

Generic Quantitative Process Monitoring and Accountability Methodology for Fuel Cycle Facilities

Benjamin B. Cipiti
Sandia National Laboratories
PO Box 5800, MS 074
Albuquerque, NM, 87185-0747, USA
Tel: 1-505-284-8757, Email: bbcipit@sandia.gov

Thomas Severynse
Savannah River National Laboratory
Savannah River Site
Aiken, SC, 29808, USA

Candido Pereira
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL, 60439, USA

James Bresee
Office of Nuclear Energy
U.S. Department of Energy
Washington, DC, 20585 USA

Abstract – Nuclear material accountability in fuel cycle facilities is typically based upon the use of sampling and process status measurements to estimate material inputs, outputs, and in-process inventories. These measurements may include destructive or non-destructive analysis of process samples. However the time required for high precision analysis tends to limit the number of measurements that are practical, resulting in long material balance periods and hampering the timely detection of plant anomalies. Traditional nuclear material balance approaches are well-defined, but the availability of new process monitoring technologies suggests revisiting the methodology. Process monitoring technologies may include bulk measurements like flow, mass, density, temperature, voltage, etc. or chemical composition via at-line spectroscopies such as Ultraviolet-Visible-Near Infrared (UV-Vis-NIR) or gamma analysis. The accumulated data may also feed into process models to provide better estimates of in-process inventories prior to ultimate confirmation via sample analysis. Because process monitoring measurements can be made more often (or continuously) with minimal process impact, they make practical near-real-time accountancy (NRTA). One of the challenges to this approach is determining how the data can be used without creating more burden for the operator. The purpose of this work was to develop a generic quantitative methodology for integrating process monitoring measurements and advanced process modeling with traditional accounting to identify potential improvements in process control and safeguards monitoring. This includes developing a scheme for determining an appropriate NRTA balance period based on the implementation of process monitoring, and the use of the collected data to make qualitative decisions in a quantitative environment. Experimental work at the H-Canyon reprocessing facility at Savannah River National Laboratory, including testing of process monitoring technologies, was used to inform this work. However, the larger goal was to create a generic methodology that would be applicable to any fuel cycle facility and possibly other chemical processing facilities that have similar needs.

I. INTRODUCTION

Traditional nuclear material accounting at fuel cycle facilities is limited by both the timeliness and uncertainty of safeguards measurements. Precision measurements typically require sampling and laboratory analysis, which takes time. A full material balance requires measurements of inputs and outputs and the in-process inventory change, which can be a considerable challenge for an analytical laboratory.

The measurement uncertainty is also limited by technology and the buildup of error from multiple sampling points. The need for a quicker turn-around-time may require non-destructive analysis (NDA) which has higher uncertainty, further complicating the analysis.

The purpose of this work is to develop a safeguards approach and methodology that uses process monitoring to both improve the detection timeliness of abnormal events and improve detection sensitivity. This work moves closer to the ultimate goal of NRTA of providing timely

safeguards data that allows an operator to pinpoint problems and respond as soon as possible. This work is focused on domestic safeguards since it is reliant on process monitoring data, which can be difficult to authenticate for international safeguards. However, the approach presented here could be adapted for international use.

II. BACKGROUND

Nuclear material accounting at U.S. facilities and facilities under international safeguards is based predominately on a Physical Inventory Taking (PIT).¹ The PIT may be done yearly, every 6 months, or more often depending on the facility and applicable regulations. In each of the material balance areas, the sum of the inputs, outputs, and change of in-process inventory is determined.

The H-Canyon facility at Savannah River National Laboratory is an example of a facility operated under U.S. Department of Energy (DOE) regulations. This facility performs a PIT every two months.² Material is drained into vessels that can be sampled for accountancy. Depending on the solution or material type, different laboratory analyses are used. H-Canyon is being used to test new safeguards and process monitoring technologies and will provide important experimental data. Some of the technologies being tested at H-Canyon are presented in the next section.

