LA-UR- 12-00908

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Title: Status Report for Development of Advanced Process Monitoring Concepts for IAEA Safeguards Sponsored by the Next Generation Safeguards Initiative (NGSI)

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Status Report for
Development of Advanced Process Monitoring Concepts for IAEA Safeguards Sponsored by the Next Generation Safeguards Initiative (NGSI)

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February 2012

ACKNOWLEDGEMENT

This work was sponsored by the United States Department of Energy, National Nuclear Security Administration (NNSA), Office of Nonproliferation and International Security (NA-24), Next Generation Safeguards Initiative (NGSI).

EXECUTIVE SUMMARY

Goals

Process monitoring (PM) is used in international safeguards as an additional measure to nuclear material accountancy (NMA). For large throughput nuclear facilities such as commercial spent fuel reprocessing plants, it is difficult to satisfy the IAEA's goal for detection probability using NMA alone. And, for all facilities, regardless of their throughput, PM can provide redundant verification for both NMA and containment and surveillance and can be used to detect abnormal plant operation through the use of IAEA's instrumentation and selected operator's process control instrumentation. Examples of PM include (1) continuity of knowledge of nuclear material flows and inventories by monitoring of tank solution levels in reprocessing plants, (2) load cell monitoring of cylinders in GCEPs, (3) thermal power monitoring of large research reactors to detect undeclared operation, and (4) providing data to enable frequent NMA-like evaluation (near-real-time-accounting), which involves a hybrid of NMA and PM, for many facility types. Sensors required for PM include for example sensors for measuring flow, volume, density, level, temperature, and other diverse sensors such as portal monitors, video cameras, motion detectors, and gamma and neutron detectors. Additionally, PM lends itself to remote and/or unattended monitoring which has the potential to reduce the IAEA inspection burden and intrusiveness to the operator.

Despite the ongoing successes of PM, a quantitative measure of PM's contribution to overall safeguards effectiveness has not been well developed. Therefore, the United States National Nuclear Security Administration (NNSA) has initiated a PM project through its Next Generation Safeguards Initiative (NGSI) to advance its use for international safeguards. The primary purpose of the NGSI PM project has been to demonstrate an approach/methodology for estimating the added value for safeguarding a nuclear facility with PM in addition to NMA alone. An established methodology to estimate the added value for PM will enable the designer and the IAEA to measure its contribution to the overall safeguards effectiveness for a facility, thus making possible a "cost versus effectiveness" tradeoff study. This tradeoff study could aid a Safeguards by Design (SBD) approach. The NGSI PM project objectives are consistent with future goals for the IAEA identified at the "Consultancy Meeting on Proliferation Resistance Aspects of Process Management and Process Monitoring/Operating Data" held in Vienna, 28-30 Sept 2011. These IAEA goals included a "proof-of-principle study on a well-known facility, demonstrating impact on efficiency and effectiveness" and a "proof-of-principle study on an advanced (future) facility, demonstrating that safeguards goals could be met using extended PM." Additionally, the PM project objective reported here is consistent with the NGSI goal of "Implementation of safeguards at declared facilities can be made more efficient and effective by incorporating advances in automation, measurement, and information technology" and "promoting Safeguards by Design as an international standard."

Accomplishments

Results to date for the NGSI PM project include (1) development of a methodology to determine the added value for PM in addition to NMA alone, and (2) use of the new methodology to identify existing operator and potential IAEA instrumentation not currently used for PM in nuclear facilities, that could enhance effectiveness of the facility safeguards design. For example, one approach for determining the added value for PM in addition to NMA alone can be quickly illustrated in the special case where PM and NMA measurements are essentially independent. Under the independence assumption, it is well known that the overall probability (P) of detecting material loss for a specific diversion is related to the combined probabilities of failing to detect loss with either NMA or PM as follows.

$$P(success_{combined}) = 1 - P(failure_{NMA})P(failure_{PM}).$$

Applying this approach (which relies on independent PM and NMA measurements) to a hypothetical diversion scenario described in this report, it is shown that combined NMA and PM can significantly increase the probability of detection over that for NMA alone. Additionally, it is shown how this approach can be used for SBD. The NGSI project team also considered examples where PM and NMA shared instrumentation in such a manner that the independence assumption is not appropriate; and consequently, other methods are being developed.

A spent fuel reprocessing facility was selected as the basis for developing the approach/methodology for several reasons. First, not only do reprocessing facilities have high plutonium throughput, but they also have a high in-process plutonium holdup during operation, which both significantly contribute to a large overall measurement uncertainty which leads to difficulty in satisfying the IAEA NMA-based detection goal. Second, although reprocessing facilities are heavily reliant on operator's process control data, historically the IAEA has not had full access to this data because of operator's proprietary concerns. The operator's existing process control data beyond that currently provided can provide a new data source for enhanced safeguards through PM, if proprietary and authentication issues can be overcome. Additionally, new PM techniques for reprocessing facilities are being developed within the DOE that can lead to enhanced safeguards. And finally, based on the Rokkasho experience, reprocessing plants consume an inordinate amount of IAEA resources, and PM is well-suited to remote and/or unattended monitoring which could reduce this burden.

To develop an approach to determine the added value for the use of PM in addition to NMA alone, specific capabilities required development. To begin, plant operating data for the facility of interest, that includes a diversion, is required to demonstrate the PM detection approach/methodology. This could be accomplished with actual plant data or simulated plant data. Actual plant data was collected and assessed for the Barnwell plant, Idaho Chemical Processing Plant (ICPP), Savannah River facilities, and Tokai pilot plant in Japan. Simulated data was derived from an approach using novel concepts for a model-based prediction of a monitored stream such as the leached hulls in the dissolver or the effluent stream from a separations column. In one approach, PM algorithms that are diversion path dependent were augmented with a path-independent algorithm that has the potential to detect any off-normal operation by using a multivariate outlier detection scheme that responds to any change from normal operation.

Specific tasks and the related accomplishments required to develop a methodology to determine the added value for PM in addition to NMA alone included: (1) develop a partial reprocessing plant model to simulate operating data representative of specific diversions, (2) identify diversion paths that can be used to demonstrate combined PM and NMA detection algorithms, (3) design and prototype PM detection algorithms using currently-available or potentially-available PM data, (4) develop advanced instrumentation for PM such as the Raman/UV-vis-NIR spectroscopic monitor and the Multi-Isotope Process (MIP) monitor with related authentication assessment, and (5) review actual facility data that included deliberate diversions and recover modest amounts of historic real facility data. These products and accomplishments are described in separate sections in this report and separate technical appendices for each sub-topic. This report also includes a survey of current uses for PM, possible application of the work to other (non-aqueous) bulk facilities, technology gaps, and ideas for future work.

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INTRODUCTION

The United States National Nuclear Security Administration (NNSA) has initiated a process monitoring (PM) project through its Next Generation Safeguards Initiative (NGSI) to advance its use for international safeguards. NGSI has five main pillars of which two are directly supported by the PM project. These two pillars are (1) Concepts and Approaches and (2) Technology Development. Both Technology Development and Concepts and Approaches are supported by the PM task through developing a methodology to measure the contribution of PM to the overall facility safeguards design. In future work, this measure could be used for "cost versus effectiveness" tradeoff studies to support "Safeguards by Design (SBD)."

PM has been defined by the IAEA as an element of a safeguards approach that monitors material, processes, and equipment (nuclear and non-nuclear) in all types of nuclear facilities, through independent and/or shared safeguards-relevant operator measurements[1]. It involves collection and evaluation of data on the flow of nuclear and other material, or on the status of a nuclear facility or its equipment. It is normally continuous and unattended, and the data may be transmitted to a central on-site location or back to IAEA headquarters. The use of this data for PM, as a central part of a safeguards approach, provides a higher level of operational transparency in a timely manner.

