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Advancing the State of the Art in Materials Accountancy through Safeguards Performance Modeling

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Advancing the State of the Art in Materials Accountancy through Safeguards Performance Modeling

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Abstract

Using a Safeguards Performance Model, an advanced instrumentation scenario has been developed for materials accountability in a UREX+1a reprocessing plant. This scenario takes advantage of new technology to both advance the state of the art in materials accountability and reduce plant construction and operational costs. The model was used to determine the overall standard error of the inventory difference, the instrumentation response to misuse scenarios or off-normal plant processes, and which areas of the plant may be better served with advanced instrumentation. Some candidate new instrumentation is suggested along with the expected improvements in overall accountability. The conclusions of this work are as follows: (1) As little as five additional plutonium in-process measurement points will provide enough information to determine the change of in-process inventory with time—true near real-time accountability of plutonium can drastically shorten the detection time of a protracted diversion by eliminating the need for a plant flush-out. (2) Additional measurement points will require either rapid, low-error mass spectrometry measurements or rapid on-line non-destructive measurements. Emerging technologies that may be best suited include TARIS (Thermal Atomization Resonance Ionization Spectroscopy) and Ultra-High Resolution Spectroscopy. (3) A rapid non-destructive measurement will also be useful for measuring actinides in spent fuel assemblies, a continuing gap in materials accountancy. (4) Uncertainty still exists as to how well the rest of the minor actinides and some of the key fission products need to be measured. Experimental work is needed to determine the measurement precision of these species in mixed solutions.

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Acronyms

| | |
|----------|---|
| AFCI | Advanced Fuel Cycle Initiative |
| CCD-PEG | Cesium/Strontium Extraction |
| CFR | Code of Federal Regulations |
| GNEP | Global Nuclear Energy Partnership |
| HKED | Hybrid K-Edge Densitometry |
| IAEA | International Atomic Energy Agency |
| IDMS | Isotope Dilution Mass Spectrometry |
| KMP | Key Measurement Point |
| L | Liter |
| MT | Metric Tons |
| NDA | Non-Destructive Analysis |
| NRC | Nuclear Regulatory Commission |
| PWR | Pressurized Water Reactor |
| RE | Rare Earth |
| SEID | Standard Error of the Inventory Difference |
| SNF | Spent Nuclear Fuel |
| TALSPEAK | Rare Earth Fission Product Extraction |
| TARIS | Thermal Atomization Resonance Ionization Spectroscopy |
| TITR | Titration |
| TRU | Transuranic Isotopes (Np, Pu, Am, Cm) |
| TRUEX | Transuranic Actinide Extraction |
| UDS | Undissolved Solids |
| UREX | Uranium Extraction |

Elements

| | |
|----|------------|
| Am | Americium |
| Cm | Curium |
| Cs | Cesium |
| Np | Neptunium |
| Pu | Plutonium |
| Sr | Strontium |
| Tc | Technetium |
| U | Uranium |

Advancing the State of the Art in Materials Accountancy through Safeguards Performance Modeling

1.0 Introduction

Nuclear fuel reprocessing presents the most challenging area within the nuclear fuel cycle to safeguard. A potential UREX+1a plant, which has been proposed as part of the Global Nuclear Energy Partnership (GNEP), will be the most complicated plant design to date due to an increased number of separation steps. The purpose of this work was to develop an instrumentation mapping for the plant along with the statistical analyses that demonstrate the ability of the plant to achieve materials accountancy goals.

Plenty of information exists regarding accountancy instrumentation for existing reprocessing plants throughout the world. What sets this work apart is the development of a Safeguards Performance Model that allows for virtual testing of the instrumentation mapping. The model was used as a tool for performing a gap analyses and determining instrumentation response to off-normal events or diversion pathways. The model allows for instrumentation design optimization to save costs, to develop advanced data correlation strategies, and to determine the effect of advanced instrumentation.

This tool will allow plant designers to incorporate Safeguards by Design in the next generation of reprocessing plants. It also can be used to advance the state-of-the-art in material safeguards by testing new data correlation strategies. Beyond the scope of this work, it is also possible to extend this model to incorporate plant safety and security as well.

2.0 Background

2.1 Materials Accountability Goals

Because of the difficulties of measuring elemental content in mixed reprocessing solutions, accountability goals as opposed to requirements have been set up. The most applicable Nuclear Regulatory Commission (NRC) regulation, 10CFR74 [1], can be used as a basis for domestic safeguards until new guidance is issued. 10CFR74.59(f) requires a 6-month shut-down and flush-out to calculate the material balance inventory difference. The standard error of the inventory difference (SEID) must be estimated for any material mass balance—and an SEID > 0.1% of the active inventory must be examined along with any ID that is greater than three times the SEID and 200 g of Pu. 10CFR74.53 requires a statistical test for abrupt diversion that has at least a 95% chance of detecting the loss of 2 kg of Pu within three working days for Category 1A material and seven working days for Category 1B material.

The International Atomic Energy Agency (IAEA) has requirements which are similar and focus on the detection of a diversion of one significant quantity of plutonium (8 kg) with 95% detection probability. International safeguards agreements are different than domestic safeguards in that they must protect against diversion by the host country. Verification and data authentication play a large role.

Since nuclear material is held up in the plant piping and processing vessels, a flush-out is required at the end of a campaign to complete the mass balance. Discrepancies in the mass balance will occur at startup and flushout due to the material hold up. It is possible that a protracted diversion would not be detected until the next flush-out occurs.

For large reprocessing plants (~2000 MT/yr), if the goal is to detect the loss of one significant quantity of plutonium in one month, the combined error on the plutonium measurement must be smaller than 0.1% to reach the accountability goal. This is an incredibly difficult level of precision to achieve in a large plant. Current reprocessing plants around the world do not achieve this goal which is why additional measures (physical protection, containment, and surveillance) are used.

2.2 Advancing the State-of-the-Art

The purpose of this work is to help advance the state-of-the-art in the design of a materials accountability system for a reprocessing plant. Current plants already have their accountability practices established and approved, and the argument could be made that existing systems are secure enough. However, new technologies are continuously being developed which can improve overall measurement uncertainties and/or decrease the costs of meeting accountability requirements. This modeling work provides a tool for determining the effect of these potential improvements.

Current accountability systems suffer from two weaknesses:

1. Precision measurements of elemental quantities in mixed isotope reprocessing samples take a great deal of time, thus limiting how many areas of the plant can be feasibly sampled.
2. Precision measurements are taken from the plant inputs and the plant outputs, but the in-process inventory change is not accounted for to a high level of precision.

What this means is that current plants have accurate measurements of the amount of material entering and leaving, but there are no elemental measurements to show how the material builds up in the processing vessels. Various predictive models can be used along with bulk process monitoring measurements (flow rates, tank volumes, etc.), but a plant flushout is required to close out the mass balance.

A true near real-time accountability system would be able to track the change of the plant inventory (for elemental quantities) along with the input and output measurements. Such a system, if it achieved a high enough precision, could possibly eliminate the need for a plant flushout and interim inventory measurements. In effect, a mass balance could be completed on the order of every 8 hours. This paper will show how this goal could be achieved—specifically what the properties of new instrumentation must be to move toward this type of an advanced accountability system.

3.0 Safeguards Performance Model

The Safeguards Performance Model is a tool for examining advanced accountancy systems and new technology in a reprocessing plant. Simulink (a simulation software package that runs under MATLAB) has been used to develop a transient flow model of a UREX+1a reprocessing plant. This model tracks cold chemicals, bulk fluid flow, solids, and the individual elemental quantities of U, Pu, Np, Am, Cm, Cs, and Sr. Expected separation efficiencies are specified to determine the quantity of nuclear material going into different streams. Measurement models are used to simulate an expected measurement from a particular piece of instrumentation. The following sections describe the mass balance, measurement models, and statistical analyses in more detail.

3.1 Mass Balance

The Simulink model is broken down into five different sub-models based on the UREX+1a process: Front end, uranium extraction (UREX), cesium/strontium extraction (CCD-PEG), transuranic actinide extraction (TRUEX), and rare earth fission product extraction (TALSPEAK). Dividing the plant up in this manner will be useful for potential changes to the separation steps in the future. An entire plant simulation runs each model in sequence. The model currently does not include product conversion at the end of each separation step, but this can be added later if appropriate. The final product from each separation step (in dissolved fluid form) is used as the plant output.

A complete mass balance was used as the basis of the model, so at any instant during a simulation, the total mass entering a processing vessel equals the total mass leaving it unless the vessel inventory is changing. More detailed information about the mass balance is given in reference 2. The data used to build the model and assumptions comes from the AMUSE code developed at Argonne National Laboratory. Every stream in the model contains information about the volume and mass flow rate, the concentrations of the 7 key elements tracked, and the solids flow rate where appropriate (at the front end). Separation efficiencies are specified in the dissolver and the contactors to determine the percentage of each element going into a specific output stream. Section 3.3 includes pertinent data about the flow rates and separation efficiencies.

Tanks are modeled assuming that the fluid phase is well mixed, a good assumption for accountability tanks but not for all areas of the plant. In other words, it is assumed that fluid samples taken from anywhere within the tank at the same time instant would have the same composition. The modeling of the contactor trains is less intuitive, but simplified since details about the individual contactors may not be known (from the materials accountability standpoint). Each contactor train is broken up into 3 different model blocks to represent the extraction, strip, and scrub operations. The specified fluid inventory within each such block is based on the number of stages within the contactor. Each block has two inputs and two outputs (organic and nitric acid), and the elemental content in the outputs is defined by specified separation efficiencies.

3.2 Measurement Models

A universal measurement block has been designed for use in any location within the model. This block can be used to simulate any type of desired measurement having to do with mass, volume, flow rate, or concentration. The block simulates a real measurement, which is imperfect, and then logs the measurement history as a variable in MATLAB. Once the measurements are collected, statistical tests can then be used to analyze the data.

Each measurement block contains parameters that are supplied by the user. The user specifies which stream variable is to be measured (such as the bulk fluid flow or an individual element's concentration), sampling period, random error, systematic error, drift, and calibration period. The data from this measurement is recorded into a matrix with one value for each time period.

3.3 UREX+1a Model

The following five figures show the complete UREX+1a model in Simulink. Plant equipment is labeled below the block, and each type of equipment shows the number of in-flows and out-flows. The main dissolver solution output from each sub-model becomes the input for the next sub-model. Measurement blocks are labeled by the measurement type below the block.

The measurement blocks at first glance will appear to be excessive. The measurements shown in Figures 1-5 are for an advanced plant that may be able to take advantage of technology advances to include more in-process measurements. For a more conservative approach that only uses measurements that are available on plants today, much of this added measurement data can be treated as if it did not exist. Sections 4 and 5 will go into more detail about the differences between a baseline and advanced instrumentation scenario.

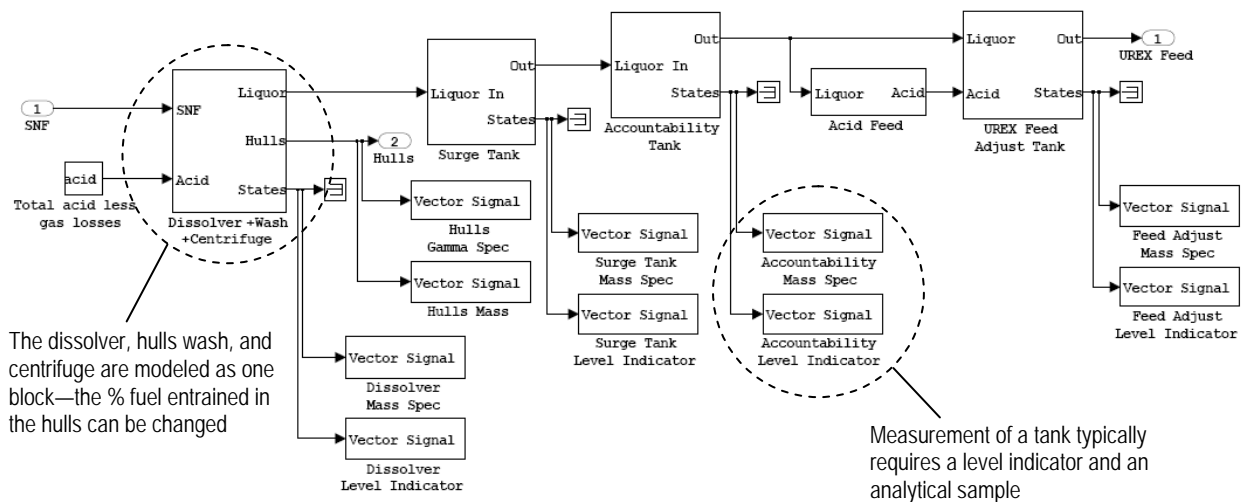


Figure 1: Front End Model

Figure 1 shows the Front End Model which begins with dissolution of the spent fuel. The dissolver tank, hulls wash tank, and centrifuge are all modeled as one block for simplicity. The dissolved fuel moves on to a surge tank, accountability tank, and UREX feed adjust tank. The accountability tank is where the first precision measurement of the solution occurs. The output from the Front End moves on to the UREX extraction.