The Rokkasho Reprocessing Plant (RRP) in Japan is an example of a facility under International Atomic Energy Agency (IAEA) regulations. A PIT is performed yearly with a plant flushout to drain in-process material. However, RRP also uses an Interim Inventory Verification (IIV) to perform a material balance on a monthly basis. This requires a combination of measurements and estimates of in-process inventory from a large number of vessels. A random sampling procedure is used to sample a small subset of the vessels every ten days. Bulk process monitoring (PM) data is included as part of the IIV via the Solution Measurement and Monitoring System.³

The approach presented here is intended to provide a plant balance much more frequently, on the order of daily or even hourly. The goal is to provide a safeguards system that is robust and that will timely detect many different types of material loss.

III. PROCESS MONITORING TECHNOLOGIES

The term “process monitoring” is often used in different ways. The DOE definition is useful, and states “Process Monitoring is a methodology to ensure that special nuclear material is in its authorized location, and when effectively implemented, it is a useful tool to detect anomalous process conditions and indicate losses of special nuclear material well before the scheduled physical inventory.”⁴

PM measurements can cover a broad range of instruments in nuclear facilities. Bulk measurements such as level, volume, density, flow, temperature, current, voltage, and pH are routinely used for process control at existing facilities. Additional technologies may include off-gas monitors or at-line spectroscopies for monitoring chemistry, activity, or isotopic content. The distinguishing feature is that these are continuous, on-line measurements.

III.A. Bulk Measurements

Bulk measurements are used routinely in nuclear facilities and have well-established uncertainties.⁵ Load cells and electronic balances can determine mass with 0.05% error. Electromanometers are used for level, density, and volume, and have uncertainties ranging from 0.05-1% depending on the accuracy required. Flowmeters can measure flow rates with uncertainty from 0.1-1%. These measurements can directly be used for bulk mass or volume balances.

Temperature, current, voltage, power use, capacitance, heating/cooling required, and off-gas monitors can also provide useful data, but in a more indirect form. Usually, some type of model or correlation developed from data from past experience is required to use this information to estimate material content.

III.B. UV-Vis-NIR Spectroscopy

Ultraviolet-Visible-Near Infrared (UV-Vis-NIR) spectroscopy⁶ uses a light source to produce a chemical spectra of a liquid process solution. It can be used to determine if the correct chemicals are present and the concentrations of specific species. The advantage of this measurement is that it is a NDA and can be applied to a sample or process stream. The counting time is on the order of seconds, so it can provide continuous monitoring.

Past work has shown the ability to determine U and Pu content to 1-2%,⁶ which is higher than other analytical techniques. However, the instrument is simple and cost-effective and only requires a fiber-optic probe in the region of interest. All data processing can occur outside of the hot cell.

III.C. MIP Monitor

The Multi-Isotope Process (MIP) monitor⁶ is a gamma analysis technology for determining change in isotopic content in a stream or sample. One key advantage is that it can be used on both liquid and solid material streams. The MIP monitor focuses on comparing the spectra with known or normal spectra in order to determine if the process stream is at the conditions expected.

The MIP monitor uses principal component analysis to automatically evaluate the patterns of gamma-emitting isotopes. In the future, the technology will be tested at H-

Canyon to determine measurement uncertainties. It may have applications in reprocessing where UV-Vis-NIR spectroscopy cannot be used.

IV. UNIT OPERATION MODELING

Computer models of unit operations can play a role in process monitoring as well. For areas that are particularly difficult to measure, models provide a way to estimate content. Dissolvers, pulsed columns, centrifugal contactors, mixer-settlers, and conversion processes all contain varying material forms, so obtaining representative and timely in-process samples could be difficult.

A fully validated unit operations model can be used to estimate effluent compositions and in-process inventory based on measured feed compositions, density, mass, pH, flowrates, temperatures, and other bulk process monitoring measurements. However, the models require extensive validation which in-turn requires a collection of experimental data obtained both at the lab-scale and on prototypic equipment. Experimental data in combination with standard statistical analysis techniques can be used to determine the uncertainties associated with model simulation results. It is unlikely that modeling will be able to determine content to less than 3-5% uncertainty, but for some areas of a plant, this uncertainty may be acceptable. Certainly, when integrated within a PM scheme, validated models can provide confirmation that a process is functioning as expected, and identify the source and mitigation of a problem when it is not.