Current Application of PM for IAEA Safeguards

Making reference to the IAEA Policy Paper 16 (reissued 2009-10-15) on "Remote monitoring (RM) for safeguarding nuclear facilities", the IAEA characterizes monitoring data in three levels:

Level 1: Equipment State–of–Health Information
The transmission of state–of–health information (Level 1) of individual or all installed components of an RM system at a facility confirms that the system is functioning properly or provides an indication of malfunction or tampering. This lowest level could be a simple "OK" or "not OK" message, or an indication of failure of a specific system component or a report of a suspected tamper event.

Level 2: Summary Data
The transmission of summary data (Level 2) provides valuable information for preparation of inspection activities. In addition to Level 1 information, summary data could include the number of events recorded by the RM system, e.g., the number of items passing a detector, number of triggered image recordings, etc.

Level 3: Detailed Data
The transmission of detailed data from individual devices (Level 3) provides information which will be used in deriving safeguards conclusions. In addition to Levels 1 and 2, these data could include digital pictures, electronic seals or sensors status reports, NDA or other measurement results and other information.

Process monitoring is not a new technique and is currently used by the IAEA in a number of types of facilities where it provides added assurance to accountancy verification measures and/or aids in the early detection of misuse of a process or facility. A variety of data sources from either independent or shared monitoring systems are used. These data sources include flow rates, temperatures, pressures, volumes, acidity, voltage, electrical current, concentration, mass, reactant volumes and concentrations, off-gases, container item identification, radiation, etc.. Some of the specific uses for PM data include:

- Continuity of knowledge (CoK) of nuclear material flows and inventories, and of Design Information Verification (DIV) results;
- Portal monitors for storages;
- Thermal power monitors for large research reactors;
- Monitoring of uranium enrichment levels;
- Determination of in-process hold-ups and un-measureable inventories (UMI):
- Added assurance to high uncertainty accountancy and timeliness measurements;
- Optimization of inspection and/or measurement/sampling plans;
- Measurement data needed any-time/on-demand, such as for electronic mailboxes for Short Notice Random Inspections (SNRI);
- Support to Near-Real-Time-Accountancy (NRTA) methods and evaluations;
- Timely detection of process disruptions or equipment mal-functions;
- Assurance that operations are as declared; and
- Reduction of on-site inspector presence (inspection effort).

There are a number of technical issues which impact the effective and efficient implementation of PM as a safeguards technique, and will need to be addressed with further development work.[1] They are:

- Authentication of monitoring data originating from the operator's systems;
- Volume of data acquired (probably will perform data processing and reduction on-site),
- Security of data transmission;
- Development costs (e.g. evaluation software, sensors, and data collection and storage);
- Minimization of operating and maintenance costs;
- Protection of confidentiality of proprietary or sensitive data;
- Need for independent conclusion capabilities, particularly when sharing data with the operator;
- Resolution of the question of qualitative assessments vs. quantitative results in the facility safeguards effectiveness evaluation;
- Possibility of automated declaration (legal obligations); and
- Validation/benchmarking of simulation models.

In addition, there is a need for more robust, reliable on-line/in-vessel measurement systems with improved sensitivity. These systems should be capable of remote, unattended operations, while still providing inspector access for data retrieval and servicing.

Future Development of PM for IAEA Safeguards

The NGSI PM project objectives are consistent with future efforts proposed by the IAEA as identified at the "Consultancy Meeting on Proliferation Resistance Aspects of Process Management and Process Monitoring/Operating Data" held in Vienna, 28-30 Sept 2011. These proposed IAEA efforts include:

- Proof-of-principle study on a well-known facility, demonstrating impact on efficiency and effectiveness,
- Proof-of-principle study on an advanced (future) facility, demonstrating that safeguards goals could be met using extended PM.

The types of facilities suggested for the proof-of-principle studies include:

- Aqueous reprocessing,

- Pyroprocessing,
- OLR (CANDU) reactor,
- Enrichment.

ACCOMPLISHMENTS

Specific tasks and the related accomplishments required to develop a methodology to determine the added value for PM in addition to NMA alone included: (1) develop a partial reprocessing plant model to simulate operating data representative of specific diversions, (2) identify diversion paths that can be used to demonstrate combined PM and NMA detection algorithms, (3) design and prototype PM detection algorithms using currently-available or potentially-available PM data, (4) develop advanced instrumentation for PM such as the Raman/UV-vis-NIR spectroscopic monitor and the Multi-Isotope Process (MIP) monitor with related authentication assessment, and (5) review actual facility data that include deliberate diversions and recover modest amounts of historic real facility data.
These 5 accomplishments are described in separate sections in this report and in cited references. This report also includes a survey of current uses for PM, possible application to other (non-aqueous) bulk facilities, technology gaps, and ideas for future work. Briefly, accomplishments 1-5 are as follows.

1. The facility models include a dissolver model in the head-end, a separations model for the main reprocessing area, and a simulation model for buffer and control tanks surrounding a separations area. The dissolver model inputs include acid concentration, batch cycle times, and temperature, and the outputs are the relative amounts of Pu going to the spent hulls as waste and with the product. The University of Glasgow extended its open-source (Python) GU-RPSP (Glasgow University Reprocessing Plant Simulation Program) model for buffer and control tanks to include a separations area model (SEPHIS) that was first developed in the 1980s. Model 1 inputs include the tank layout, schedule of flow rate changes, and constituent masses for the tracked species such as U, Pu, nitric acid, and water. Model 1 outputs are the true constituent masses in each tank at each time step. Process variation effects and measurement error effects are modeled in model 2 at LANL by post-processing the model 1 outputs.
2. Simulated diversions specify an amount of Pu and a time pattern for removal, and then are characterized by their impact on normal operating data. For example, a sensor might tend to read "high" if Pu is present in unusually large amounts, and tend to read "normal" when Pu is present in typical amounts . Or, the sensor is characterized by its numeric response as a function of the amount of Pu present .
3. Diversion detection algorithms rely on either detecting unusual sensor patterns using discretized (such as "low," "medium" and "high") data over various time frames or on detecting numeric shifts of multivariate quantities from their normal ranges.
4. Development and evaluation of on-line instrumention to support NMA or PM or both includes the multi-isotope PM and UV-Vis.
5. Historic real data from INL (ICPP), SRS, Barnwell, and TRP have been made available at the unrestricted or OUO level. These real data sets are modest size but have not yet been fully analyzed. Unfortunately, the Barnwell data which was anticipated to be particularly valuable because of the known diversions did not prove to be as useful as hoped due to issues with the data that are described.

Three scenarios were developed in which it is difficult to detect by NMA alone, but were somewhat easily detected by NMA and PM. However, a full system that properly accounts for the large number of statistical tests per unit time has not yet been developed, so the false alarm rate of the combined NMA and PM system is still under evaluation. Scenario 1 involves directing excess Pu to the hulls by improper operation of a batch dissolver in the head end. Scenario 2 involves directing excess Pu to the waste stream from a separations area. Scenario 3 is a conceptual scenario developed to provide a simple illustration of combining PM and NMA data to increase overall detection probability in the case where PM and NMA data can be assumed to be independent.

Simulated Operating Plant Data with Diversions

Here we describe two diversion scenarios considered for a PUREX-type reprocessing plant, (1) incomplete dissolution of spent fuel and (2) removal of material from a solvent extraction unit operation. A representative composition of 40 GWd, 10-yr cooled fuel was obtained from the Characteristics Data Base of the LWR Radiological Data Base program. The operating conditions for the plant are based on an annual throughput of 1000 metric tonnes. Based on the given fuel composition, that equates to a daily throughput of 4700 kg of U and 53 kg of Pu.