The UREX extraction is shown in Figure 2. This extraction removes U/Tc from the dissolved fuel. The incoming solution from the UREX feed adjust tank goes directly into a train of centrifugal contactors. It would be possible to model each contactor separately using Simulink, but for the purposes of this work it was adequate to combine contactors into three blocks representing extraction, scrub, and strip. Each block has an aqueous solution input and organic solvent input along with an extractant and raffinate. The separation efficiency of each element tracked can be specified. The separation efficiencies of the three blocks were set so that the overall separation efficiency was equal to the results of the Argonne AMUSE code.

The end of the UREX extraction includes the UREX holding tank that contains the separated U and Tc product. The organic solvent is recycled in a mixer-settler tank before being reused in the extraction. Note that measurement blocks are included on the two tanks at the end of the process as well as the first block of the extraction. These will be discussed in later sections.

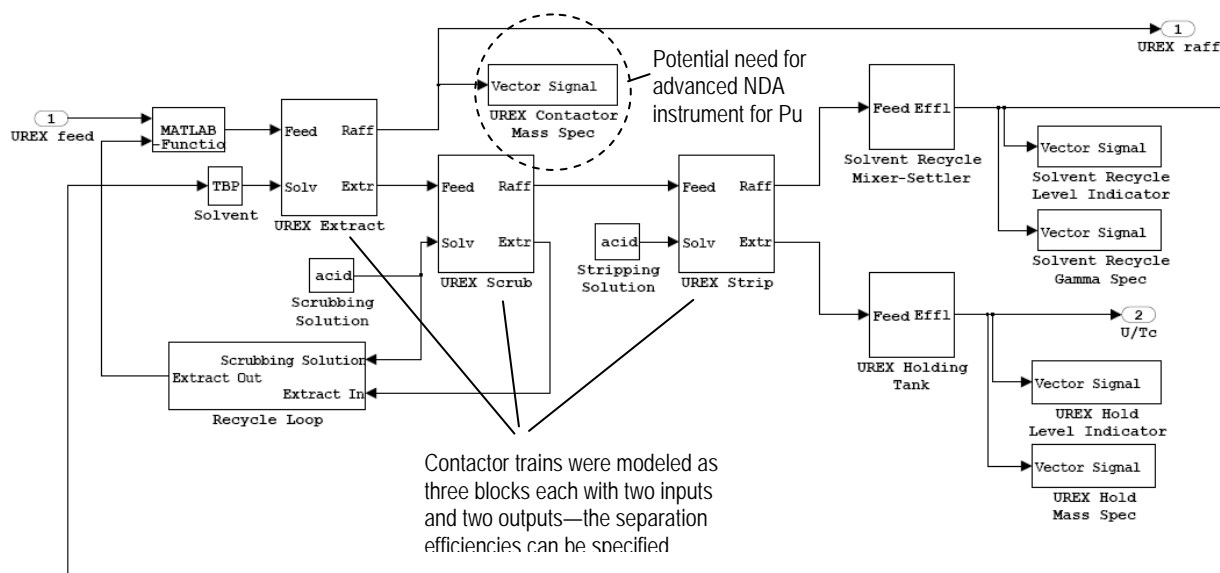


Figure 2: UREX Extraction

The leftover raffinate from the UREX extraction moves on to the CCD-PEG extraction, which is shown in Figure 3. This extraction removes the Cs/Sr from the dissolved fuel. Again, the contactors are modeled as three blocks, and the final product solution is contained in the Cs/Sr holding tank. Measurement blocks are shown for both the holding tank and the mixer-settler tank.

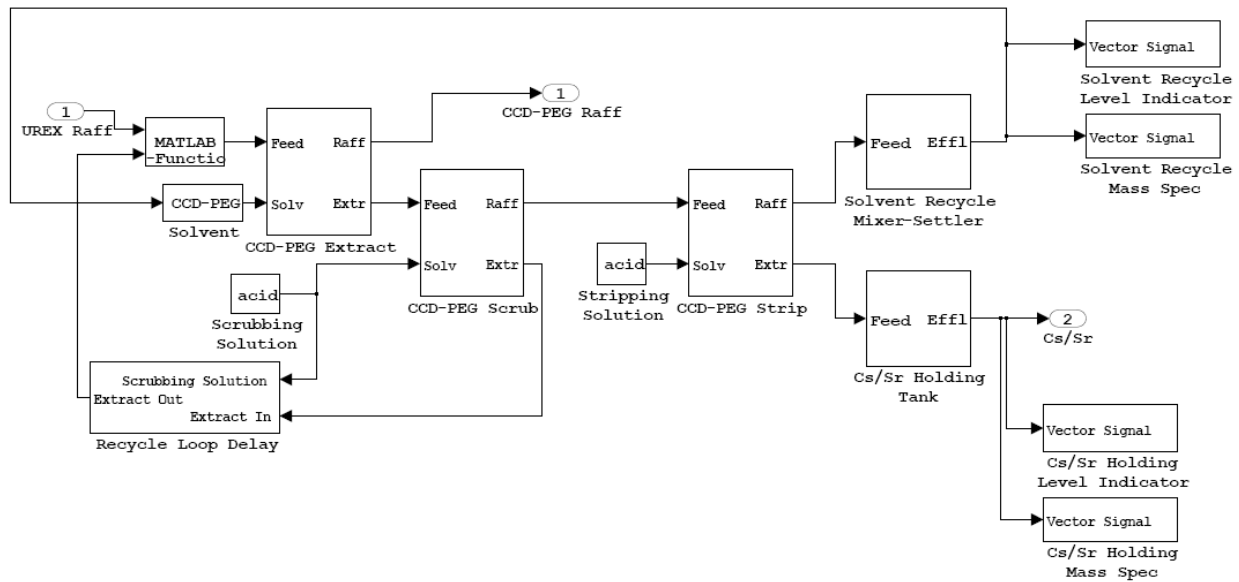


Figure 3: CCD-PEG Extraction

The leftover raffinate from the CCD-PEG extraction contains all transuranic elements and all fission products except Cs/Sr. This raffinate moves on to the TRUEX extraction shown in Figure 4. The TRUEX extraction removes the transuranic elements and rare earth fission products from the rest of the fission products. A stripper and reduction tank are required before the TRUEX contactors to adjust the chemistry for this extraction.

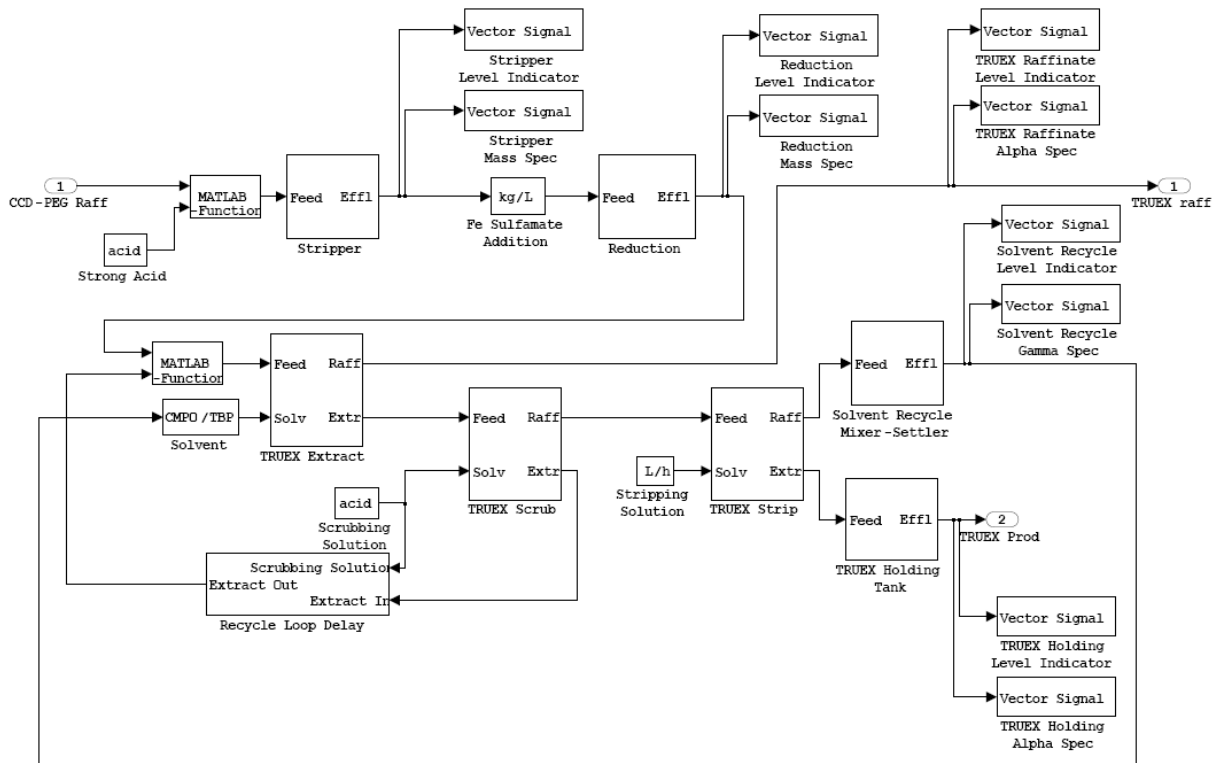


Figure 4: TRUEX Extraction

The rest of the flow chart looks similar to the previous two extractions. The TRUEX raffinate contains the left-over fission products and will be fabricated into a high level waste form. The TRUEX product goes to the TRUEX holding tank and contains the transuranics and rare earth fission products.

This solution then moves on to the final extraction, the TALSPEAK extraction, shown in Figure 5. A feed adjust tank prepares the solution for extraction, which removes the rare earth fission products from the transuranics. The rare earth fission products are collected in the RE product tank—this solution will be combined with the other fission products in fabrication of the high level waste form. The raffinate from TALSPEAK contains the final transuranic product that can be fabricated into a fuel.

A diversion block was added to the transuranic product stream for use in testing a diversion scenario. This block can be set up to divert a fraction of the entire stream or individual elements (though diversion of the entire stream is more realistic). The starting time of the diversion can be set along with the length of diversion. In this manner it can be used to test the instrumentation response to either abrupt or protracted diversion of material.

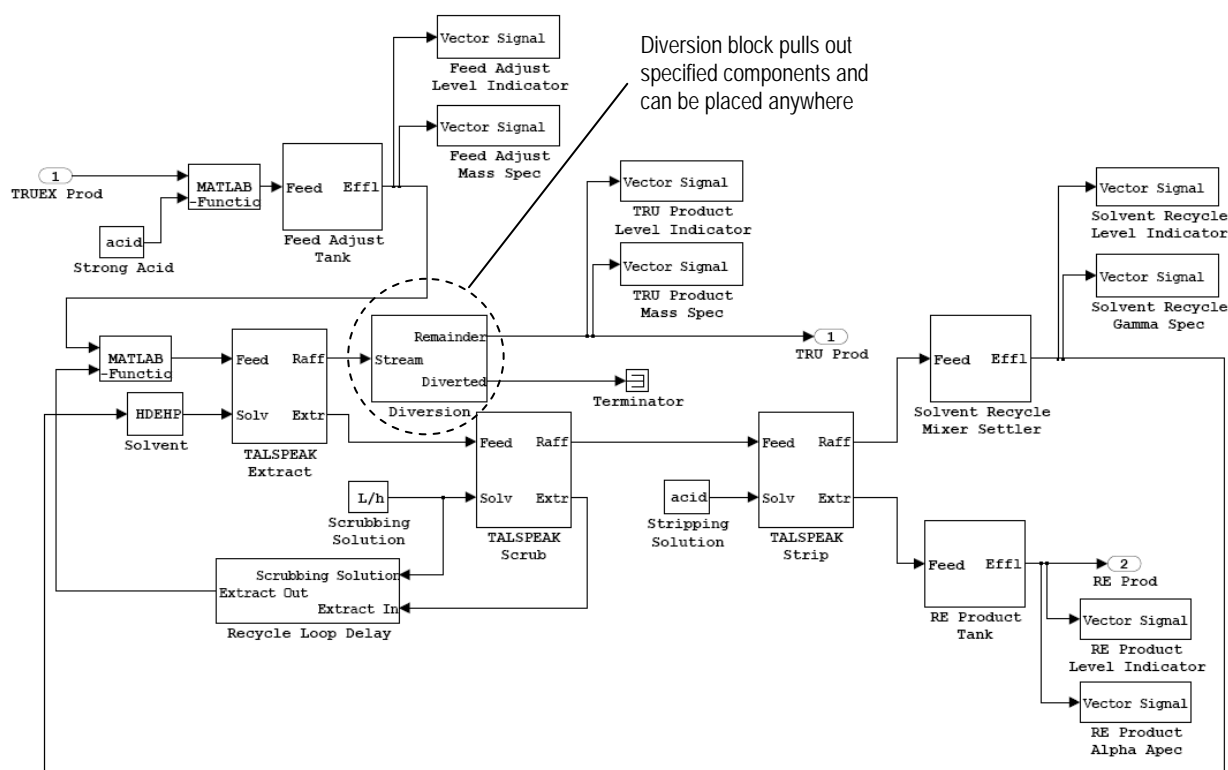


Figure 5: TALSPEAK Extraction

3.4 Time Dependence

The Safeguards Performance Model is a transient model that includes time dependence. The initial conditions of all components can be specified, and the time of the run can be set. For example, to model startup the initial elemental quantities in all components will be set to zero. After startup, nuclear material will begin to build up in the processing vessels until a steady-state condition is achieved. Likewise, to model flushout the feed is set to zero until the nuclear material within the plant gets completely flushed out of the system.