Past work has examined the integration of U.S. solvent extraction codes and unit operation models with safeguards modeling.⁷ This was done to add more fidelity to the safeguards modeling efforts, but it could be extended to directly rely on the codes to determine actinide inventories based on PM data. The AMUSE (Argonne Model for Universal Solvent Extraction)⁸ was developed for steady-state flowsheet design for the UREX+ suite of extraction processes. The Nitron code⁹ is a continuous, rotary dissolver model developed at Oak Ridge National Laboratory that was also examined for integration with safeguards modeling. These are just two examples from past work, but most bulk handling facilities will have their own models that can be integrated with the safeguards system.

V. INTEGRATION OF PM DATA

There are two approaches for using PM data to provide more timely information. A qualitative approach uses PM data to inform the operator if process conditions do not look correct. This could involve visual monitoring of the data or bounding conditions that are set on PM variables. Usually, additional sampling and analyses would be required for confirmation if an anomaly were detected. The difficulty with a qualitative approach is that

it tends to be based more on facility operation experience and therefore could be more reliant on the knowledge base of the operator or analyzer—introducing a much greater chance for human error.

The more formal approach is to directly perform frequent material balances using the PM data. This approach is preferred because it is quantitative and can be automated. Also, this more formal approach should make it possible to better identify any anomalies.

This paper uses an aqueous plant as an example to demonstrate the methodology, but the concept can be extended to other reprocessing concepts and other bulk handling facilities.

V.A. Quantitative Approach

The ultimate goal of NRTA is to complete a plant balance daily or more frequently without causing a significant burden to the operator. The first step to integrating PM data is to provide a bulk material balance across individual unit operations or process control units. For areas processing liquid solutions where density does not change over time, a volume balance is possible. Flowmeters are used for inputs and outputs, and electromanometers are used for volume.

For areas processing solids or where densities are changing, a mass balance is needed. This requires mass measurements of the inputs and outputs (or flowrates and density measurements) and load cells on the vessels. Note that load cells may not be feasible on all vessels.

These bulk balances have low overall error since the measurements themselves can have uncertainties at or below 0.1%. Therefore, the bulk balance provides excellent detection probability for direct loss of material. These balances can be calculated daily or even hourly, which provides very timely detection of anomalies. The disadvantage is that bulk balances may not detect substitution diversions, when the target is replaced with an equal volume or mass of another material.

Spectroscopies and computer models can be used to fill in the gap to provide detection of substitution diversions. For example, if UV-Vis-NIR spectroscopy is used to monitor actinide content in various streams or vessels, a substitution diversion would cause a decrease in actinide concentration. The bulk material balance would not indicate a problem, but the combination of both systems would be effective to detect both direct and substitution diversions.

The quantitative approach uses the concentration measurements from the spectroscopies along with the bulk measurements to perform a complete material balance frequently.

For areas where either the bulk measurements or the spectroscopies will not work well, computer models can be used. For example, the separations processes (either in pulsed columns, mixer-settlers, or centrifugal contactors)

cannot be conveniently sampled to determine total actinide content since the quantities in different phases vary. Computer models can be used to estimate content in those vessels using PM data such as feed density, cold chemical flows, temperature, etc. One key advantage is that the total actinide content in these processes is much lower than in the buffer and feed tanks before and after, so the measurement uncertainty does not need to be low ($\pm 3-5\%$ is adequate).

Figure 1 shows how the PM measurements can be applied to the front end of a typical PUREX reprocessing

plant. In MBA 1, the quantity of actinides in used nuclear fuel can only be determined through fuel history and burnup codes, and that data can be used to estimate content before dissolution. Mass measurements may be used throughout to provide a continuous bulk balance. The accountability tank is measured well through traditional sampling and laboratory analysis, and that datum can be used to estimate content in the buffer tanks. The solid wastes are measured for accountability, which can also be used as part of the inventory balance.