Dissolver Scenario (1)

The extent of fuel dissolution was calculated as a function of operating temperature or initial nitric acid concentration using a time-dependent dissolver model based on the equations published by Koga *et al* in 1991.[2] The equations describe the dissolution of pure UO_2; for this study Pu is assumed to be substituted into the UO_2 structure and therefore, is released from the matrix at the same rate as the U.

The target U concentration was 300 g/L with a nominal extent of reaction of 99.9%. The time required for 99.9% dissolution at 368 K and $[HNO_3]_{initial} = 6.0$ M was calculated as 765 minutes. The new reference data was focused on the loss of 8 kg of Pu from the dissolver that has been assumed to remain undissolved and is removed from the system with the hulls. Four different time periods were assumed for the material loss: 15, 30, 60 and 90 days.

Table 1 summarizes the extent of dissolution for the base operating conditions of 368 K and $[HNO3]_{initial} = 6$ M, and when the diversion period is equal to the reporting period. The 8 kg Pu lost was converted to a fractional amount by dividing by the total amount of Pu processed over the operating time period. This fractional loss was then subtracted from 99.9% to give the extent of dissolution reached that would produce the loss of Pu. The required extent of dissolution was then used to "reverse engineer" the required (off-normal) time of operation and solution density. The density of the dissolver solution can be used as a direct indication of the extent of dissolution. As can be seen from the Table 1, a densitometer with an accuracy of <0.04% would be required for tracking the 8 kg loss of Pu over a reporting (diversion) period of 90 days.

The effects of changing both the initial HNO_3 concentrations (range of 4.5 to 6.5 M) and operating temperatures (range of 353 to 373 K) were also investigated. For example, a combination of 5.5 M initial HNO_3 concentration, 366.9 K and 765 minutes will lead to a 99.5% extent of dissolution. This is a temperature difference of 1.1 K and 0.5 M HNO_3 concentration (0.02 g/cc or 1.7%). In principle,

Table 1. For scenarios when the diversion period equals the reporting period.

Diversion period	Units	15 day	30 day	60 day	90 day
Total Pu processed in reporting time	kg	801.96	1603.92	3207.85	4811.77
Total daily Pu loss to achieve 8 kg loss in reporting time	Kg-Pu/day lost	0.5333	0.2667	0.1333	0.0889
Diversion extent of dissolution		98.90%	99.40%	99.65%	99.73%
Δ operation times	minutes	291	219	154	121
Δ density	kg/m^3	3.7	1.85	0.93	0.62
% change density		0.24	0.12	0.06	0.04

these differences are easily detected by current off-the-shelf PM technology. However, this PM approach requires dissolver batch measurements of cycle time, temperature, and nitric acid concentration. It also relies on operator declaration of fuel properties (which in principle could be verified, but is not currently verified by the IAEA) and on a dissolver model that has not been validated. Therefore although we suggest that inspections should use both neutron-based hull monitoring AND a model of dissolver operation with PM measurements, there are open challenges before PM benefits can be quantified for this scenario. Nevertheless, the prospect of quantifying the benefits of PM for this scenario while relying heavily on a dissolver model appears quite promising.

Separation Scenario (2)

Results from this study indicated that extraction and scrub parameters (such as flow rates and constituent concentrations) of the primary PUREX separation flowsheet could not be adjusted to a sufficient extent to redirect Pu to the raffinate stream. Another scenario considered for removing 8 kg Pu from the first Pu cycle was diversion of a small portion of the U/Pu loaded solvent before it is partitioned. For these calculations, four different reporting periods were again used: 15, 30, 60, and 90 days. This scenario assumes that the U-Pu loaded solvent exiting the extraction/scrub flowsheet is transferred to a temporary holding tank prior to entering the first Pu cycle flowsheet. We note here that this scenario requires attaching additional piping that might be high-risk for the adversary. However, the purpose of this calculation is solely to gauge an approximate response of the system with respect to such a scenario. If the flow rate leaving the U-Pu loaded solvent tank was increased so as to maintain the base flow rate of 63.7 L/min entering the first Pu Cycle at stage 11, a faster drop in the tank height would be observed.

For this example, a turbine flow meter with an accuracy of 1% of the reading is assumed. The change in flow rates needed are all less than the assumed accuracy of the baseline flow rate reading of 0.64 L/min. Thus, it may or may not possible to detect this small of change from flow rate measurements. However, using sequential statistical analysis techniques to monitor for trends, it may be possible to disengage such small changes from the instrument noise.

Other Simulation Capabilities

In addition to the modeling and simulation capabilities such as the dissolver model and the separations area model, Glasgow University has provided GU-RPSP (Glasgow University reprocessing plant simulation package) in open-source Python for simulating classic SM data consisting of tank volumes and mass for each of U, Pu, nitric acid, water, and "other," where "other" is all the unspecified fission products and other stream constituents[4]. Also, Sandia University is developing a Matlab/Simulink simulation of an aqueous reprocessing plant, which generates SM and some PM data[5].

Combined PM and NMA Detection Algorithms

Analysis of the multivariate time series of PM and NMA scores extends sequential analysis of the MB scores from NMA. Two possible analysis approaches are being pursued: "system-centric," and "multivariate sequential pattern recognition." Also, for illustration a simple approach with a numerical example that assumes NMA and PM scores are independent is provided in the separate Example Section.

For diversion scenario 1, solution monitoring (SM) as an example of PM could help verify declared dissolution time in the dissolver by confirming the dissolver cycle time. This requires attention to "solution monitoring scoring systems," because many tank cycle features will be monitored frequently[10]. There are various SM approaches[6-8] with a range of data assumptions, and some of the versions exist only as prototypes. However, for this scenario, we anticipate a mistake rate of essentially zero in recognizing for example the difference between a dissolver dissolution time of 765 minutes (nominal) and 561 minutes (which could send an extra 0.4% of Pu to the hulls if there were no processing change except for the shorter dissolution time).

For scenarios 1 and 2, Pu mass measurements in waste streams are a component of the material balance (MB, also known as the material unaccounted for, MUF), and these same measurements of waste stream Pu mass can be compared to the model-based predicted value, resulting in two correlated "scores," one score being the MB and another score being the comparison between predicted and measured waste stream Pu mass. In this case, scores from NMA and PM cannot be assumed to be independent, so options to combine non-independent scores are being developed in a prototype SM version[9].

System-Centric

Garcia et al.[10] describe a possible way ("system-centric") to combine multiple subsystems that relies on "anomalies unaccounted for (AUF)" The approach currently assumes that each subsystem is independent and uses a very specific alarm rule involving various sensors reporting either abnormal or normal status. It allows for the partial observation case, in which missing sensor information is inferred from other sensors. In addition, each sensor is characterized by a reliability defined by its false-pass and false-fail rates. It uses a discrete event model of operations and allows for inference of missing sensor values. This is an example of an overall system, and one could add NMA as a subsystem and treat NMA on the same footing, but not independent of SM.