The feed can also be changed during a run to represent a switch to a different burnup or even different type of fuel. Then the transient concentrations throughout the plant can be tracked. For all measurement blocks throughout the plant, the measurement will log both the actual value and the simulated measurement value.

For this model, the dissolver was assumed to be a batch process, while the contactors are continuous. The surge tank, accountability tank, and UREX adjust tank at the front end were used to maintain a continuous flow to the UREX extraction. The Simulink model is able to adequately mimic how this would occur in a real plant by allowing the tank volumes to fill and empty.

The time dependence makes it possible to simulate the turn around time for measurements throughout the process. The measurements are logged as a function of time in Matlab arrays, and then these measurements can be analyzed later for SEID or instrumentation response to normal operation and misuse scenarios. The following chapter discusses the instrumentation in more detail.

4.0 Baseline Instrumentation List

The model was configured with two instrumentation lists, a baseline and an advanced mapping. These two lists were used to compare the effect of adding additional instrumentation to the overall safeguards performance of the system. The baseline instrumentation list includes materials accountancy instrumentation that would likely be installed on a reprocessing plant built today. This list is based on established instrumentation in use around the world and is made up of instrumentation that is currently available off-the-shelf.

4.1 Previous Work

The *International Target Values 2000* reference [3] forms the basis for much of the baseline instrumentation in the model. This reference provides typically achievable uncertainties for mass, volume, and density measurements as well as a detailed list of the uncertainties of U and Pu concentration measurements using various laboratory techniques. The difficulty with the current practice is that all existing reprocessing plants only separate U and Pu, so they do not need to deal with challenge of measuring the other minor actinides or Cs/Sr. The accountability of a UREX+1a plant is further complicated since the requirements for measuring the minor actinides and Cs/Sr are not set yet.

The instrument locations for a UREX+1a plant are based on a number of past references, mostly funded through GNEP or AFCI projects of the past [4,5,6,7,8,9]. In some cases the instrumentation choices are estimates given the uncertainty regarding future regulations.

4.2 Instrumentation Mapping

Figure 6 shows the instrumentation mapping for the baseline design. This figure shows the UREX+1a flow chart with the key instrumentation points shown in red. Traditional materials accountability includes a relatively small number of measurement points. In this case, the input measurement is taken at the accountability tank, while the outputs are taken from the five product/waste forms indicated. One additional measurement is taken of the washed hulls. Traditional accounting requires the mass balance to be completed at plant flushout since the in-process inventory is not measured.

For measurement points 1-6, the total plutonium measurement is found using a level indicator on a tank for volume plus a concentration measurement in a sample. For all cases the level indicator was assumed to give a volume measurement with 0.1% random and systematic errors. Note that these errors are only possible if great care is taken in calibration and measurement procedures [3].

Table 1 shows more detail about the instrumentation parameters for the concentration measurements including the expected instrument, random and systematic errors, and the measurement turn around time. The table is broken down into each element to make it easier to see how many measurements go into a mass balance for a particular element.

The measurement technique for the minor actinides is somewhat uncertain as the accounting requirements for these species have not been set. Alpha spectroscopy has been proposed as part of the Engineering Alternatives Study [7], but it would require a significant amount of time in the radiochemistry lab to separate the components and perform measurements. Hybrid K-edge densitometry may be more suited for these measurements. Either way, the measurement of the minor actinides is a gap that needs to be closed with further research and development. The measurement uncertainties listed for alpha spectroscopy are only estimates based on one reference [10], so more work is needed.

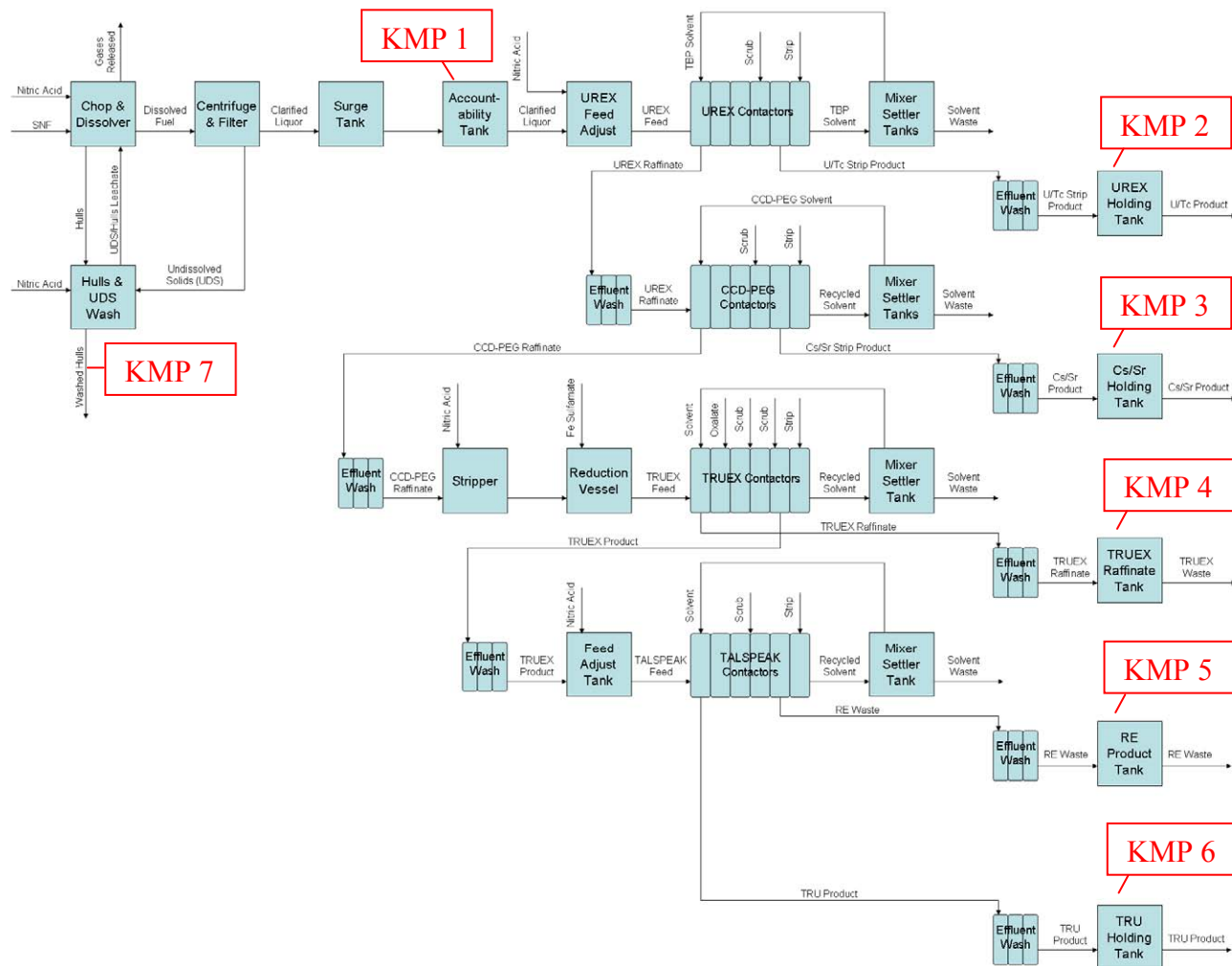


Figure 6: Baseline Instrumentation Mapping

The standard measurement technique for U and Pu at the key accountability points is Isotope Dilution Mass Spectrometry (IDMS)—this measurement requires lengthy chemical separation before the measurement. Titration (TITR) is used for separated U products. Laboratory alpha spectroscopy (α spec) may be used for minor actinides, but it also requires lengthy chemical separation. Gamma spectroscopy (γ spec) using high purity germanium detectors can be used for some of the product or waste forms to varying degrees of uncertainty. Liquid scintillation may

be used for measuring the Tc and Sr product. Finally, in many areas of the plant, gross alpha and gamma measurements (gross α,γ) may be used for rough measurement of material.

| Measurement Point | Uranium | Plutonium | Neptunium | Americium | Curium | Cesium | Strontium | Technetium |
|-----------------------|----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|--------------------|
| 1 | IDMS | IDMS | α spec | α spec | α spec | | | |
| Accountability | $\sigma_{ran}=0.2\%$ | $\sigma_{ran}=0.2\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | | | |
| Tank | $\sigma_{sys}=0.2\%$ | $\sigma_{sys}=0.2\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | | | |
| | 8 hours | 8 hours | 8 hours | 8 hours | 8 hours | | | |
| 2 | TITR | gross α,γ | gross α,γ | gross α,γ | gross α,γ | gross α,γ | gross α,γ | Liq Scint |
| UREX | $\sigma_{ran}=0.1\%$ | | | | | | | $\sigma_{ran}=5\%$ |
| Holding | $\sigma_{sys}=0.1\%$ | | | | | | | $\sigma_{sys}=2\%$ |
| Tank | 8 hours | | | | | | | 8 hours |
| 3 | | gross α | gross α | gross α | gross α | γ spec | Liq Scint | |
| Cs/Sr | | | | | | $\sigma_{ran}=5\%$ | $\sigma_{ran}=5\%$ | |
| Holding | | | | | | $\sigma_{sys}=2\%$ | $\sigma_{sys}=2\%$ | |
| Tank | | | | | | x hours | x hours | |
| 4 | IDMS | α spec | α spec | α spec | α spec | | | |
| TRUEX | $\sigma_{ran}=0.2\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | | | |
| Raffinate | $\sigma_{sys}=0.2\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | | | |
| Tank | 8 hours | 8 hours | 8 hours | 8 hours | 8 hours | | | |
| 5 | | gross α | gross α | gross α | gross α | | | |
| RE | | | | | | | | |
| Product | | | | | | | | |
| Tank | | | | | | | | |
| 6 | | IDMS | α spec | α spec | α spec | | | |
| TRU | | $\sigma_{ran}=0.2\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | $\sigma_{ran}=3\%$ | | | |
| Holding | | $\sigma_{sys}=0.2\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | $\sigma_{sys}=1\%$ | | | |
| Tank | | 8 hours | 8 hours | 8 hours | 8 hours | | | |
| 7 | | γ spec | | γ spec | | γ spec | | |
| Hulls | | $\sigma_{ran}=5\%$ | | $\sigma_{ran}=5\%$ | | $\sigma_{ran}=5\%$ | | |
| | | $\sigma_{sys}=2\%$ | | $\sigma_{sys}=2\%$ | | $\sigma_{sys}=2\%$ | | |
| | | x hours | | x hours | | x hours | | |

Table 1: Baseline Instrumentation Parameters

4.3 Standard Error of the Inventory Difference

Using the expected measurement errors as shown in Table 1, it is possible to determine the SEID for each elemental mass balance. Only the actinides have enough information to complete a mass balance—the fission product measurements are taken for the purpose of waste characterization.

It is useful to note that this SEID only covers the input and output measurements in the plant—in-process inventory is not included. Therefore this SEID is based on the assumption that all in-process inventory is liberated from the plant during flushout. It also means that this error can only be calculated at plant flushout, so a potential diversion may take a while to detect.

The SEID is calculated as a mass rate in kg/hr. Table 2 shows example measurement values for the actinides along with the measurement uncertainty. The bottom row shows the calculation of the inventory difference between the plant outputs and the plant input. The uncertainty on this inventory difference is the SEID for the five actinides.

| Measurement Point | Uranium | Plutonium | Neptunium | Americium | Curium |
|---------------------------|------------------------|-------------------------|---------------------------|---------------------------|---------------------------|
| 1 Accountability Tank | 121.9 ± 0.512 kg/hr | 1.932 ± 0.008 kg/hr | 0.151 ± 0.005 kg/hr | 0.106 ± 0.0035 kg/hr | 0.029 ± 0.0009 kg/hr |
| 2 UREX Holding Tank | 120.5 ± 0.337 kg/hr | | | | |
| 3 Cs/Sr Holding Tank | | | | | |
| 4 TRUEX Raffinate Tank | 1.23 ± 0.0052 kg/hr | 0.019 ± 0.0006 kg/hr | 0.0015 ± 0.00005 kg/hr | 0.0011 ± 0.00004 kg/hr | 0.0003 ± 0.00001 kg/hr |
| 5 RE Product Tank | | | | | |
| 6 TRU Holding Tank | | 1.916 ± 0.008 kg/hr | 0.149 ± 0.005 kg/hr | 0.104 ± 0.0034 kg/hr | 0.029 ± 0.0009 kg/hr |
| Inventory Difference | -0.17 ± 0.85 kg/hr | 0.003 ± 0.017 kg/hr | -0.0005 ± 0.010 kg/hr | -0.0009 ± 0.0069 kg/hr | -0.0003 ± 0.0018 kg/hr |

Table 2: Standard Error for the Baseline Mass Balance

For example, the SEID for plutonium was 0.017 kg/hr. In order for a protracted diversion to be detected with a 95% detection probability, the diversion would need to be greater than 3.3 x SEID, or 0.0561 kg/hr. If a protracted diversion of material is kept below this mass flow rate, then it is unlikely to be detected. Significant quantities of the other minor actinides are not defined, but the quantities of Np, Am, and Cm even in a large plant are small in comparison to the amount of Pu.