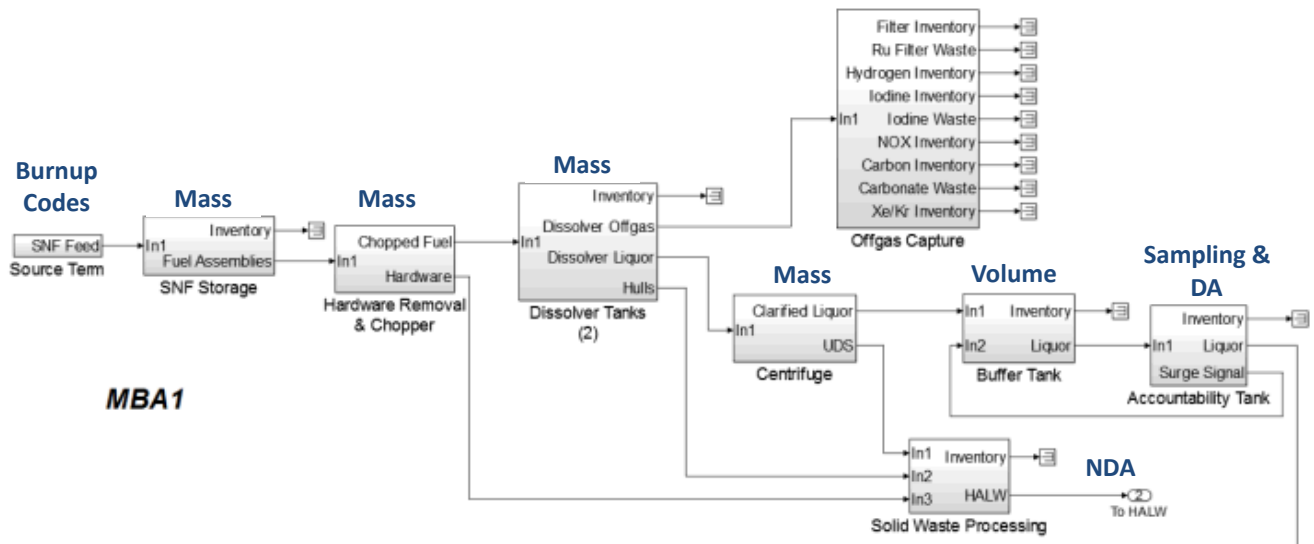


Fig. 1. MBA 1 PM locations.