Garcia et al.[10] report high DPs for Diversion Scenario 1 in the dissolver that NMA alone can detect only with very low DP. This approach works with categorical data from each sensor, such as L (low), M (medium), and H (high), allowing for tuned time-delays from some sensors to model a temporal trend. Figure 4 (a-c) illustrates high DP results for Diversion Scenario 1 for three values of sensor reliability. Sensor reliability determines, for example, a sensor's probability of correct classification into the L, M, or H categories. An example AUF would then be an "H" reading on a sensor that should read "M." Although the estimated DPs are very high, and there is clear separation between the non-anomalous and anomalous data (unlike in Figure 3 where group separation is not so dramatic), real plants are quasi-continuous, the approach is tuned to this particular scenario, and subsystems are currently assumed to operate independently (not true for SM and NMA for example). Therefore, additional development is required to modify the current system-centric approach applied to Diversion Scenario 1 using the dissolver model from Bakel et al.[3]

Complex nuclear fuel processing facilities need to be closely monitored for timely detection of process or equipment anomalies and assurance that plant operations are as declared. Using observation platforms for tracking and interpreting anomalies, optimal collection and evaluation of data are considered on the in-process flow of material and status of unit operations (UO) and the target monitored facility at hand. An example of an observation platform considered here for processing monitoring (PM) is shown in Figure 1.

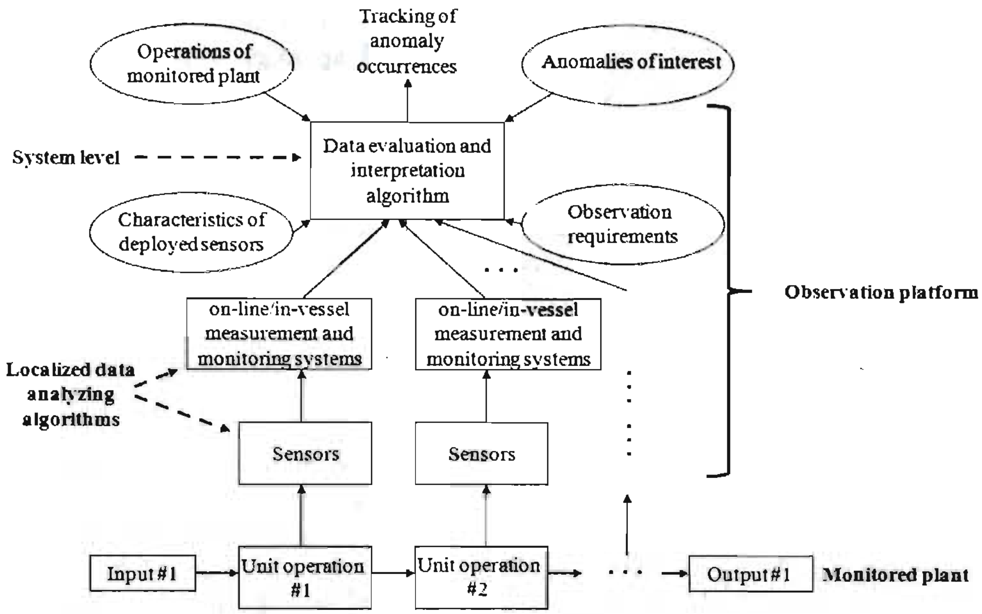

Figure 1. Example of an observation platform considered here for PM.

As illustrated in Fig. 1, sensors observe UO in the monitored facility. These sensors may include, for example, process monitors for measuring diverse process variables (e.g., flow, volume, density, level, temperature), on-line chemical component analysis, portal monitors (e.g., video cameras, motion detectors), and radiation monitors (e.g., gamma and neutron detectors). The sensor data are normally continuous and collected by unattended on-line/in-vessel measurement and monitoring systems (MMS), which may have automated data collection and computer-assisted data analysis capabilities. For example, if a unit operation is a chemical reaction vessel, an MMS may be an in-vessel solution MMS.
Data processing and reduction may occur by sensors at their locations (e.g., smart sensors) or at the MMS. For example, a sensor can report raw data (e.g., temperature of a vessel or γ-ray spectra of observed material), which is simply collected by the MMS without further analysis. Or, the raw data might be analyzed either at the sensor or at the MMS (e.g., using principal component analysis of γ-ray spectra to estimate constituents of observed material) to produce higher-level information. The concept of the raw sensor data being analyzed at the sensor location or at the MMS gives rise to the notion of (local) in-field diagnostics, and the data-fusion field recognizes situations where individual sensors should report either raw data, numeric "scores" such as "low," or an interval-valued score such as -2.5, or decisions, such as "alarm" or "no alarm."

Data collected and/or analyzed by MMS are then processed at the (global) system level by a data evaluation and integration (DEI) algorithm to detect and keep track of occurrences of anomalies of interest. The objective of a DEI algorithm for PM is to satisfy user specified observation requirements (e.g., probabilities of detection and false alarm) of anomalies being tracked. In order to accomplish this objective, the DEI algorithm utilizes knowledge regarding characteristics of deployed sensors and operations of the monitored facility. While the former may be specified by their manufacturers or determined by experiments, the latter is specified by reference signatures for the monitored UO, which are series of events in given logical, temporal sequences. Plant operation characterizations may be determined by the physical arrangement of the monitored facility at hand or from operational experiences.

Anomalies of interest for detection may represent undesired plant operations (e.g., loss of material) and are defined by specific signatures, such as patterns of events occurring at different locations of the monitored facility and at different stages of operations. The intent under PM is to detect these operational anomalies in real time rather than detecting them by measuring mass balance inconsistencies resulting from abnormal activities. PM methods and algorithms are being developed to improve the effectiveness and/or efficiency of anomaly detection and interpretation. The development of a rigorous framework is also being pursued for integrating data and optimizing observation platforms in order to effectively deal with complexity and optimization, amenable to synthesis and analysis.
There are two major challenges in developing DEI algorithms. The first is that information reported to these algorithms from sensors is often partial (e.g., it can be the case that no sensor is deployed to observe a specific process variable, such as temperature of a unit operation) and unreliable (e.g., the true temperature of a unit operation is low but, occasionally, the associated sensor may report a high temperature reading or even fail to report an observation). Part of the reason for the partial and unreliable information is that some process variables may be inherently difficult or impossible to measure accurately. A DEI algorithm for PM thus needs to effectively process the partial and unreliable information typically collected from multiple sensors. The second challenge is that facilities often consist of multiple UO. Anomalies in these multi-unit systems are often defined as patterns of events occurring at different UO and time instances (e.g., a high temperature indication in a particular UO, followed by a low concentration alarm of a specific material in another UO, and, eventually, an abnormal report triggered at a given output port). A DEI algorithm needs to recognize anomalies defined in this manner, where the events constituting them may span several UO and occur apart in time. Here, DEI algorithms for PM are being developed to process partial and unreliable information for detecting and tracking occurrences of anomalies defined as patterns of events. Additional detail is given in Appendix A.

To illustrate the benefits of PM for improved safeguards, consider a PUREX reprocessing facility with a daily total throughput of about 5 MTHM. Assume he anomaly of interest for detection manifests itself as a loss of a significant quantity (SQ) of Pu (i.e., 8 kg-Pu) caused by abnormally operating a dissolver in support of a protracted diversion of Pu to a retained (unconditioned) waste. For detecting the occurrence of this anomaly, consider two different DEI approaches. The first DEI approach is based on NMA, computing the MUF. The second DEI approach is based on PM, which does not directly do mass balance calculations, but rather monitors for the possible occurrence of anomaly patterns related to potential loss of nuclear material. It is thus assumed that the loss of a given mass amount of nuclear material can be directly associated with the execution (by the facility operator) of proliferation-driven activities that trigger the occurrence of an anomaly pattern consisting of series of events or signatures occurring at different unit operations and time instants. Instances of these anomaly events under the considered material diversion example are low temperature or low nitric acid readings at the dissolver, high neutron count readings on cladding hull batches, significant discrepancy between fuel batch estimations calculated at the Input Accountability Tank and corresponding shipper's fuel batch characterizations, and high neutron count readings when removing full hull drums with metal waste. By effectively integrating these events in time and space, the PM-based DEI approach tries to infer whether this specific pattern of events has occurred and how many times within a given time period, as these counts can be mapped into a certain amount of material unaccounted for. Making a correct inference is challenging considering that these constituting events may be unobservable or observed unreliably due to the absence of corresponding sensors and/or sensor unreliability characteristics.