4.4 Response to Off-Normal Events

The difficulty with measuring only the inputs and outputs is that there are large differences at the beginning and end of the run. To demonstrate this discrepancy, the Pu mass balance was used (column 2 in Table 2). At startup, Pu is going to accumulate in the process vessels, so the Pu inputs will be greater than the sum of the Pu outputs. At the end of the run, during flush-out, Pu will still be coming out of the plant even though no fuel is being fed.

The Safeguards Performance Model was used to demonstrate this effect using a one month run assuming standard pressurized water reactor (PWR) fuel. Flush-out was started at hour 620.

Figure 7 shows the result. A large negative inventory difference is present at the beginning of the run, and a large positive inventory difference is present during flush-out. In between, the process is at steady-state so the inventory difference varies slightly around zero. After flushout, a balance of the net inventory difference will be very close to zero during normal operation.

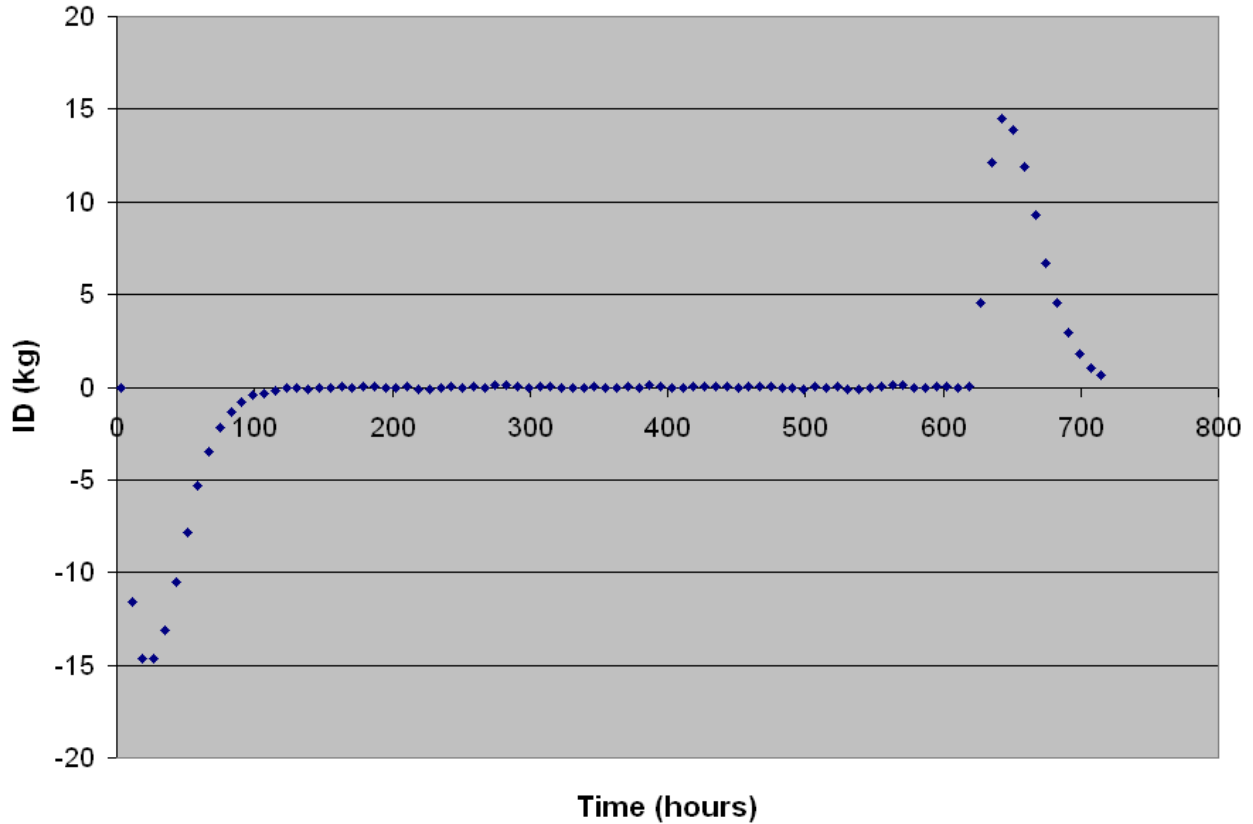


Figure 7: Inventory Difference using Traditional Accounting

The performance measure of the baseline instrumentation is how well it responds to an off-normal event. Such events could include leaks, problems with the separations steps, or misuse scenarios. The diversion block described in section 3.3 was used to simulate a misuse scenario. Figure 8 shows the effect of a diversion of 8 kg of Pu spread out over 100 hours. The model assumed the diversion of 4.2% of the TRU product stream starting at hour 50 and ending at hour 150. Although the diversion is evident on this plot, it is not difficult to see how a diversion could be hidden in the plant startup or flushout phase or during any other plant transient. Such a diversion would not be detected until the full mass balance was performed.

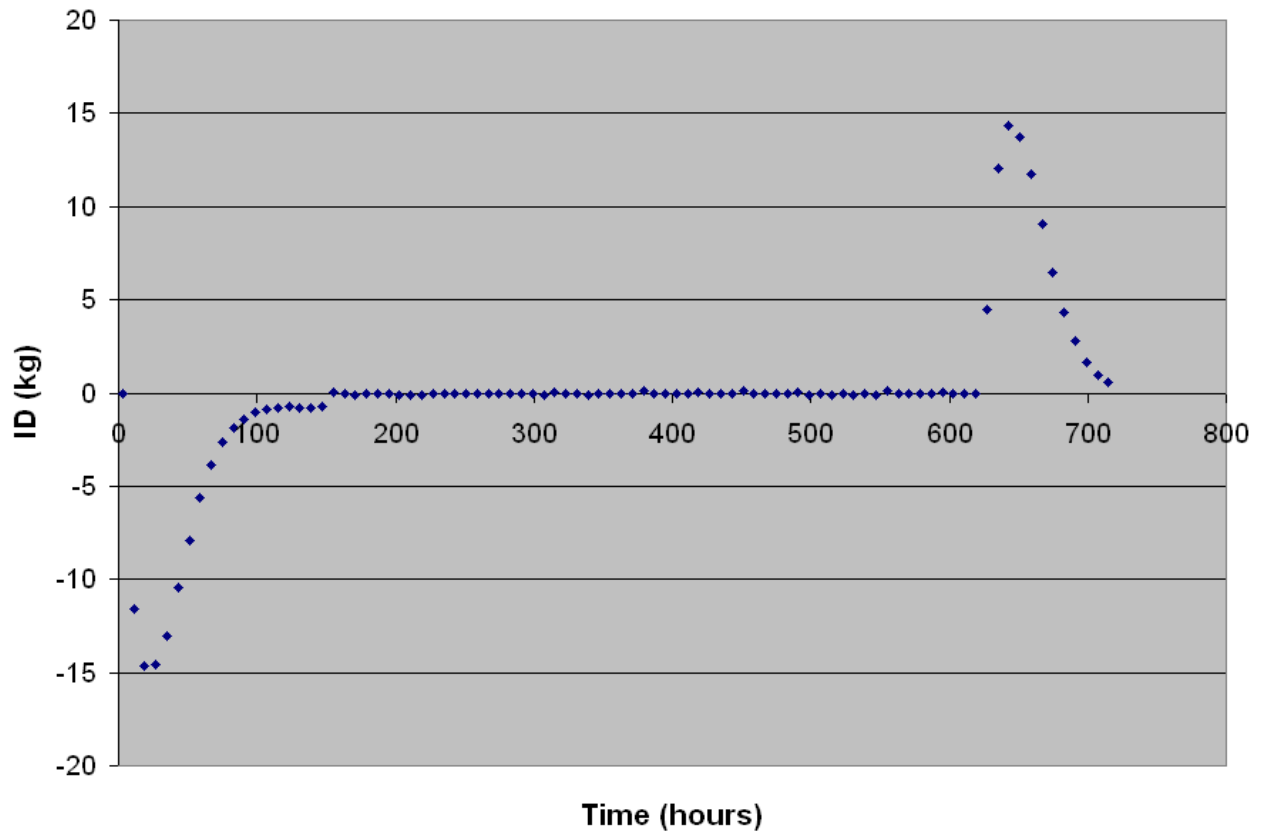


Figure 8: Inventory Difference with a Diversion Event using Traditional Accounting

The goal of this work was to determine instrumentation gaps and improvements which can more quickly and easily detect these types of misuse scenarios. The following section describes advanced instrumentation which may drastically improve the state-of-the-art of materials accountability.

5.0 Advanced Instrumentation List

Three goals have been identified in developing advanced instrumentation for future reprocessing plants:

1. Identify gaps in traditional accounting and determine if new instrumentation can help fill those gaps.
2. Identify ways to reduce plant costs or use new instrumentation to influence the overall cost of the plant design.
3. Determine the measurement characteristics required to help guide instrumentation development.

Sections 5.1 and 5.2 describe the lessons learned from this analysis. Sections 5.3, 5.4, and 5.5 describe an advanced instrumentation list and the model runs that were performed. Finally, section 5.6 summarizes these results into an advanced instrumentation requirement list that others in the safeguards community can use as a guide in developing new measurement techniques.

5.1 Input and Output Measurements

The current uncertainty of the measurements on the accountability tank and the outputs from mass spectrometry are about as good as they are going to get. It is unlikely that new instrumentation will push the uncertainties much below 0.1% simply because of the realities of doing laboratory measurements. Advances in these locations should focus instead on decreasing the measurement time. Given that a typical mixed isotope reprocessing sample may have measure turn-around time of about 8 hours, accountancy measurements require a large amount of time and money to keep the laboratories staffed.

A large gap in accounting is the measurement of the plutonium in the spent fuel coming into the plant. No technique exists for measuring elemental content in spent fuel rods with low uncertainty until the fuel is dissolved at the accountability tank. For this reason, current plants use the accountability tank as the input and rely on physical protection and other barriers to protect spent fuel at the front end of the plant.

5.2 In-Process Inventory

The in-process inventory is another gap in accountability. While current plants do keep track of bulk material flow (total volumes or masses of solution), elemental content from the plant processing vessels is not measured. Therefore, there is no data on the change of Pu inventory within the plant as a function of time—leading to the need for a plant flush-out to complete the mass balance. If it is possible to determine the complete plant inventory change based on in-process measurements, it might be possible to completely eliminate the need for a periodic plant flushout.

Measuring every internal tank and contactor could be expensive. Therefore, as a first step, it is useful to determine whether some areas in the plant would not require measurement for various statistical reasons. Many locations in the plant may only have small volumes being processed or

may be expected to only contain trace quantities of nuclear material. These areas can potentially be ignored.

Figure 9 shows the plant schematic with the expected tank or contactor volumes (in Liters) shown in blue. All of the effluent wash stages and the CCD-PEG, TRUEX, and TALSPEAK contactors contain very small quantities of material in comparison to the tanks. Therefore, the relative amount of fissionable material in these processing steps can be neglected from a mass balance, which in turn eliminates the need for measurement instrumentation at these locations.

All of the mixer-settler tanks, which are used for the recycled solvent, are expected to only contain trace quantities of nuclear material. Gross gamma and alpha measurements of these tanks will likely be required for process monitoring to ensure nuclear material is not present. In this sense, these areas act as confirmatory measurements to make sure the amount of nuclear material is negligible. By taking advantage of this process control information, four more in-process measurements have been eliminated.