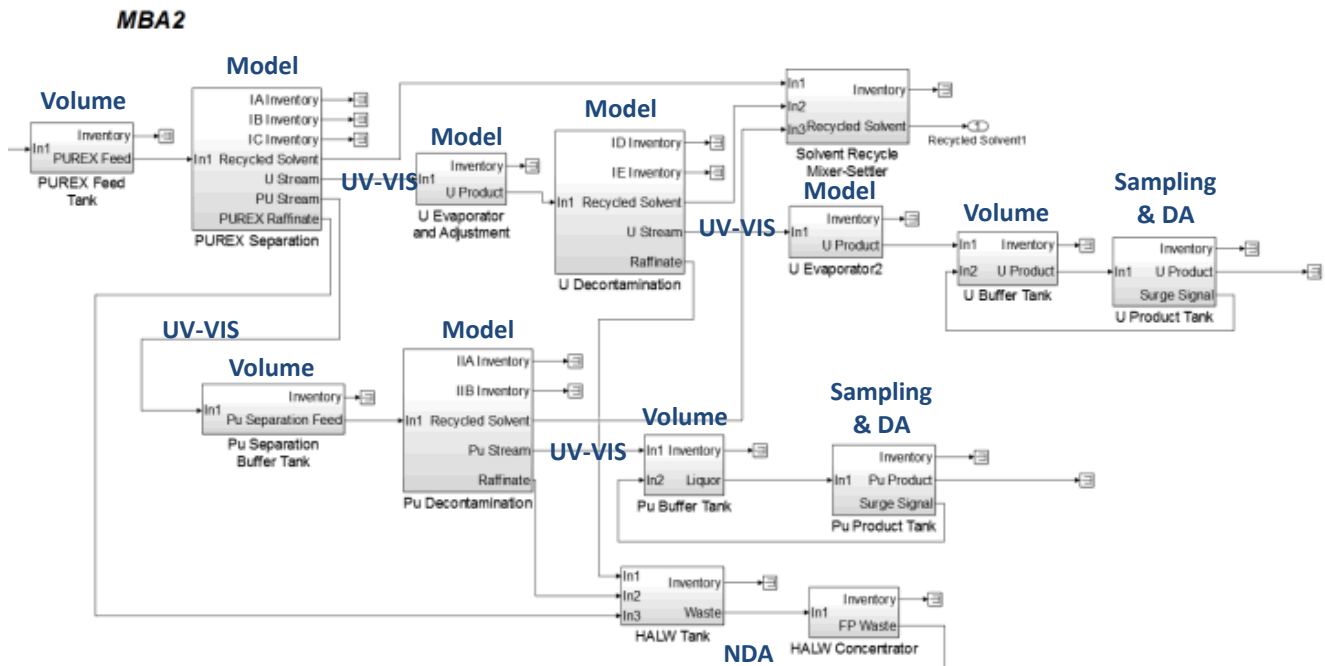


Fig. 2. MBA 2 PM locations.

MBA 1 continues to be limited by the high uncertainty on the input. Mass balances can provide protection from direct loss, but substitution losses would be difficult to detect. For this reason, containment and surveillance is relied on heavily in this area. Also, the fuel form is not an attractive target in this location.

Figure 2 shows how PM can be applied to MBA 2, or the separations area of a PUREX plant. This area has excellent measurements of inputs and outputs since the material is in solution throughout.

Bulk mass or volume balances can be setup on each unit operation in MBA 2. UV-VIS-NIR spectroscopy is more easily applied to flow streams, so it can be used to monitor concentration going into tanks. Computer models would be used for the separation steps and for evaporators based on PM data.

Product conversion is the next step of this process, and is being evaluated currently. There are a number of challenges in implementing continuous monitoring of oxide products. RRP uses the Plutonium Inventory Measurement System (PIMS) to keep track of material in this area, but uncertainties are limited to around 6%.¹⁰ Future work will examine this area in more detail.

VI. SAFEGUARDS PERFORMANCE MODELING

The Separation and Safeguards Performance Model (SSPM),^{11,12} developed at Sandia National Laboratories, is being used to model this PM approach. The SSPM is a transient reprocessing plant model built in the Matlab Simulink software. It models the various unit operations and tracks nuclear and bulk materials through the processes, but the main purpose of the model is to examine safeguards measurements and approaches. It provides the ability to set up various inventory balances, examine the integration of PM data, and model material loss scenarios to examine safeguards performance.

In past work the SSPM was used to model a traditional PIT or IIV,¹³ and bulk PM balances were added to the model. In this work, the on-line use of spectroscopies and unit operation models are being added to the SSPM to evaluate more frequent material balances.

A large number of measurement blocks are included in the model to represent both PM and traditional accountability measurements. These blocks include the random and systematic errors which can be changed by the user. The model uses those errors to simulate a measurement with errors while the model runs. All of the measurements are used to calculate a material balance. A great deal of attention goes into the delays of measurements so that the material balance is set for a certain point in time.

Diversion blocks are included to test the performance of the safeguards system against material loss. These blocks allow the user to set the starting time, stopping time,

location, fraction of material diverted, and choice of direct or substitution scenario.