To evaluate PM effectiveness, the 3 sigma (standard deviation) of the estimated mass loss is computed under both DEI approaches as a function of the number of input batches processed by the facility. As shown in Fig. 2, the PM-based algorithm performs better. Specifically, the 3 sigma for the PM-based method does not grow as fast as the 3 sigma-MUF for the NMA-based method. While the comparison here is done considering a scenario with a specific set of sensors and given sensor reliabilities, performance results obtained under the PM-based method still look more favorable for scenarios with more or less sensors. The main reason for this improved performance is that the PM-based method is designed specifically to detect the anomaly pattern considered, while the NMA

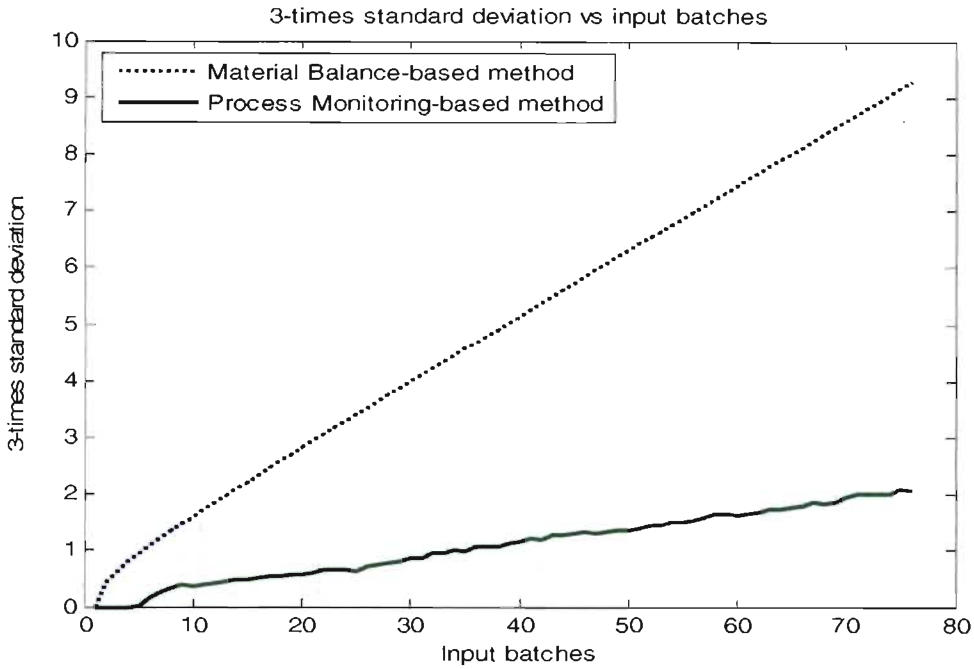

Figure 2. 3-sigma of mass loss estimates as function of number of batches processed.

method may detect any anomaly of the same nature (i.e., loss of mass). A related second reason for improved performance involves the fact that some PM can involve monitoring streams having small amounts of Pu, provided a model-based prediction for those streams is available. Obviously, the PM-based method can be accordingly extended to achieve much broader detectability characteristics.
The systems-centric approach assumes each subsystem is independent and uses a very specific alarm rule involving various sensors reporting either abnormal or normal status. It allows for the partial observation case, in which missing sensor information is inferred from other sensors. In addition, each sensor is characterized by a reliability defined by its false-pass and false-fail rates. It uses a discrete event model of operations and allows for inference of missing sensor values. This is an example of an overall system, and one could add NMA as a subsystem and treat NMA on the same footing, but not independent of SM. Garcia et al.[10] report high DPs for Diversion Scenario 1 in the dissolver that NMA alone can detect only with very low DP. This approach works with categorical data from each sensor, such as L (low), M (medium), and H (high), allowing for tuned time-delays from some sensors to model a temporal trend. Although the estimated DPs are very high, and there is clear separation between the non-anomalous and anomalous data, real plants are quasi-continuous, the approach is tuned to this particular scenario, and subsystems are currently assumed to operate independently (not true for SM and NMA for example). Therefore, additional development is required to modify the current system-centric approach applied to Diversion Scenario 1 using the dissolver model previously described.

Multivariate sequential pattern recognition applied to residuals/scores

As an example of PM, solution monitoring (SM) is a type of PM in which masses and volumes are estimated from frequent in-process level, density, and temperature measurements[6-9,11-17]. If each tank is regarded as a sub-MBA (material balance area), then transfers between tanks can be identified, segments of which can then be compared to generate volume and mass transfer differences (TDs) between tanks. A safeguards concern might then be raised if either these TDs or deviations in mass or volume data during "wait" modes (non-transfer modes) become significant. Average mass and volume TDs should be zero (perhaps following a bias adjustment) to within a historical limit that is a multiple of the standard deviation of the mass or volume TD, as should deviations during "wait" modes.

A residual (residual = measured – predicted) is generated each time a mode (transfer or wait) is completed by any tank. Such residuals ("scores") can be analyzed over time and over tanks. Analogously, in NMA one can analyze MBs for trends over time. Another approach to SM that relies on having consistent tank cycles defines a template signature for each tank and monitors each cycle for agreement with the historical template[8]. This alternate SM approach also generates residuals or scores.

Figure 3 plots example simulated NMA and PM scores for a 30-day balance period, with MBs every 10 days and PM scores from wait and transfer modes plus residuals from model-based predictions for some flow streams. Figure 4 is a two-dimensional representation of the scores in Figure 3 for a moderate and large loss. Reference [9] gives more details about analysis of much multivariate scores. Although these results are preliminary, the approach does allow for non-independence of the NMA and PM residuals.

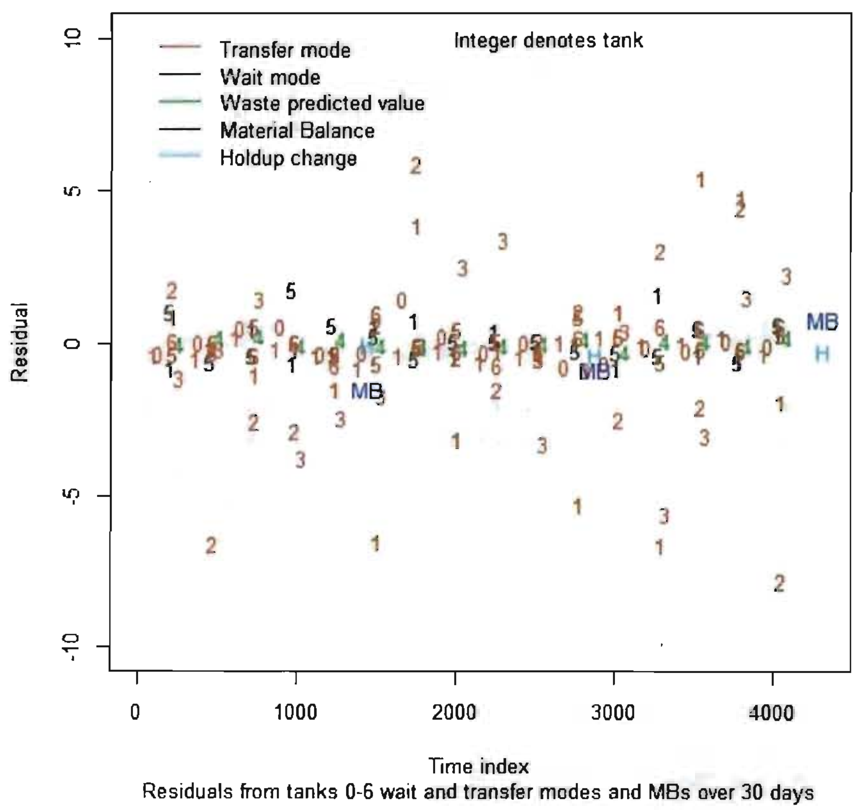

Figure 3. Residuals or "scores" from NMA and PM for a 7-tank MBA (tanks 0 to 6).