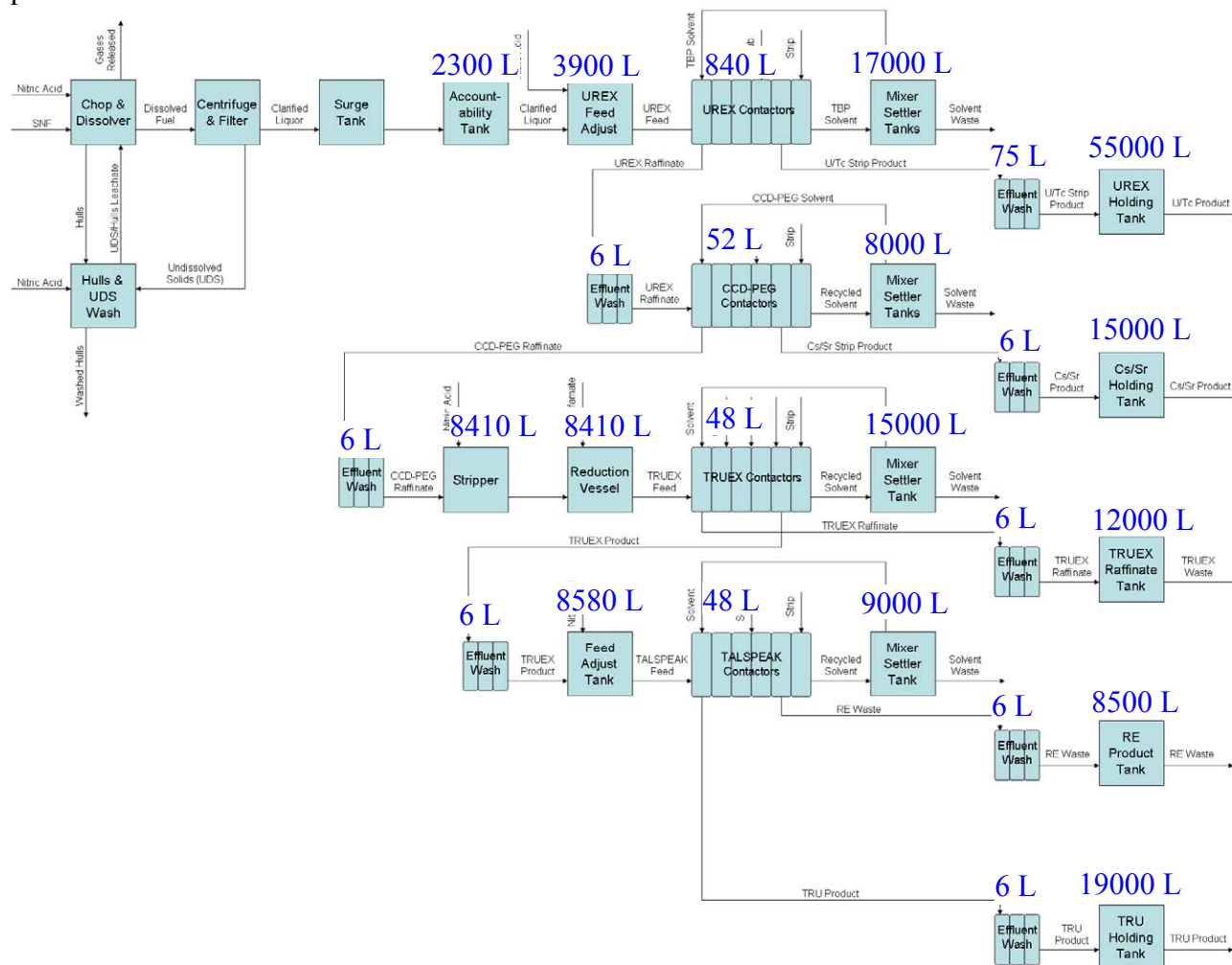


Figure 9: In-Process Volumes

That leaves four additional tanks and the UREX Contactor train that must be measured with accuracy to determine the in-process inventory. New measurement techniques that can drastically speed up a mass spectrometry measurement may open the door to taking additional samples from the four additional tanks. A non-destructive measurement technique will be required to measure the amount of material in the UREX contactor train since sampling is nearly impossible due to the nature of contactor operation. Fortunately, the total volume of the UREX contactors is small compared to the rest of the tanks, so the measurement technique does not require low uncertainty (<5% will be adequate).

Finally, it is worth noting that the front end of the plant (before the accountability tank) was excluded from this exercise. The accountability tank is the first time that precise data on the plant inputs is received. If new measurement techniques are able to precisely measure actinides in spent fuel (non-destructively with uncertainty < 0.5%), then the in-process measurements of the front end will become important. Future work will investigate the front end in more detail.

5.3 Advanced Instrumentation Mapping

Figure 10 shows the advanced instrumentation mapping with new measurement areas indicated in green and blue. These measurement points would be required to determine the change in in-process Pu inventory with time (to prevent the need for plant flushout).

Key measurement points 8, 9, 10, and 11 are the mixer-settler tanks, which are expected to only contain trace quantities of nuclear material. Gross alpha and gamma measurements with a relatively high uncertainty are adequate at these locations since the amount of actinides is so low compared to other areas of the plant. The gross alpha and gamma measurements combined with the level indicator will provide confirmation that only very small quantities of actinides should exist in these locations. This instrumentation will be required for process monitoring, so it does not represent an increase in instrumentation costs.

Key measurement points 12, 13, 14, 15, and 16 represent new capability. These locations all contain statistically significant quantities of plutonium so must be either sampled or measured non-destructively to determine the plant inventory change. The UREX feed adjust tank, TRUEX stripper, TRUEX reduction vessel, and TALSPEAK feed adjust tank all contain significant volumes and significant quantities of plutonium. These tanks should be sampled and measured with low uncertainty in the advanced accountability scenario. The UREX contactors will contain plutonium only in the first set of stages for which the total volume of material would be about 280 L. The quantity of plutonium at this location is small compared to the rest of the plant, so a plutonium measurement with uncertainty around 5% would be adequate. A non-destructive technique could be used in this location.

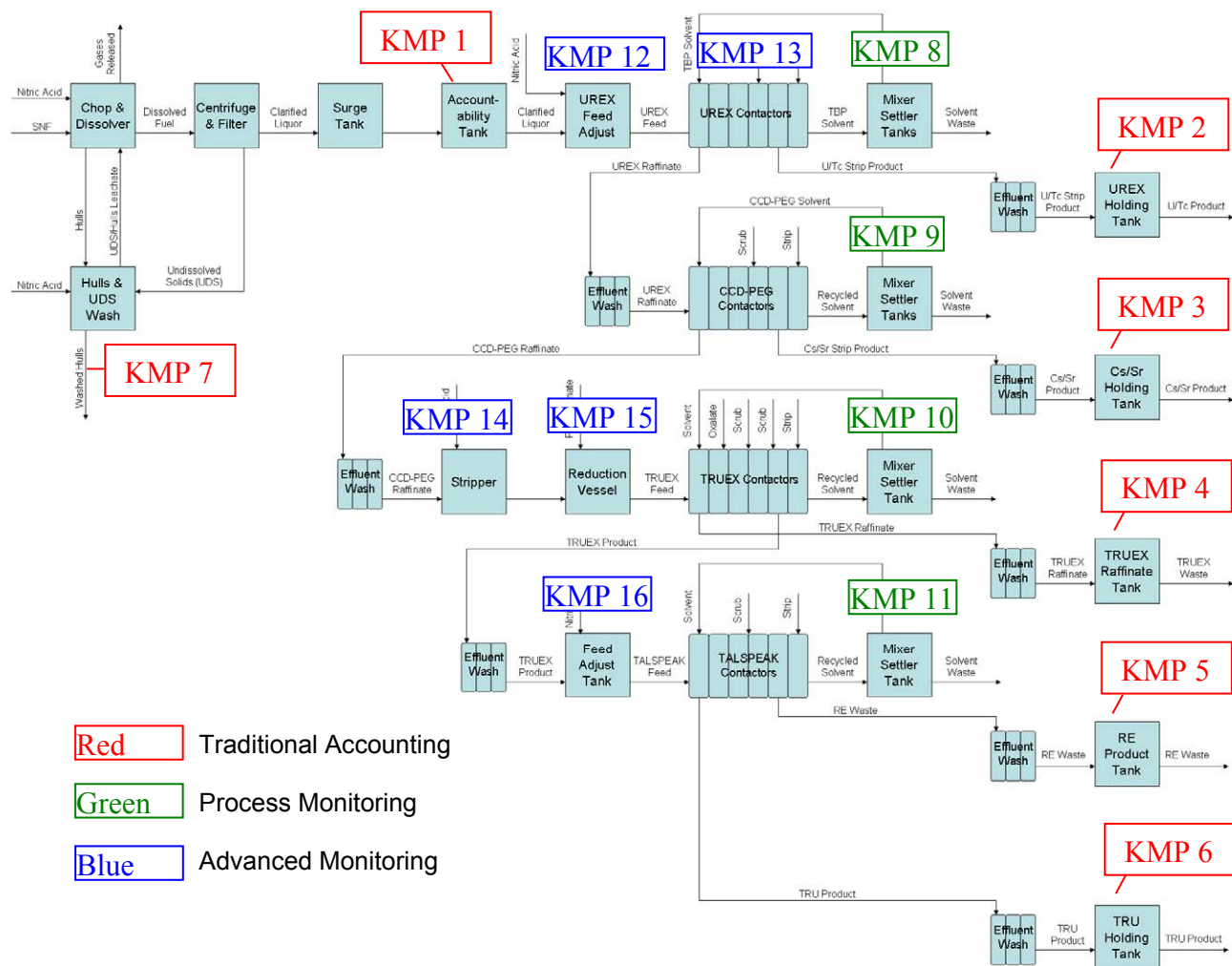


Figure 10: Advanced Instrumentation Mapping

Table 3 shows the desired plutonium concentration measurement parameters for the additional measurement points. These parameters point to the need for either rapid on-line analysis of plutonium (at low error) or more rapid laboratory analyses that allow for more samples to be taken. The UREX contactors require some type of NDA measurement that can determine the plutonium content from an unusual geometry, but fortunately the error does not need to be very low.

| Measurement Point | Plutonium |
|-------------------|-----------------------------|
| 12 | ??? |
| UREX Feed | $\sigma_{\text{ran}}=0.2\%$ |
| Adjust Tank | $\sigma_{\text{sys}}=0.2\%$ |
| | 8 hours |
| 13 | ??? |
| UREX | $\sigma_{\text{ran}}=5\%$ |
| Contactors | $\sigma_{\text{sys}}=2\%$ |
| | 8 hours |
| 14 | ??? |
| TRUEX | $\sigma_{\text{ran}}=0.2\%$ |
| Stripper | $\sigma_{\text{sys}}=0.2\%$ |
| | 8 hours |
| 15 | ??? |
| TRUEX | $\sigma_{\text{ran}}=0.2\%$ |
| Reduction | $\sigma_{\text{sys}}=0.2\%$ |
| | 8 hours |
| 16 | ??? |
| TALSPEAK | $\sigma_{\text{ran}}=0.2\%$ |
| Feed Adjust | $\sigma_{\text{sys}}=0.2\%$ |
| Tank | 8 hours |
| | ??? |
| SNF | $\sigma_{\text{ran}}=0.2\%$ |
| | $\sigma_{\text{sys}}=0.2\%$ |
| | per batch |

Table 3: Advanced Measurement Parameter Requirements

5.4 Standard Error of the Inventory Difference

Under the advanced instrumentation scenario, the standard error can be calculated at any time as opposed to just at plant flushout. Not only does this make it much quicker to detect off-normal events, but it may prevent the need for plant flushout to save operating costs. The model assumptions for the five advanced measurement locations are shown in Table 3, although the equipment was not specified since this is a requirements list.

An assumption was made that only Pu would need to be measured in near-real time using this advanced instrumentation scenario. In other words, all of the additional instrumentation would only focus on Pu measurements as opposed to all of the actinides. This would likely be sufficient since Pu is the critical material for safeguards, since the rest of the minor actinides are present in much lower quantities, and since the uranium coming from a reprocessing plant is not an ideal material for weapon-making. The overall system, then, will account for Pu in near-real

time taking into account input, output, and in-process measurements, but all other species will only take into account input and output measurements.

Table 4 shows the results of a 360 hour run using the model along with the advanced instrumentation. Each column represents a different processing point and the plutonium measurement at that point. All measurements are given in kg of Pu at that particular point in time. The final four columns show the plant input, sum of the plant outputs, the in-process inventory change, and finally a calculation of the inventory difference (all in kg/hr).

At plant startup as the in-process inventory is changing, the inventory difference is on the order of 0.1 to 0.2 kg/hr. Once steady-state is reached, the inventory difference is around 0.01 to 0.02 kg/hr. Of more importance is the uncertainty in the measurement, around 0.5 kg/hr. Although this uncertainty is somewhat high, a complete mass balance is being completed every 8 hours without a plant flushout. In 8 hours, there is not enough time to hide a diversion of 8 kg of Pu within the measurement uncertainty.

The inventory difference from this run is shown in Figure 11. As expected, the inventory difference hovers very close to zero since the discrepancy between input and output measurements is offset by the additional measures of the change of in-process inventory.