VI.A. Generic Methodology

The methodology for integrating PM data involves setting up much more frequent material balances using both traditional accounting and PM data and applying a statistical test for detecting material loss. The SSPM uses a joint Page's test which is tuned for detecting both abrupt and protracted loss. Page's test is an accepted statistical test for this situation.^{14,15}

Precision measurements are taken for each input and output accountability batch though sampling and laboratory analysis. These are key measurements for completing a PIT or IIV, but the analysis time makes it difficult to rely on for a frequent PM balance. As an example, the plutonium product tank in RRP has a five day cycle time. A daily balance would need to be delayed by five days plus the sample turn-around-time in order to use that information. This is part of the reason why an IIV is unlikely to be performed any more frequently than every ten days.

A daily PM balance would need to instead rely on bulk balances and on-line spectroscopy measurements. In the SSPM, flowmeters and tank level measurements or mass measurements are assumed on every vessel. A bulk balance is performed for each unit operation as the model runs. Since the data can be generated continuously, the material balance period can be as frequent as desired, but false alarm probabilities associated with the statistical tests will be important to consider.

Throughout the MBA, spectroscopy can be installed on key product streams to monitor the concentration of Pu (and U if needed). Installation on streams may be more feasible than on tanks since the technologies only need to see a small cross-section of solution. The concentrations can be correlated with the vessel volumes on either side of the measurements, allowing for a calculation of the inputs, outputs, and many of the in-process inventories.

Bulk PM measurements can be used with computer models of separation processes and evaporators to determine in-process inventories in those locations.

This overall system will be able to provide a near real time actinide balance that can detect both direct and substitution loss. The overall measurement uncertainty for the material balance will be higher than for an IIV or PIT because the measurement uncertainties are higher. However, the main purpose of the PM balance is to provide more timely detection. The PM balance will mainly be looking for shorter diversion scenarios. One of the goals of this work is to determine the protracted limit of the PM balance.

For protracted, direct diversion scenarios, the PM balance will provide better sensitivity than the IIV or PIT. For protracted, substitution diversion scenarios, the IIV and

PIT can provide detection—since the material is being removed over a longer time, the IIV would have time to respond. This system will be limited by the measurement uncertainty for very long diversions, but that has always been the case for safeguards.

VI.B. Material Balance Calculation

The bulk material balance across each unit operation is straightforward. The Material Unaccounted For (MUF) is equal to inputs minus outputs minus the change in the inventory. Page's test is applied to each bulk balance to look for material loss.

Page's test can be applied directly to the series of MUF values using the following equations:¹⁵

$$S_i = MUF_i \quad (1)$$

$$S_i = \max(S_{i-1} + MUF_i - k, 0) \quad (2)$$

An alarm condition is reached when:

$$S_i > h \quad (3)$$

The h, k variables are tuned to achieve an optimal balance between sensitivity and false alarm probability. An abrupt loss will lead to a large MUF value that will pass the h threshold. For protracted loss over more than one balance period, if the average MUF is greater than k , the test may eventually reach the threshold. In the model, a no diversion case is run into order to set an optimal h, k to achieve a false alarm probability that is consistent with regulatory requirements.

The frequent plutonium balance is performed across each MBA, and so uses a large number of measurements. The spectroscopy data for plutonium concentration is used along with the bulk measurements to determine the input and inventories where applicable. For areas relying on models, the SSPM assumes the ability to estimate plutonium content with a specified uncertainty. (Note that the models are not integrated in the current model, but past work has examined this integration.)

In all cases, error propagation is tracked to determine overall measurement uncertainty. The errors are also required for the Page's test calculation. The Page's test results for all of the bulk PM balances and the Pu balances for each MBA are plotted as the model runs. Message blocks appear when a threshold condition is reached.

VI.C. Preliminary Results

Preliminary model runs were setup to examine the response to both direct and substitution material loss scenarios. Figure 3 shows the bulk material balance for a protracted diversion where plutonium nitrate was directly removed from a tank. About 1% of the solution was removed for 3000 hours, and the bulk PM balance was

easily able to detect the loss 200 hours into the diversion event. The figure shows the Page's test result for the bulk balance over that tank—the yellow line is the S value from equation 2, and the magenta line is the h value, or the threshold condition. This was a very protracted direct diversion, and any more abrupt diversion of this type would also be detected well.