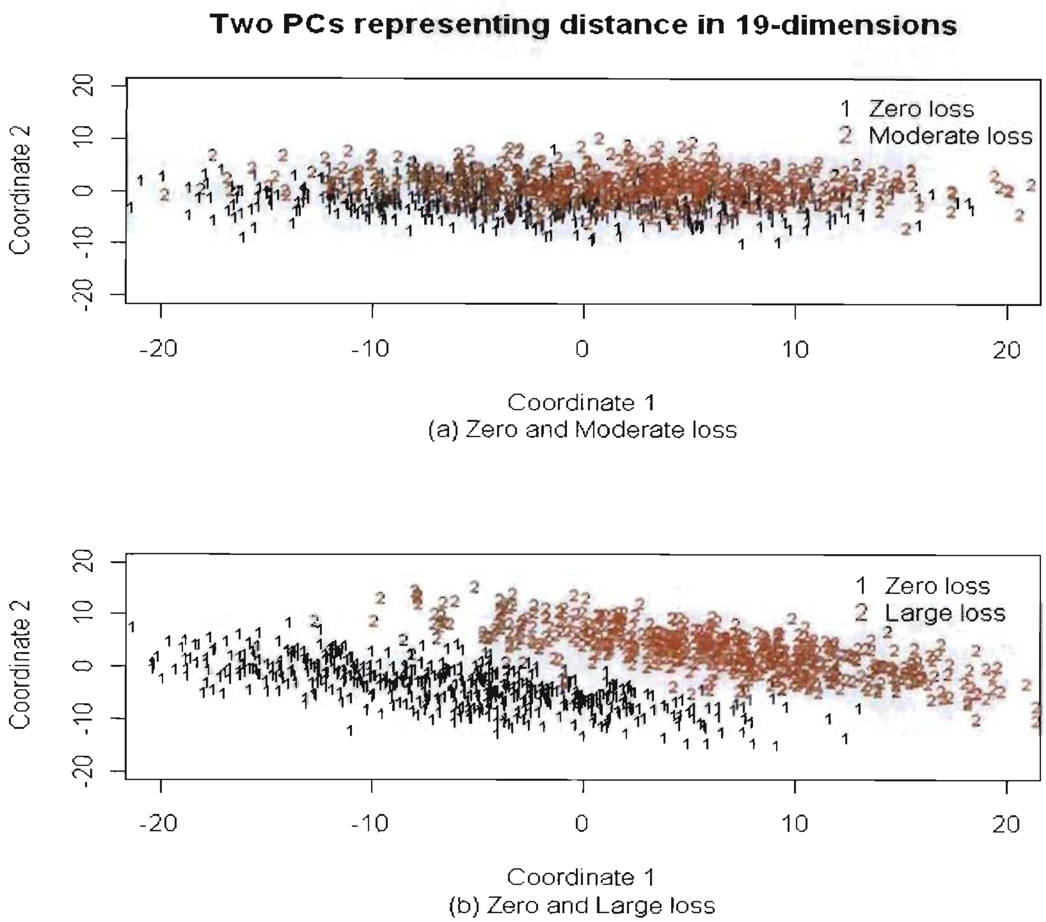

Figure 4. Qualitative assessment of the ability to detect moderate or large loss using scores as in Figure 3 from NMA and PM data. Two principal coordinates (similar to principal components) are used to show distances between 19-component realizations. Because Page's sequential test checks for temporal trends over the 30 days, Figure 4 is not intended as a check for trends, but is intended only to evaluate how detectable a moderate or a large loss is with one particular pattern recognition option.

Advanced Instrumentation and Monitoring Techniques Development

Advanced instrumentation and associated new monitoring techniques are being developed that provide new PM options[18-32]. Identifying physical characteristics the methods are measuring, along with the errors associated with the measurement, are essential tasks.

Three plots in this section illustrate accomplishments completed during FY11. Figure 5 illustrates optical PM techniques' ability to detect material diversion in near real-time. Figure 6 illustrates our preliminary understanding of uncertainties associated with the calibrated multivariate techniques used in the Multi-Isotope Process Monitor (MIPM). The error bars on the points show the relative error in the predictions and the uncertainty in the original measurements. The final section outlines preliminary investigations into using potentiometric sensors for inline spent fuel solution acid concentration measurements (Figure 7).

Spectroscopic Process Monitoring

Liquid-liquid extraction is a separation technique commonly employed for the processing of the dissolved spent nuclear fuel. Real time monitoring of the liquid-liquid extraction flowsheets provides opportunity to quickly detect unwanted manipulations with fissile isotopes present in the radiochemical streams during reprocessing activities. The instrumentation used to monitor these processes must be robust, require little or no maintenance, and be able to withstand harsh environments such as high radiation fields and aggressive chemical matrices. In previous years, the team experimentally assessed the potential of Raman and vis-NIR spectroscopic techniques for on-line real-time monitoring of the U(VI)/nitrate ion/nitric acid and Pu(IV)/Np(V), respectively, in solutions relevant to spent fuel reprocessing. Recently, the team demonstrated the real-time, on-line capability to use spectroscopic monitoring for safeguarding a continuous feed extraction system.

The ability to identify material intentionally diverted from a liquid-liquid extraction contactor system was successfully tested using on-line PM as a means to detect the amount of material diverted. A chemical diversion and detection from a liquid-liquid extraction scheme was demonstrated using a centrifugal contactor system operating with the simulant PUREX extraction system of $Nd(NO_3)_3$/nitric acid aqueous phase and TBP/dodecane organic phase. During a continuous extraction experiment, a portion of the feed from a counter-current extraction system was diverted while the spectroscopic on-line PM system was simultaneously measuring the feed, raffinate and organic products streams.

To test the on-line and near real-time aspects of PM, a counter-current liquid-liquid extraction testing apparatus was instrumented with visible, NIR and Raman spectroscopic probes. The counter-current extractor is based on multiple banks of 2-cm centrifugal contactors. The bank, consisting of four centrifugal contactors for this study, was installed and instrumented with vis-NIR and Raman spectroscopy probes, and with flow meters. Fibre-optic cables are used to connect the spectroscopic instrumentation to the solution probes attached to the centrifugal contactors. Flow testing of the centrifugal contactor system used feed solutions containing $Nd(NO_3)_2$, $NaNO_3$, and nitric acid. The organic solvent system used for this demonstration was 30% TBP/dodecane with an aqueous feed containing constant concentrations of $Nd(NO_3)_3$ and nitric acid.

Figure 5 contains a schematic representation of the bank of four contactors used in our initial study. The locations of feed, raffinate, organic inlet, and loaded organic product streams are shown. The vis-NIR and Raman monitoring probes are positioned on the feed, raffinate, organic inlet, and organic product streams. After the steady state flow for both aqueous and organic streams was sustained for 87 minutes, a diversion valve was opened at the entrance into the contactor feed inlet, and a fraction of the feed solution was diverted over the time of diversion. After approximately 47 minutes, the diversion of material was stopped (i.e., 134 minutes after the start of experiment), and the normal feed flow (i.e., with no diversion) into the contactor system was re-established.

