2%</td>
<td>1%</td>
</tr>
<tr>
<td>PUREX Separation</td>
<td>10%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>U Decontamination</td>
<td>10%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>U Surge Tank</td>
<td>5%</td>
<td>2%</td>
<td>5%</td>
</tr>
<tr>
<td>U Product Tank</td>
<td>2%</td>
<td>2%</td>
<td>2%</td>
</tr>
<tr>
<td>Recycle Mixer-Settler</td>
<td>7%</td>
<td>7%</td>
<td>7%</td>
</tr>
<tr>
<td>Pu Decontamination</td>
<td>10%</td>
<td>10%</td>
<td>10%</td>
</tr>
<tr>
<td>Pu Surge Tank</td>
<td>2%</td>
<td>2%</td>
<td>1%</td>
</tr>
<tr>
<td>Pu Product Tank</td>
<td>0.3%</td>
<td>0.3%</td>
<td>0.1%</td>
</tr>
<tr>
<td>Waste Tank</td>
<td>7%</td>
<td>7%</td>
<td>7%</td>
</tr>
<tr>
<td>Waste Calciner</td>
<td>7%</td>
<td>7%</td>
<td>7%</td>
</tr>
</tbody>
</table>

For the “Traditional” base case, HKED was assumed to be used for the Pu measurement at the input accountability tank and at the Pu product tank. Solution monitoring techniques were assumed to estimate Pu content to 2% for both random and systematic errors in the PUREX feed tank (right after accountability) and the Pu surge tank (right before accountability). The low uncertainty accountability measurement right before or after these tanks should provide reasonable estimates of the content in those tanks. However, these numbers will need to be reviewed in future work.
For the “hiRX” case, the measurement uncertainties at input accountability and the Pu product tank were decreased to 0.1%. This value is believed to be achievable with the hiRX technology, but experimental testing will be required for confirmation. As the experimental progress on hiRX develops, this number can be changed as needed. Due to this improved measurement, it was also assumed that the solution monitoring techniques could estimate Pu content at the PUREX feed tank and Pu surge tank to 1%.

For the “Expanded hiRX” case, it was assumed that hiRX was used for additional sampling of the PUREX feed tank and Pu surge tank to give a Pu inventory measurement at 0.1%. Since both of these tanks also contain large quantities of Pu, they may be additional locations to consider sampling with hiRX, which would be enabled by the improved timeliness and lower costs of hiRX measurements.

For each of the three model setups, five protracted diversion cases were examined. In all cases, material was removed right before Pu decontamination. The fraction of material removed was modified and the diversion time was increased in order to remove a total of 8 kg of Pu for each case.

Table 2 shows a summary of the runs performed with the SSPM. A 95% detection probability is a typical goal in regulations, and achieving that goal would mean that an alarm was triggered in 95 out of the 100 runs. The first column of values corresponds to the base case using HKED, and 95% detection probably was not achieved until above a 3% diversion. The second column of values corresponds to using hiRX technology at the accountability tanks instead of HKED—as can be seen, a 1% diversion was detected with greater than 95% detection probability. If hiRX is implemented at more locations than just the accountability tanks (represented by the third column), the improvement is even more substantial. This improvement shows that the use of hiRX for the routine verification measurements provides at least a three-fold improvement in the ability to detect protracted diversions, and more substantial gains may be possible if hiRX can be used for additional sampling throughout the plant.

The false alarm probability was also examined for both the “HKED” case and the “hiRX” case in order to make sure that the Page’s Test was not leading to higher than normal detection probabilities. For the false alarm test, a 1600 hour run was performed without any diversion of material. For the “HKED” case 3/100 of the runs led to alarm, and for the “hiRX” case 4/100 of the runs led to alarm. Thus, the false alarm probability is below 5%.
Table 2: Probability of detection for five different levels of diversions

<table>
<thead>
<tr>
<th>Diversion Scenario</th>
<th>HKED</th>
<th>hiRX</th>
<th>Advanced hiRX</th>
</tr>
</thead>
<tbody>
<tr>
<td>3% Diversion (over 100 hrs)</td>
<td>92%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td>2% Diversion (over 150 hrs)</td>
<td>71%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td>1% Diversion (over 300 hrs)</td>
<td>39%</td>
<td>98%</td>
<td>100%</td>
</tr>
<tr>
<td>0.5% Diversion (over 600 hrs)</td>
<td>24%</td>
<td>65%</td>
<td>98%</td>
</tr>
<tr>
<td>0.25% Diversion (over 1200 hrs)</td>
<td>21%</td>
<td>33%</td>
<td>75%</td>
</tr>
</tbody>
</table>
5.0 Conclusion

The SSPM model has been modified to represent a PUREX reprocessing plant in order to test an advanced accountancy and verification measurement technology. The hiRX technology shows promise in the ability to provide a better measurement than HKED for routine measurements. The SSPM was used to determine the potential improvement to safeguards under various diversion cases. The modeling indicates that the HKED technology may not detect a diversion of less than 3% of a Pu product stream with greater than 95% detection probability. The hiRX technology may provide an increased ability to detect a 1% diversion with 98% detection probability, leading to an improvement in the ability to detect protracted diversions. More extensive sampling and analyses throughout the plant will certainly result in improved detection probability for all methods.

Future work will focus on the model assumptions (such as adequate representation of tank and column sizes), and a general review of the model will be performed to ensure that it is representative of existing plants. A notable change will be to reduce the material throughput to 1000 MT/yr, even though this is unlikely to change the results. Diversion from more specific locations will be examined. Also, the ability to achieve more of an on-line measurement capability will be examined to determine potential impact to safeguards.
6.0 References

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