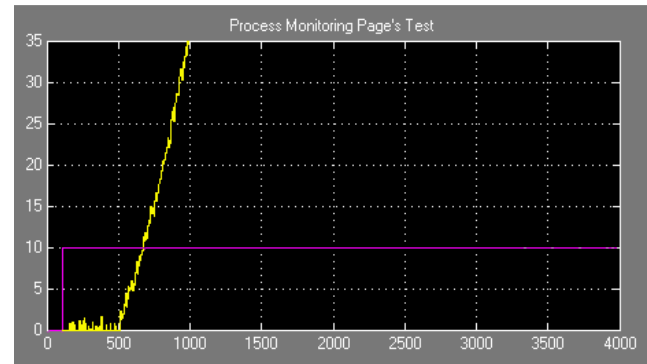


Fig. 3. Bulk PM balance during a direct loss.

Figure 4 shows the overall daily plutonium balance for a protracted substitution diversion lasting for 1000 hours. The assumption was that plutonium nitrate was removed and replaced with uranium nitrate. The figure shows the Page's test result for the actinide balance across MBA2 assuming the use of spectroscopy to provide more frequent material balances. The blue line is the threshold condition, magenta is for plutonium, and yellow is for uranium. In this case, the overall plutonium balance indicated the material loss about 250 hours from when the diversion started. The bulk PM balance (not shown) presented normal operation since material was substituted.

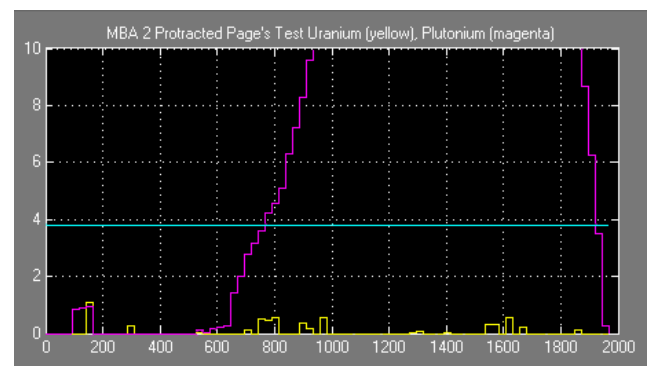


Fig. 4. Pu PM balance during a substitution loss.

These two examples show how the integrated PM system has the ability to detect both direct and substitution scenarios. Both of these examples were protracted cases—abrupt cases would have even better detection sensitivity, but future work will need to examine more scenarios.

V.I.D. Modification of the IIV

With a frequent PM balance, the requirement for an IIV needs to be revisited. Effectively, the PM data allows for a much more frequent IIV. The overall measurement uncertainty may be slightly higher, but the improvement to timeliness is useful.

The PIT will continue to require precision measurements for all input and output accountability tank batches. At the end of a campaign, after a plant flushout, this data will continue to be used to calculate the overall MUF.

There should still be an interim balance in order to detect moderately protracted substitution loss that might not be detected by the daily balance, but the approach could be easier than current implementation. The bulk material balance will continue to be effective for detecting direct loss even in protracted scenarios. A substitution scenario will cause the output accountability measurement to decrease while the input measurements stay constant. As long as the bulk balance is showing that the plant is in a steady-state mode, a drop in the output without a drop in the input indicates a substitution loss.

More formally, the precision measurements from sampling the input and output accountability tank could be used retroactively to update the daily PM balance. There would be a delay due to the batch sizes and sample turn-around-time, but the lower uncertainty data would increase the detection sensitivity for small material loss.

VII. DISCUSSION

The preliminary results used a simplified modeling approach that will need to be modified for future work. The proper integration of the bulk measurements with the spectroscopy data will need to be modeled more realistically. In addition, a comparison of the frequent PM balance along with the IIV and PIT will be useful to examine. A larger variety of diversion scenarios will be modeled to show robustness against different scenarios. Future work will also need to examine the product conversion area for potential use of daily PM balances.