Spectroscopic monitoring of the feed, raffinate, and organic product steams were recorded during the entire flow test. Fig. 6A contains the series of vis-NIR spectra measured at the organic product location during the extraction experiment. The vis-NIR spectra show the typical absorbance spectra associated with Nd^{3+} ion in solution, and show the increase in Nd^{3+} in the extractant phase after the initiation of the experiment and a plateau of absorbance after about 20 min after the start of the experiment. At the approximate point of diversion (87 min) the absorbance value for the Nd^{3+} band significantly decreases, and stays at a suppressed level until the diversion was stopped (at time = 134 min), after which the measured absorbance increased to its pre-diversion value.

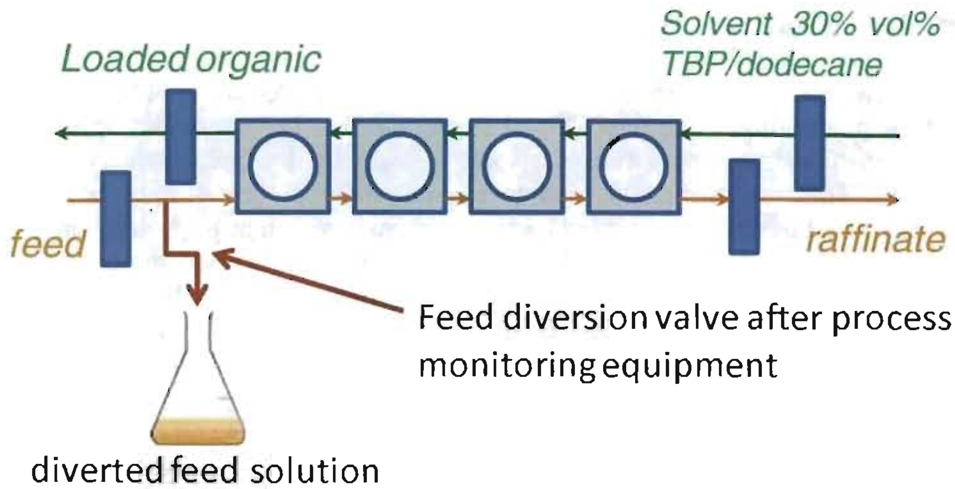

Figure 5. Schematic representation of the bank of contactors used in our study; the feed, raffinate, and loaded organic product streams are instrumented with vis-NIR and Raman probes as well as flow meters.

By combining the flow rate information (recorded using in-line flow meters) with the concentration of Nd^{3+} in each phase (determined from the spectroscopic data taken over the course of the experiment), we are able to determine the cumulative total (integrated amount) of Nd^{3+} in each contactor inlet and outlet stream. Fig. 6B shows the integrated total of Nd measured in the feed, and raffinate plus organic product during the entire experiment. The sum of the quantity (in mmol) values for the organic product and raffinate steams (labeled "organic + raffinate", in Figure 6B) should equal the total quantity (in mmol) of Nd^{3+} measured for the feed. The curve for the "organic + raffinate" is parallel with that for the feed measurement prior to the diversion point at 87 min into the experiment. There is a constant difference between the "organic + raffinate" and feed due to in-process amount of Nd^{3+} which is still within the contactor system and not yet measured by the spectroscopic probes on the outlet of the system. Fig. 6B also shows the difference (delta) between the inlet (Feed + Solvent) minus the outlet (Raffinate + Organic Product) streams during the solvent extraction experiment. This difference in measurement is labeled "delta from in-process" within Fig. 6B, and is the difference in mmol between the two curves (delta-y on xy-plot). After the start of diversion, at 87 min, the "organic + raffinate" curve in Figure 2B further deviates from the "feed" curve, and during the time in which diversion is occurring (between 87 min and 134 min) the two curves are no longer parallel. After diversion is stopped (at 134 min) the "organic + raffinate" curve then returns to being parallel with the "feed" curve. By extrapolating a line from the "organic + raffinate" curve prior to diversion, we are able to measure the amount of Nd^{3+} material diverted by subtracting the extrapolated value (prior to diversion) from the measured value after diversion. The "delta from diversion" is also shown within Figure 6B. This difference was measured to be 3×10^{-3} mol Nd^{3+} diverted based on the graphical analysis of the data in Figure 6A. This value is in excellent agreement with the value based on mass balance values, which is 2.9×10^{-3} mol Nd^{3+}.

The amount observed to be diverted by on-line spectroscopic PM was in excellent agreement with values based from the known mass of sample directly taken (diverted) from system feed solution. We conclude that near real-time spectroscopic PM is a useful tool for the immediate detection of diverted material. This summary details our methodology of on-line PM and shows results of this specific example.

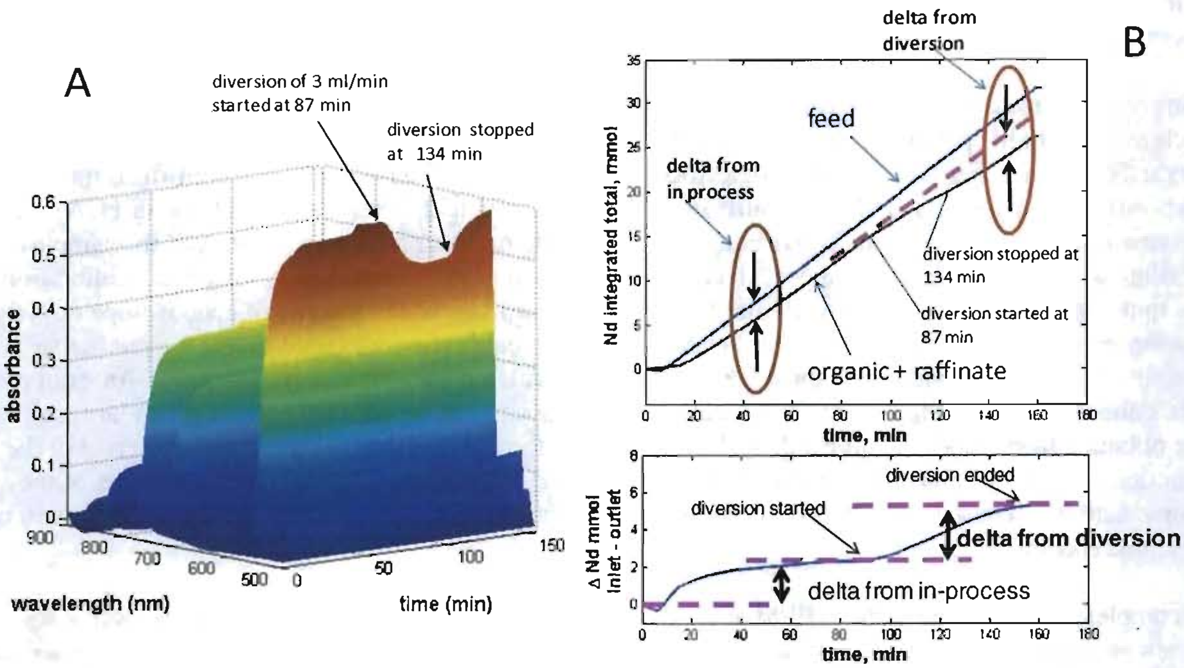

Figure 6. Demonstration of diversion detection and quantification, using on-line spectroscopic PM equipment. Diversion experiment contained PUREX simulant, $Nd(NO_3)_3$, HNO_3, $NaNO_3$, TBP/dodecane, with instrumented 2-cm centrifugal contactors extraction system.