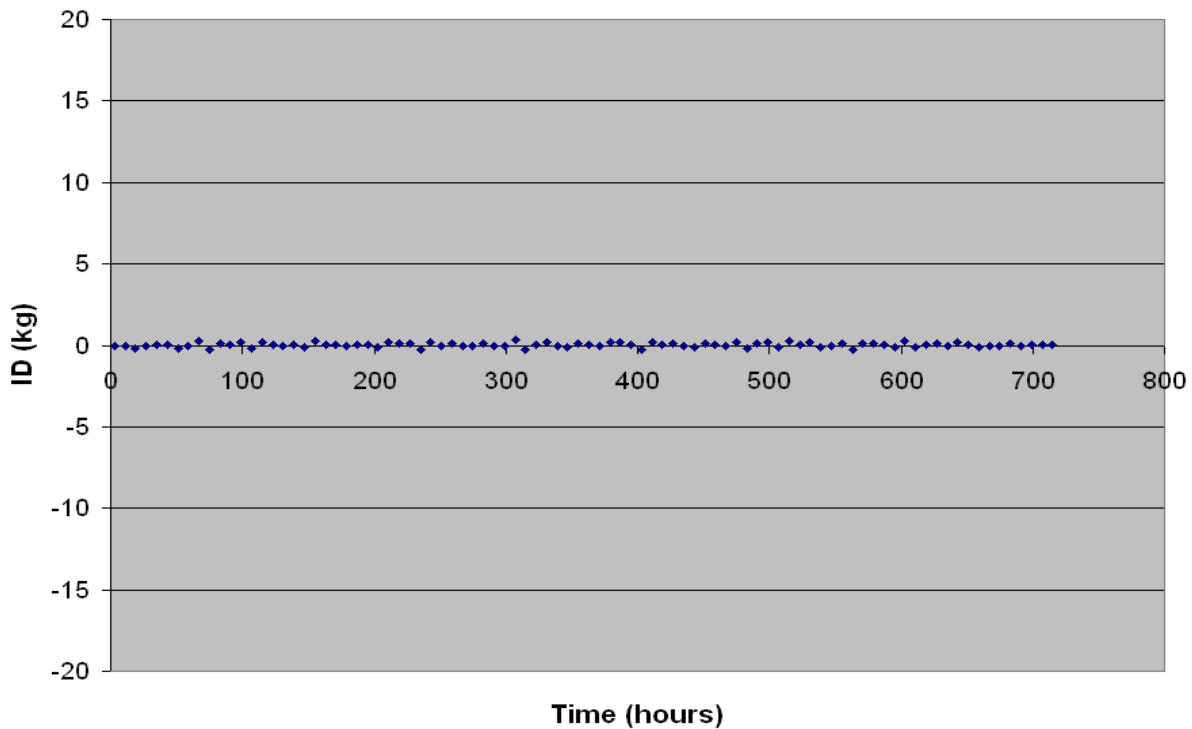


Figure 11: Inventory Difference using Advanced Accounting

| Hour | Accountability Tank | UREX Adjust Tank | UREX Contactors | UREX Mixer Settler Tank | UREX Holding Tank | CCD-PEG Mixer Settler | Cs/Sr Holding Tank |
|------|---------------------|------------------|-----------------|-------------------------|-------------------|-----------------------|--------------------|
| 8 | 0.00 ± 0.000 | 0.00 ± 0.000 | 0.00 ± 0.000 | 3.6E-29 ± 1.9E-30 | 1.2E-28 ± 3.8E-30 | 3.0E-30 ± 1.6E-31 | 4.5E-25 ± 1.5E-26 |
| 16 | 11.63 ± 0.049 | 9.83 ± 0.042 | 0.48 ± 0.026 | 1.1E-08 ± 6.2E-10 | 3.7E-08 ± 1.2E-09 | 7.0E-10 ± 3.8E-11 | 1.2E-04 ± 4.1E-06 |
| 24 | 14.80 ± 0.063 | 15.05 ± 0.064 | 0.77 ± 0.042 | 4.2E-08 ± 2.3E-09 | 1.4E-07 ± 4.5E-09 | 2.8E-09 ± 1.5E-10 | 4.2E-04 ± 1.4E-05 |
| 32 | 15.28 ± 0.065 | 16.18 ± 0.069 | 0.78 ± 0.042 | 6.9E-08 ± 3.7E-09 | 2.2E-07 ± 7.4E-09 | 4.4E-09 ± 2.4E-10 | 7.3E-04 ± 2.4E-05 |
| 40 | 15.48 ± 0.066 | 16.37 ± 0.069 | 0.75 ± 0.040 | 8.6E-08 ± 4.6E-09 | 2.8E-07 ± 9.1E-09 | 6.0E-09 ± 3.2E-10 | 8.9E-04 ± 2.9E-05 |
| 48 | 15.44 ± 0.065 | 16.43 ± 0.070 | 0.82 ± 0.044 | 1.1E-07 ± 5.8E-09 | 3.5E-07 ± 1.1E-08 | 7.0E-09 ± 3.8E-10 | 9.5E-04 ± 3.1E-05 |
| 56 | 15.46 ± 0.066 | 16.45 ± 0.070 | 0.80 ± 0.043 | 1.0E-07 ± 5.6E-09 | 3.4E-07 ± 1.1E-08 | 6.8E-09 ± 3.7E-10 | 1.1E-03 ± 3.6E-05 |
| 64 | 15.50 ± 0.066 | 16.48 ± 0.070 | 0.67 ± 0.036 | 1.1E-07 ± 5.8E-09 | 3.5E-07 ± 1.2E-08 | 7.5E-09 ± 4.0E-10 | 1.1E-03 ± 3.5E-05 |
| 72 | 15.55 ± 0.066 | 16.46 ± 0.070 | 0.76 ± 0.041 | 1.1E-07 ± 6.1E-09 | 3.7E-07 ± 1.2E-08 | 6.8E-09 ± 3.7E-10 | 1.2E-03 ± 3.9E-05 |
| 80 | 15.47 ± 0.066 | 16.46 ± 0.070 | 0.73 ± 0.039 | 1.1E-07 ± 5.7E-09 | 3.4E-07 ± 1.1E-08 | 7.3E-09 ± 3.9E-10 | 1.3E-03 ± 4.3E-05 |
| 88 | 15.51 ± 0.066 | 16.51 ± 0.070 | 0.76 ± 0.041 | 1.1E-07 ± 6.1E-09 | 3.7E-07 ± 1.2E-08 | 7.1E-09 ± 3.8E-10 | 1.1E-03 ± 3.6E-05 |
| 96 | 15.45 ± 0.066 | 16.49 ± 0.070 | 0.78 ± 0.042 | 1.1E-07 ± 5.9E-09 | 3.5E-07 ± 1.2E-08 | 7.2E-09 ± 3.9E-10 | 1.1E-03 ± 3.6E-05 |
| 104 | 15.48 ± 0.066 | 16.51 ± 0.070 | 0.71 ± 0.039 | 1.1E-07 ± 5.9E-09 | 3.5E-07 ± 1.2E-08 | 7.2E-09 ± 3.9E-10 | 1.1E-03 ± 3.6E-05 |
| 112 | 15.50 ± 0.066 | 16.47 ± 0.070 | 0.73 ± 0.040 | 1.1E-07 ± 5.9E-09 | 3.6E-07 ± 1.2E-08 | 7.4E-09 ± 4.0E-10 | 1.1E-03 ± 3.8E-05 |
| 120 | 15.55 ± 0.066 | 16.46 ± 0.070 | 0.74 ± 0.040 | 1.2E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 7.0E-09 ± 3.8E-10 | 1.2E-03 ± 4.0E-05 |
| 128 | 15.50 ± 0.066 | 16.46 ± 0.070 | 0.81 ± 0.044 | 1.2E-07 ± 6.3E-09 | 3.8E-07 ± 1.2E-08 | 7.7E-09 ± 4.2E-10 | 1.2E-03 ± 3.8E-05 |
| 136 | 15.51 ± 0.066 | 16.48 ± 0.070 | 0.80 ± 0.043 | 1.1E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 7.8E-09 ± 4.2E-10 | 1.2E-03 ± 4.1E-05 |
| 144 | 15.51 ± 0.066 | 16.49 ± 0.070 | 0.87 ± 0.047 | 1.1E-07 ± 6.1E-09 | 3.6E-07 ± 1.2E-08 | 7.9E-09 ± 4.3E-10 | 1.1E-03 ± 3.7E-05 |
| 152 | 15.52 ± 0.066 | 16.52 ± 0.070 | 0.79 ± 0.043 | 1.1E-07 ± 5.8E-09 | 3.5E-07 ± 1.2E-08 | 6.8E-09 ± 3.6E-10 | 1.2E-03 ± 3.8E-05 |
| 160 | 15.48 ± 0.066 | 16.48 ± 0.070 | 0.79 ± 0.043 | 1.2E-07 ± 6.3E-09 | 3.8E-07 ± 1.3E-08 | 7.0E-09 ± 3.8E-10 | 1.2E-03 ± 4.0E-05 |
| 168 | 15.46 ± 0.066 | 16.42 ± 0.070 | 0.80 ± 0.043 | 1.1E-07 ± 5.9E-09 | 3.5E-07 ± 1.2E-08 | 6.8E-09 ± 3.6E-10 | 1.2E-03 ± 3.8E-05 |
| 176 | 15.47 ± 0.066 | 16.54 ± 0.070 | 0.75 ± 0.040 | 1.1E-07 ± 6.0E-09 | 3.6E-07 ± 1.2E-08 | 7.2E-09 ± 3.9E-10 | 1.2E-03 ± 4.0E-05 |
| 184 | 15.52 ± 0.066 | 16.44 ± 0.070 | 0.76 ± 0.041 | 1.2E-07 ± 6.6E-09 | 3.9E-07 ± 1.3E-08 | 7.7E-09 ± 4.2E-10 | 1.1E-03 ± 3.8E-05 |
| 192 | 15.51 ± 0.066 | 16.50 ± 0.070 | 0.73 ± 0.039 | 1.1E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 7.2E-09 ± 3.9E-10 | 1.1E-03 ± 3.6E-05 |
| 200 | 15.49 ± 0.066 | 16.55 ± 0.070 | 0.78 ± 0.042 | 1.1E-07 ± 6.1E-09 | 3.6E-07 ± 1.2E-08 | 7.5E-09 ± 4.1E-10 | 1.2E-03 ± 3.8E-05 |
| 208 | 15.54 ± 0.066 | 16.50 ± 0.070 | 0.71 ± 0.039 | 1.1E-07 ± 5.7E-09 | 3.4E-07 ± 1.1E-08 | 7.6E-09 ± 4.1E-10 | 1.2E-03 ± 3.9E-05 |
| 216 | 15.48 ± 0.066 | 16.51 ± 0.070 | 0.72 ± 0.039 | 1.2E-07 ± 6.3E-09 | 3.8E-07 ± 1.2E-08 | 8.1E-09 ± 4.4E-10 | 1.1E-03 ± 3.7E-05 |
| 224 | 15.50 ± 0.066 | 16.50 ± 0.070 | 0.77 ± 0.042 | 1.1E-07 ± 5.8E-09 | 3.5E-07 ± 1.2E-08 | 7.8E-09 ± 4.2E-10 | 1.2E-03 ± 3.8E-05 |
| 232 | 15.47 ± 0.066 | 16.50 ± 0.070 | 0.77 ± 0.041 | 1.1E-07 ± 5.9E-09 | 3.5E-07 ± 1.2E-08 | 7.0E-09 ± 3.8E-10 | 1.0E-03 ± 3.4E-05 |
| 240 | 15.52 ± 0.066 | 16.46 ± 0.070 | 0.85 ± 0.046 | 1.1E-07 ± 6.1E-09 | 3.7E-07 ± 1.2E-08 | 7.5E-09 ± 4.0E-10 | 1.2E-03 ± 3.9E-05 |
| 248 | 15.46 ± 0.066 | 16.46 ± 0.070 | 0.80 ± 0.043 | 1.2E-07 ± 6.7E-09 | 4.0E-07 ± 1.3E-08 | 7.1E-09 ± 3.8E-10 | 1.2E-03 ± 3.9E-05 |
| 256 | 15.50 ± 0.066 | 16.50 ± 0.070 | 0.76 ± 0.041 | 1.1E-07 ± 6.0E-09 | 3.6E-07 ± 1.2E-08 | 7.1E-09 ± 3.9E-10 | 1.1E-03 ± 3.7E-05 |
| 264 | 15.53 ± 0.066 | 16.52 ± 0.070 | 0.74 ± 0.040 | 1.2E-07 ± 6.4E-09 | 3.8E-07 ± 1.3E-08 | 7.1E-09 ± 3.8E-10 | 1.2E-03 ± 4.1E-05 |
| 272 | 15.51 ± 0.066 | 16.51 ± 0.070 | 0.77 ± 0.042 | 1.1E-07 ± 6.1E-09 | 3.7E-07 ± 1.2E-08 | 7.9E-09 ± 4.2E-10 | 1.1E-03 ± 3.6E-05 |
| 280 | 15.49 ± 0.066 | 16.48 ± 0.070 | 0.75 ± 0.040 | 1.0E-07 ± 5.6E-09 | 3.3E-07 ± 1.1E-08 | 7.9E-09 ± 4.3E-10 | 1.1E-03 ± 3.6E-05 |
| 288 | 15.53 ± 0.066 | 16.53 ± 0.070 | 0.80 ± 0.043 | 1.1E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 6.9E-09 ± 3.7E-10 | 1.0E-03 ± 3.5E-05 |
| 296 | 15.60 ± 0.066 | 16.48 ± 0.070 | 0.74 ± 0.040 | 1.2E-07 ± 6.3E-09 | 3.8E-07 ± 1.2E-08 | 7.0E-09 ± 3.8E-10 | 1.2E-03 ± 3.8E-05 |
| 304 | 15.53 ± 0.066 | 16.54 ± 0.070 | 0.75 ± 0.041 | 1.2E-07 ± 6.3E-09 | 3.8E-07 ± 1.2E-08 | 6.8E-09 ± 3.7E-10 | 1.2E-03 ± 3.8E-05 |
| 312 | 15.48 ± 0.066 | 16.45 ± 0.070 | 0.76 ± 0.041 | 1.1E-07 ± 5.8E-09 | 3.5E-07 ± 1.1E-08 | 6.8E-09 ± 3.7E-10 | 1.2E-03 ± 3.9E-05 |
| 320 | 15.48 ± 0.066 | 16.50 ± 0.070 | 0.81 ± 0.043 | 1.1E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 7.1E-09 ± 3.9E-10 | 1.2E-03 ± 3.9E-05 |
| 328 | 15.53 ± 0.066 | 16.52 ± 0.070 | 0.82 ± 0.044 | 1.1E-07 ± 5.9E-09 | 3.5E-07 ± 1.2E-08 | 7.6E-09 ± 4.1E-10 | 1.1E-03 ± 3.8E-05 |
| 336 | 15.46 ± 0.066 | 16.50 ± 0.070 | 0.79 ± 0.042 | 1.1E-07 ± 6.1E-09 | 3.6E-07 ± 1.2E-08 | 7.9E-09 ± 4.3E-10 | 1.1E-03 ± 3.6E-05 |
| 344 | 15.54 ± 0.066 | 16.50 ± 0.070 | 0.75 ± 0.041 | 1.1E-07 ± 6.1E-09 | 3.7E-07 ± 1.2E-08 | 7.1E-09 ± 3.8E-10 | 1.1E-03 ± 3.7E-05 |
| 352 | 15.52 ± 0.066 | 16.52 ± 0.070 | 0.76 ± 0.041 | 1.1E-07 ± 6.2E-09 | 3.7E-07 ± 1.2E-08 | 7.1E-09 ± 3.8E-10 | 1.1E-03 ± 3.7E-05 |
| 360 | 15.43 ± 0.065 | 16.53 ± 0.070 | 0.80 ± 0.043 | 1.0E-07 ± 5.7E-09 | 3.4E-07 ± 1.1E-08 | 7.2E-09 ± 3.9E-10 | 1.2E-03 ± 4.0E-05 |