VIII. CONCLUSION

This work presents a generic methodology for the integration of PM data with traditional nuclear material accounting. The approach uses bulk material balances along with spectroscopy technologies to perform a frequent (hourly or daily) material balance that will provide timely detection of process instability and/or material loss. This approach is designed to be robust for detecting both abrupt and protracted material loss, for both direct and substitution scenarios.

The precision input and output accountability measurements can be added to the balance retroactively to

increase detection sensitivity for protracted diversion scenarios. Future work will model this approach in more detail.

ACKNOWLEDGMENTS

This work was funded through the Fuel Cycle Technologies program under the U.S. Department of Energy Office of Nuclear Energy. Sandia is a multiprogram laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000.

NOMENCLATURE

IIV	Interim Inventory Verification
MBA	Material Balance Area
MIP	Multi-Isotope Process
MUF	Material Unaccounted For
NDA	Non-Destructive Analysis
NRTA	Near Real Time Accountancy
PIT	Physical Inventory Taking
PM	Process Monitoring
RRP	Rokkasho Reprocessing Plant

REFERENCES

1. *IAEA Safeguards Glossary, 2001 Edition*, IAEA/NVS/3, International Atomic Energy Agency (2002).
2. T. F. SEVERYNSE, "Baseline Accountability Measurement Error and Uncertainty for H Canyon Operations," FCRD-SWF-2014-000599, Savannah River National Laboratory (2014).
3. S. J. JOHNSON, "Development of the Safeguards Approach for the Rokkasho Reprocessing Plant," IAEA-SM-367/8/01, International Atomic Energy Agency (2001).
4. U.S. DOE Standard, DOE-STD-1194-2011 Change 3, "Nuclear Materials Control and Accountability," paragraph 6.2.4.5 (2013).
5. K. Zhao et al., "International Target Values 2010," STR-368, International Atomic Energy Agency (2010).
6. L. M. Arrigo et al., "FY2009 Progress: Process Monitoring Technology Demonstration at PNNL," PNNL-19136, Pacific Northwest National Laboratory (2009).

7. B. B. Cipiti and M. D. McDaniel, "Reprocessing Plant Scale Model Integration," SAND2012-7779, Sandia National Laboratories (2012).
8. M. C. Regalbuto et al., "Solvent Extraction Process Development for Partitioning and Transmutation of Spent Fuel," *Actinide and Fission Product Partitioning and Transmutation, Eighth Information Exchange Meeting*, Nuclear Energy Agency Organization for Economic Co-operation and Development, Las Vegas, Nevada (2004).
9. V. F. de Almeida, "Plant-Level Modeling and Simulation of Used Nuclear Fuel Dissolution," ORNL/TM-2012/375, Oak Ridge National Laboratory (2012).
10. D. Parvin, "Validation and Performance Test of the Plutonium Inventory Measurement System (PIMS) at the Rokkasho Reprocessing Plant (RRP)." British Nuclear Group (2007).
11. B. B. Cipiti et al., "Fully Integrated Safeguards and Security for Reprocessing Plant Monitoring," SAND2011-7292, Sandia National Laboratories (2011).
12. B. B. Cipiti, "Separations and Safeguards Performance Modeling for Advanced Reprocessing Facility Design," *Journal of Nuclear Materials Management*, 39/2 pp. 4-14 (2011).
13. B. B. Cipiti et al., "Cost Effective Process Monitoring using UV-VIS-NIR Spectroscopy," IAEA Symposium on International Safeguards: Linking Strategy, Implementation, and People, Vienna, Austria (2014).
14. B. B. Cipiti, M. McDaniel and T. Burr, "Incorporation of Page's Test in the Separation and Safeguards Performance Model," INMM Information Analysis Techniques and Methods for Safeguards, Nonproliferation and Arms Control Verification, Portland, Oregon (2014).
15. B. J. Jones, "Near Real Time Materials Accountancy using SITMUF and a Joint Page's Test: Comparison with MUF and CUMUF Tests," *ESARDA Bulletin*, 15 (1988).