Table 4: Advanced Instrumentation Run (Values in kg/hr)

| TRUEX Stripper | TRUEX Reduction | | TRUEX Mixer Settler | | TRUEX Raffinate Tank | | TALSPEAK Feed Adjust | | TALSPEAK Mixer Settler | |
|----------------|-----------------|-------------------|---------------------|---------------|----------------------|-------------------|----------------------|-------------------|------------------------|--|
| 0.00 ± 0.000 | 0.00 ± 0.000 | 0.00 ± 0.000 | 1.0E-25 ± 5.6E-27 | 0.00 ± 0.000 | 0.00 ± 0.000 | 0.00 ± 0.000 | 0.00 ± 0.000 | 7.5E-30 ± 4.1E-31 | | |
| 2.33 ± 0.010 | 0.35 ± 0.001 | 4.0E-07 ± 2.2E-08 | 0.01 ± 0.000 | 0.00 ± 0.000 | 0.00 ± 0.000 | 5.6E-13 ± 3.0E-14 | | | | |
| 7.46 ± 0.032 | 3.48 ± 0.015 | 1.4E-05 ± 7.5E-07 | 0.05 ± 0.002 | 0.30 ± 0.001 | 2.0E-10 ± 1.1E-11 | | | | | |
| 11.83 ± 0.050 | 7.75 ± 0.033 | 4.9E-05 ± 2.7E-06 | 0.12 ± 0.004 | 1.58 ± 0.007 | 1.6E-09 ± 8.8E-11 | | | | | |
| 13.94 ± 0.059 | 11.14 ± 0.047 | 9.6E-05 ± 5.2E-06 | 0.17 ± 0.006 | 3.89 ± 0.016 | 5.8E-09 ± 3.1E-10 | | | | | |
| 14.90 ± 0.063 | 13.27 ± 0.056 | 1.3E-04 ± 7.0E-06 | 0.21 ± 0.007 | 6.68 ± 0.028 | 1.3E-08 ± 7.2E-10 | | | | | |
| 15.31 ± 0.065 | 14.44 ± 0.061 | 1.7E-04 ± 8.9E-06 | 0.23 ± 0.008 | 9.28 ± 0.039 | 2.0E-08 ± 1.1E-09 | | | | | |
| 15.39 ± 0.065 | 15.00 ± 0.064 | 1.9E-04 ± 1.0E-05 | 0.24 ± 0.008 | 11.34 ± 0.048 | 2.6E-08 ± 1.4E-09 | | | | | |
| 15.46 ± 0.066 | 15.18 ± 0.064 | 2.0E-04 ± 1.1E-05 | 0.24 ± 0.008 | 12.84 ± 0.054 | 3.9E-08 ± 2.1E-09 | | | | | |
| 15.38 ± 0.065 | 15.33 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.25 ± 0.008 | 13.77 ± 0.058 | 3.9E-08 ± 2.1E-09 | | | | | |
| 15.52 ± 0.066 | 15.42 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 14.37 ± 0.061 | 4.2E-08 ± 2.3E-09 | | | | | |
| 15.54 ± 0.066 | 15.49 ± 0.066 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 14.76 ± 0.063 | 4.8E-08 ± 2.6E-09 | | | | | |
| 15.50 ± 0.066 | 15.49 ± 0.066 | 2.3E-04 ± 1.2E-05 | 0.25 ± 0.008 | 14.95 ± 0.063 | 5.0E-08 ± 2.7E-09 | | | | | |
| 15.51 ± 0.066 | 15.44 ± 0.065 | 2.3E-04 ± 1.3E-05 | 0.24 ± 0.008 | 15.10 ± 0.064 | 5.1E-08 ± 2.8E-09 | | | | | |
| 15.57 ± 0.066 | 15.43 ± 0.065 | 2.3E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.21 ± 0.064 | 5.3E-08 ± 2.9E-09 | | | | | |
| 15.52 ± 0.066 | 15.45 ± 0.066 | 2.3E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.27 ± 0.065 | 5.7E-08 ± 3.1E-09 | | | | | |
| 15.60 ± 0.066 | 15.43 ± 0.065 | 2.3E-04 ± 1.2E-05 | 0.25 ± 0.008 | 15.24 ± 0.065 | 5.5E-08 ± 3.0E-09 | | | | | |
| 15.51 ± 0.066 | 15.39 ± 0.065 | 2.4E-04 ± 1.3E-05 | 0.24 ± 0.008 | 15.23 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.60 ± 0.066 | 15.47 ± 0.066 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.31 ± 0.065 | 5.5E-08 ± 3.0E-09 | | | | | |
| 15.56 ± 0.066 | 15.46 ± 0.066 | 2.4E-04 ± 1.3E-05 | 0.23 ± 0.008 | 15.30 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.56 ± 0.066 | 15.43 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.25 ± 0.008 | 15.30 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.49 ± 0.066 | 15.45 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.29 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.48 ± 0.066 | 15.50 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.27 ± 0.065 | 5.8E-08 ± 3.2E-09 | | | | | |
| 15.52 ± 0.066 | 15.44 ± 0.065 | 2.2E-04 ± 1.2E-05 | 0.25 ± 0.008 | 15.29 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.49 ± 0.066 | 15.45 ± 0.066 | 2.1E-04 ± 1.1E-05 | 0.23 ± 0.008 | 15.28 ± 0.065 | 5.3E-08 ± 2.8E-09 | | | | | |
| 15.52 ± 0.066 | 15.45 ± 0.065 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.23 ± 0.065 | 5.6E-08 ± 3.0E-09 | | | | | |
| 15.53 ± 0.066 | 15.47 ± 0.066 | 2.3E-04 ± 1.2E-05 | 0.23 ± 0.008 | 15.27 ± 0.065 | 5.0E-08 ± 2.7E-09 | | | | | |
| 15.59 ± 0.066 | 15.48 ± 0.066 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.23 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.53 ± 0.066 | 15.44 ± 0.065 | 2.1E-04 ± 1.2E-05 | 0.25 ± 0.008 | 15.25 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.50 ± 0.066 | 15.43 ± 0.065 | 2.0E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.23 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.55 ± 0.066 | 15.46 ± 0.066 | 2.3E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.31 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.58 ± 0.066 | 15.48 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.29 ± 0.065 | 5.5E-08 ± 3.0E-09 | | | | | |
| 15.55 ± 0.066 | 15.54 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.33 ± 0.065 | 5.3E-08 ± 2.9E-09 | | | | | |
| 15.55 ± 0.066 | 15.50 ± 0.066 | 2.1E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.29 ± 0.065 | 5.3E-08 ± 2.9E-09 | | | | | |
| 15.52 ± 0.066 | 15.47 ± 0.066 | 2.1E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.30 ± 0.065 | 5.5E-08 ± 2.9E-09 | | | | | |
| 15.53 ± 0.066 | 15.49 ± 0.066 | 2.1E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.30 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.48 ± 0.066 | 15.43 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.34 ± 0.065 | 4.8E-08 ± 2.6E-09 | | | | | |
| 15.55 ± 0.066 | 15.44 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.25 ± 0.008 | 15.30 ± 0.065 | 5.4E-08 ± 2.9E-09 | | | | | |
| 15.51 ± 0.066 | 15.44 ± 0.065 | 2.0E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.25 ± 0.065 | 4.8E-08 ± 2.6E-09 | | | | | |
| 15.54 ± 0.066 | 15.44 ± 0.065 | 2.0E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.26 ± 0.065 | 5.7E-08 ± 3.1E-09 | | | | | |
| 15.53 ± 0.066 | 15.46 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.25 ± 0.008 | 15.28 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.47 ± 0.066 | 15.47 ± 0.066 | 2.1E-04 ± 1.1E-05 | 0.24 ± 0.008 | 15.28 ± 0.065 | 5.5E-08 ± 3.0E-09 | | | | | |
| 15.54 ± 0.066 | 15.42 ± 0.065 | 2.1E-04 ± 1.1E-05 | 0.23 ± 0.008 | 15.24 ± 0.065 | 5.2E-08 ± 2.8E-09 | | | | | |
| 15.51 ± 0.066 | 15.47 ± 0.066 | 2.2E-04 ± 1.2E-05 | 0.23 ± 0.008 | 15.26 ± 0.065 | 5.5E-08 ± 3.0E-09 | | | | | |
| 15.51 ± 0.066 | 15.43 ± 0.065 | 2.2E-04 ± 1.2E-05 | 0.24 ± 0.008 | 15.30 ± 0.065 | 5.7E-08 ± 3.1E-09 | | | | | |

Table 4 (continued): Advanced Instrumentation Run (Values in kg/hr)

| RE Product Tank | | TRU Holding Tank | | Plant Inputs (kg/hr) | | Plant Outputs (kg/hr) | | In-Process Change (kg/hr) | | Inventory Difference (kg/hr) | |
|-------------------|---------------|------------------|---------------|----------------------|----------------|-----------------------|---------------|---------------------------|---------------|------------------------------|--|
| 7.8E-25 ± 2.6E-26 | 0.00 ± 0.000 | 0.00 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | 0.000 ± 0.000 | |
| 5.5E-08 ± 1.8E-09 | 0.00 ± 0.000 | 1.453 ± 0.006 | 0.001 ± 0.000 | 3.079 ± 0.129 | -1.626 ± 0.135 | 2.2E-05 ± 7.4E-07 | 0.45 ± 0.002 | 1.851 ± 0.008 | 0.042 ± 0.000 | -0.410 ± 0.228 | |
| 1.8E-04 ± 6.0E-06 | 2.33 ± 0.010 | 1.909 ± 0.008 | 0.207 ± 0.001 | 1.682 ± 0.279 | 0.020 ± 0.288 | 5.6E-04 ± 1.8E-05 | 5.72 ± 0.024 | 1.935 ± 0.008 | 0.499 ± 0.003 | -0.017 ± 0.339 | |
| 1.3E-03 ± 4.2E-05 | 9.83 ± 0.042 | 1.931 ± 0.008 | 0.850 ± 0.004 | 1.266 ± 0.376 | -0.186 ± 0.388 | 2.2E-03 ± 7.4E-05 | 13.68 ± 0.058 | 1.932 ± 0.008 | 1.179 ± 0.006 | -0.254 ± 0.423 | |
| 2.9E-03 ± 9.6E-05 | 16.74 ± 0.071 | 1.938 ± 0.008 | 1.439 ± 0.007 | 0.715 ± 0.428 | -0.216 ± 0.443 | 3.8E-03 ± 1.2E-04 | 18.88 ± 0.080 | 1.944 ± 0.008 | 1.620 ± 0.007 | -0.178 ± 0.465 | |
| 4.9E-03 ± 1.6E-04 | 20.35 ± 0.086 | 1.934 ± 0.008 | 1.746 ± 0.008 | 0.296 ± 0.458 | -0.107 ± 0.474 | 4.8E-03 ± 1.6E-04 | 21.28 ± 0.090 | 1.939 ± 0.008 | 1.824 ± 0.008 | -0.121 ± 0.484 | |
| 5.1E-03 ± 1.7E-04 | 21.79 ± 0.092 | 1.932 ± 0.008 | 1.866 ± 0.008 | 0.113 ± 0.472 | -0.048 ± 0.489 | 5.6E-03 ± 1.9E-04 | 22.16 ± 0.094 | 1.936 ± 0.008 | 1.899 ± 0.009 | -0.029 ± 0.488 | |
| 4.5E-03 ± 1.5E-04 | 22.29 ± 0.095 | 1.938 ± 0.008 | 1.909 ± 0.009 | 0.027 ± 0.473 | 0.002 ± 0.490 | 4.9E-03 ± 1.6E-04 | 22.40 ± 0.095 | 1.944 ± 0.008 | 1.919 ± 0.009 | -0.018 ± 0.492 | |
| 5.3E-03 ± 1.7E-04 | 22.39 ± 0.095 | 1.937 ± 0.008 | 1.918 ± 0.009 | 0.003 ± 0.478 | 0.016 ± 0.495 | 5.5E-03 ± 1.8E-04 | 22.48 ± 0.095 | 1.939 ± 0.008 | 1.926 ± 0.009 | -0.005 ± 0.496 | |
| 5.9E-03 ± 1.9E-04 | 22.53 ± 0.096 | 1.939 ± 0.008 | 1.929 ± 0.009 | -0.003 ± 0.482 | 0.013 ± 0.499 | 5.4E-03 ± 1.8E-04 | 22.59 ± 0.096 | 1.940 ± 0.008 | 1.935 ± 0.009 | -0.031 ± 0.496 | |
| 5.8E-03 ± 1.9E-04 | 22.53 ± 0.096 | 1.935 ± 0.008 | 1.929 ± 0.009 | -0.027 ± 0.478 | 0.032 ± 0.495 | 5.6E-03 ± 1.8E-04 | 22.58 ± 0.096 | 1.933 ± 0.008 | 1.934 ± 0.009 | 0.003 ± 0.496 | |
| 5.6E-03 ± 1.8E-04 | 22.58 ± 0.096 | 1.933 ± 0.008 | 1.934 ± 0.009 | -0.004 ± 0.479 | 0.003 ± 0.496 | 5.6E-03 ± 1.9E-04 | 22.55 ± 0.096 | 1.934 ± 0.008 | 1.931 ± 0.009 | 0.007 ± 0.493 | |
| 5.7E-03 ± 1.9E-04 | 22.62 ± 0.096 | 1.940 ± 0.008 | 1.937 ± 0.009 | 0.008 ± 0.477 | -0.005 ± 0.494 | 6.1E-03 ± 2.0E-04 | 22.59 ± 0.096 | 1.939 ± 0.008 | 1.935 ± 0.009 | 0.005 ± 0.492 | |
| 6.7E-03 ± 2.0E-04 | 22.59 ± 0.096 | 1.939 ± 0.008 | 1.935 ± 0.009 | 0.000 ± 0.475 | 0.005 ± 0.492 | 5.7E-03 ± 1.9E-04 | 22.58 ± 0.096 | 1.936 ± 0.008 | 1.933 ± 0.009 | 0.000 ± 0.495 | |
| 5.7E-03 ± 1.9E-04 | 22.61 ± 0.096 | 1.942 ± 0.008 | 1.936 ± 0.009 | -0.008 ± 0.474 | 0.014 ± 0.491 | 5.6E-03 ± 1.8E-04 | 22.49 ± 0.095 | 1.935 ± 0.008 | 1.926 ± 0.009 | -0.012 ± 0.474 | |
| 5.3E-03 ± 1.7E-04 | 22.55 ± 0.096 | 1.937 ± 0.008 | 1.931 ± 0.009 | 0.020 ± 0.477 | -0.014 ± 0.494 | 5.4E-03 ± 1.8E-04 | 22.53 ± 0.096 | 1.934 ± 0.008 | 1.930 ± 0.009 | -0.015 ± 0.477 | |
| 6.1E-03 ± 2.0E-04 | 22.49 ± 0.095 | 1.940 ± 0.008 | 1.927 ± 0.009 | 0.000 ± 0.481 | 0.014 ± 0.498 | 5.7E-03 ± 1.9E-04 | 22.61 ± 0.096 | 1.932 ± 0.008 | 1.937 ± 0.009 | 0.019 ± 0.479 | |
| 5.7E-03 ± 1.9E-04 | 22.61 ± 0.096 | 1.932 ± 0.008 | 1.937 ± 0.009 | 0.019 ± 0.479 | -0.024 ± 0.496 | 5.7E-03 ± 1.9E-04 | 22.66 ± 0.096 | 1.938 ± 0.008 | 1.941 ± 0.009 | 0.016 ± 0.477 | |
| 5.5E-03 ± 1.8E-04 | 22.64 ± 0.096 | 1.941 ± 0.008 | 1.938 ± 0.009 | 0.009 ± 0.477 | -0.007 ± 0.494 | 6.0E-03 ± 2.0E-04 | 22.52 ± 0.095 | 1.938 ± 0.008 | 1.929 ± 0.009 | -0.024 ± 0.478 | |
| 5.2E-03 ± 1.7E-04 | 22.61 ± 0.096 | 1.936 ± 0.008 | 1.937 ± 0.009 | -0.004 ± 0.476 | 0.002 ± 0.493 | 5.6E-03 ± 1.8E-04 | 22.57 ± 0.096 | 1.941 ± 0.008 | 1.934 ± 0.009 | 0.017 ± 0.479 | |
| 5.6E-03 ± 1.9E-04 | 22.55 ± 0.096 | 1.950 ± 0.008 | 1.932 ± 0.009 | -0.016 ± 0.476 | 0.034 ± 0.493 | 6.3E-03 ± 2.1E-04 | 22.54 ± 0.096 | 1.942 ± 0.008 | 1.932 ± 0.009 | 0.007 ± 0.477 | |
| 5.6E-03 ± 1.8E-04 | 22.52 ± 0.096 | 1.934 ± 0.008 | 1.929 ± 0.009 | -0.033 ± 0.476 | 0.038 ± 0.493 | 5.7E-03 ± 1.9E-04 | 22.61 ± 0.096 | 1.935 ± 0.008 | 1.936 ± 0.009 | 0.028 ± 0.479 | |
| 5.2E-03 ± 1.7E-04 | 22.54 ± 0.096 | 1.932 ± 0.008 | 1.930 ± 0.009 | -0.022 ± 0.478 | 0.024 ± 0.495 | 5.5E-03 ± 1.8E-04 | 22.56 ± 0.096 | 1.942 ± 0.008 | 1.931 ± 0.009 | 0.006 ± 0.476 | |
| 5.6E-03 ± 1.9E-04 | 22.62 ± 0.096 | 1.939 ± 0.008 | 1.936 ± 0.009 | 0.012 ± 0.477 | -0.009 ± 0.494 | 5.5E-03 ± 1.8E-04 | 22.58 ± 0.096 | 1.928 ± 0.008 | 1.934 ± 0.009 | -0.008 ± 0.479 | |

Table 4 (continued): Advanced Instrumentation Run (Values in kg/hr)

5.5 Diversion Scenario Response

The same diversion scenario used in Chapter 4 for the baseline instrumentation list was also used here for the advanced instrumentation case. This diversion was a protracted diversion of 8 kg of Pu spread over 100 hours. Figure 12 shows the mass balance during the run.

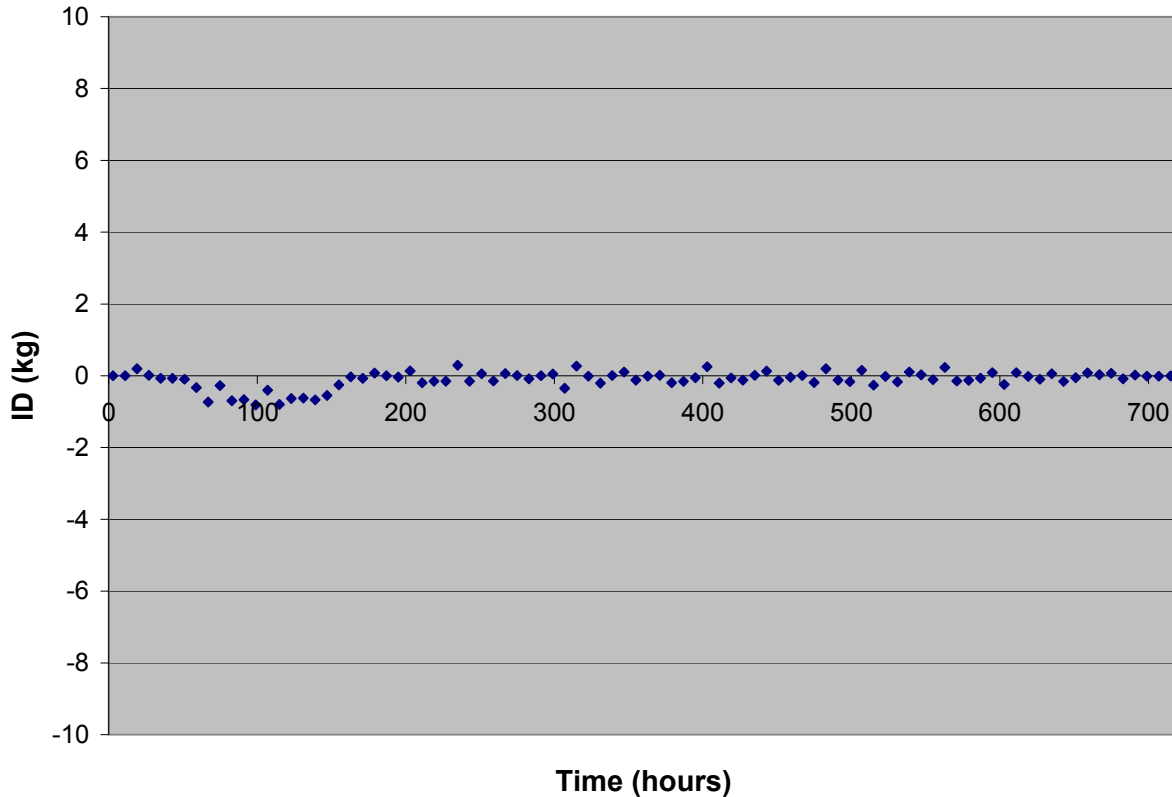


Figure 12: Inventory Difference with a Diversion Event using Advanced Accounting

Figure 12 clearly shows that the protracted diversion event will be detected. The detection will occur almost immediately when the inventory difference passes an alarm condition. Future work will put the statistics into the model to include the alarm conditions.

5.6 Advanced Instrumentation Requirements

As a result of this study, the following list of conclusions has been drawn to help guide the priorities for the development of new instrumentation:

- Existing measurements of U and Pu at accountability tanks and the front and back end of the process reach low levels of uncertainty and are unlikely to improve.
- Accountability advances at the front and back ends should instead focus on drastically reducing the measurement time through new instrumentation, more automation, or both.

- Low uncertainty measurements of solid spent fuel continues to be a gap in accounting—new instrumentation capable of measuring Pu and other species with less than 0.5% uncertainty should be a priority in the research.
- Measurements of in-process inventory, specifically Pu, can drastically improve the timeliness of detection while at the same time reducing plant construction and operating costs—these additional measurements will likely only be practical if a rapid, low uncertainty measurement technique is developed.
- The measurement of Pu in the UREX contactors will require an NDA technique due to the unique geometry and operation of contactors—5% uncertainty should be adequate for this measurement.
- Depending on the plant configuration, additional measurements of the other minor actinides (Np/Am/Cm) and some fission products (Tc/Cs/Sr) may be required—techniques like hybrid K-Edge densitometry should be investigated for use in measuring minor actinides, but traditional gamma spectrometry should be adequate for measurement of fission products of interest.

The advanced measurement technologies most likely to achieve these goals include the following:

- Ultra-High Resolution Gamma Spectroscopy or Microcalorimetry [11] – A non-destructive gamma ray spectrometer using arrays of microcalorimeters for measuring radioactive species, this technology has already been demonstrated to achieve a dramatic improvement in resolution compared to existing high purity germanium detectors. Such a technique may allow for low uncertainty measurements of solid spent fuel as well as solution from any processing vessel.
- Thermal Atomization Resonance Ionization Spectroscopy (TARIS) [12] – A destructive technique for rapidly measuring mixed reprocessing solutions with low uncertainty, TARIS could dramatically reduce the turn around time for analytical samples and open the door to additional sampling of in-process vessels.
- Hybrid K-Edge Densitometry (HKED) – HKED is well-established for measurements of U and Pu, but additional research is required to determine its use for measuring the other minor actinides in the transuranic product.

6.0 Conclusions

Some key qualitative conclusions have been drawn from the development and testing of the Safeguards Performance Model:

- As little as five additional plutonium measurement points will provide enough information to determine the plant inputs and outputs along with the change of in-process inventory with time.
- These additional measurement points will require either rapid, low-error mass spectrometry measurements or rapid on-line non-destructive measurements. Emerging technologies that may be best suited include TARIS (Thermal Atomization Resonance Ionization Spectroscopy) and Ultra-High Resolution Spectroscopy.
- A near real-time mass balance of the in-process Pu inventory change can drastically shorten the detection time of a misuse scenario by eliminating the need for a plant flush-out. At the same time, eliminating the need for a plant flush-out can save considerable operational costs.
- If a low error measurement of plutonium in spent fuel assemblies is available, it would also be useful to determine the in-process inventory of the vessels before accountability. Ultra High Resolution Spectroscopy may be best suited as an advanced measurement for this purpose.
- Uncertainty still exists as to how well the rest of the minor actinides and some of the key fission products need to be measured. Hybrid K-Edge Densitometry could be useful for these measurements. Experimental work is needed to determine the measurement precision of these species in mixed solutions.

Future work will expand the model to include more detail at the front and back end of the plant. In-process measurements of the front end need to be included along with the effect of adding a low-error measurement of spent fuel assemblies. The back end of the plant is missing the product conversion equipment to convert the liquid processing solutions to solid products and wastes. These areas will also need to be included in the in-process inventory analysis.

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