Additional research is necessary to investigate the effectiveness of optical spectroscopic methods for PM of additional nuclides of interest. While the optical sensors and techniques have been successfully tested on spent fuel solutions previously, these solutions have been limited to due cost. The development of this method would benefit from being tested on a flowing spent fuel separation process. This has not been possible previously due to lack of access and/or cost of creating/accessing an operating reprocessing facility. Additionally, these methods could be, but have not yet been tested on additional nuclear processes other than reprocessing. These activities are recommended to further develop the technique for safeguards.

Uncertainty Associated with the MIP Monitor Technique

The MIP Monitor uses the inherent gamma ray signal from spent fuel processing streams to monitor process conditions. It consists of collecting gamma-ray spectra and using multivariate algorithms to analyze the spectra for pattern changes that would indicate a change in process conditions. It has the potential to uniquely identify and quantify the process change (e.g. acid concentration) based on the way the spectral pattern changes as a result of the process change. Because it uses highly penetrating (high energy) gamma rays, the collection can be outside of the tank or pipe and the analysis can by automatic providing near real-time feedback on the process. The NGSI PM project has supported application of the MIPM to international safeguards. In previous years, the PM project supported the demonstration of the MIPM on actual spent fuel samples prepared in the radiological hot cells at PNNL. These samples are valuable and have been used extensively in a wide array of safeguards related technology development. During FY11, the MIPM focused its NGSI PM effort on understanding the fundamental uncertainties associated with the multivariate analysis of gamma spectra. By understanding the uncertainty associated with the techniques employed by the MIP Monitor, one can estimate the limits of the method's performance when deployed as a PM. These estimates can then be incorporated into systems models and uncertainty analysis projects being developed by other participants within the NGSI Process Monitoring group. Understanding the uncertainties associated with a technology allows for a comparison to the current techniques used by the IAEA.

A simple experiment was executed in FY11 to test the uncertainty. Three isotopic gamma standards, which are present in spent fuel, were mixed in various respective concentrations and their spectra collected using high purity germanium detectors. The activities of the isotopes were confirmed through traditional gamma analysis, and then multivariate analysis (Principal Component Analysis, or PCA) was performed on the set of spectra. The results showed that the multivariate analysis grouped the samples according to the isotope being changed. In addition, a portion of the sample set was used as a calibration for a multivariate prediction model (Partial Least Squares, or PLS) of the activity of each isotope directly from the spectra. The portion of the data set not used in the calibration was used to test the predictive capability of the model and the isotopic activities were calculated for each reserved spectra. An example of the calibration and prediction of Co-57 using gamma spectra can be seen in Figure 7. Similar results were obtained from Am-241 and Eu-254. It is interesting to note that the root mean squared error of the predictions is actually less than the uncertainties associated with the confirmatory measurements of the isotopic activity. These results give us preliminary insight into how effective multivariate analysis can be on gamma spectra.

For complete development of the MIPM as a safeguards tool, several development areas are necessary. The above research was performed using a high purity germanium detector to represent the ideal case. In practice, it is assumed that the MIPM approach will use a more robust detector, such as lanthanum bromide, which has poorer resolution. The uncertainty analysis needs to be performed on spectra collected by different resolution detectors to assess the limits of alternative detector options. This is planned as future work. Additionally, the MIP Monitor would benefit greatly from larger collection of gamma-ray spectral data from spent fuel processing. Multivariate analysis requires large data sets in order to optimize the approach. Ideally this data would be collected from a full or pilot scale reprocessing facility; however, given limited reprocessing facilities in the world, the project has yet to obtain this data. Until such data is obtained, multivariate algorithm and approach development continues using computationally simulated spent fuel and gamma spectra. This research includes accounting for the variations in gamma signal not related to the process changes, such as irradiation or cooling time of the fuel. As the approaches are developed, however, they will eventually require field data for confirmation and optimization of their effectiveness.

Potentiometric Sensors

In FY08, NA-24 commissioned a multi-laboratory working group to assess the state of safeguards approaches currently being implemented at reprocessing facilities. The technical report [ref] dedicated one chapter to technical deficiencies that currently exist in this area. One specific technical need identified was the need to determine nitric acid concentration in process solutions, because nitric acid concentration is key parameter that if modified can lead to Pu diversion (Diversion scenario 1 for example). We have started to evaluate potentiometric sensors for meeting this challenge. We tested the efficacy of nitrate-selective electrodes and their ability to discriminate interfering anions that will coexist in reprocessing solutions. Due to limited financial resources, the full scope was not completed, so evaluation of pH sensors that are suitable for the reprocessing environment remains as future work.

Fig. 8 demonstrates the selective affinity that electrodes can have to ions of interest, such as the nitrate ion (NO_3^-), which enables the detectors to monitor select ion populations in process streams providing information useful to PM.

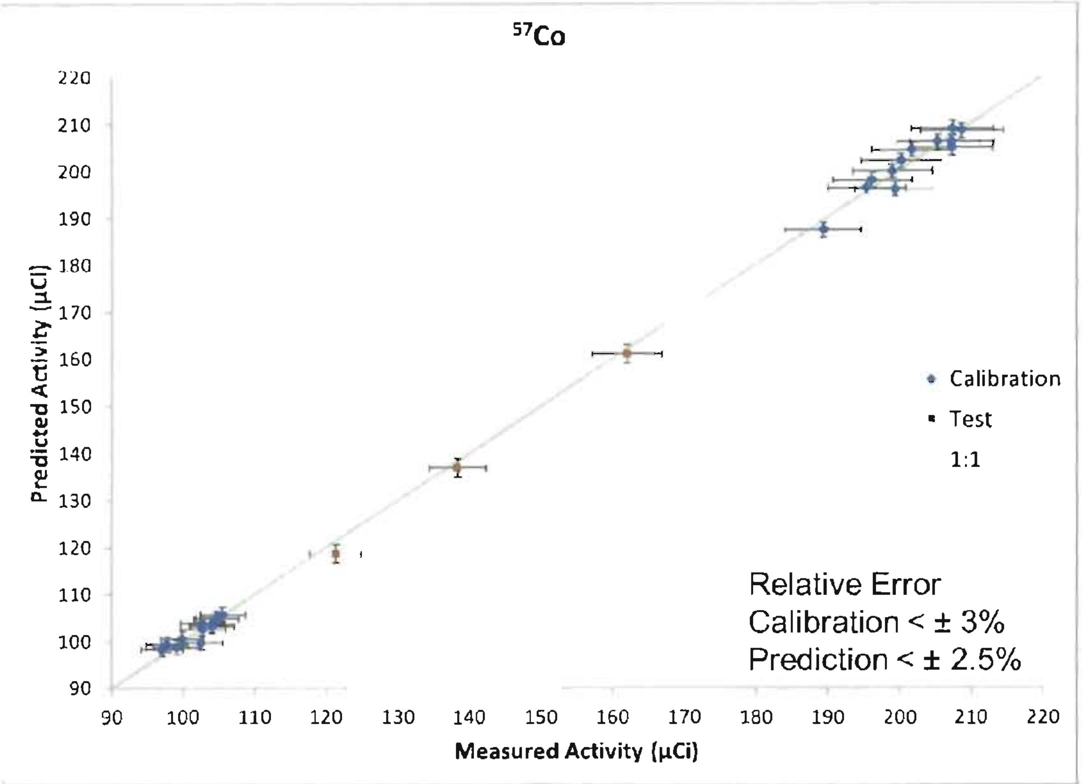

Figure 7. Multivariate predictions of ^{57}Co activity given calibration and test samples that include a variety of combinations of ^{57}Co, ^{154}Eu, and ^{241}Am compared to the measured activity using traditional gamma analysis.

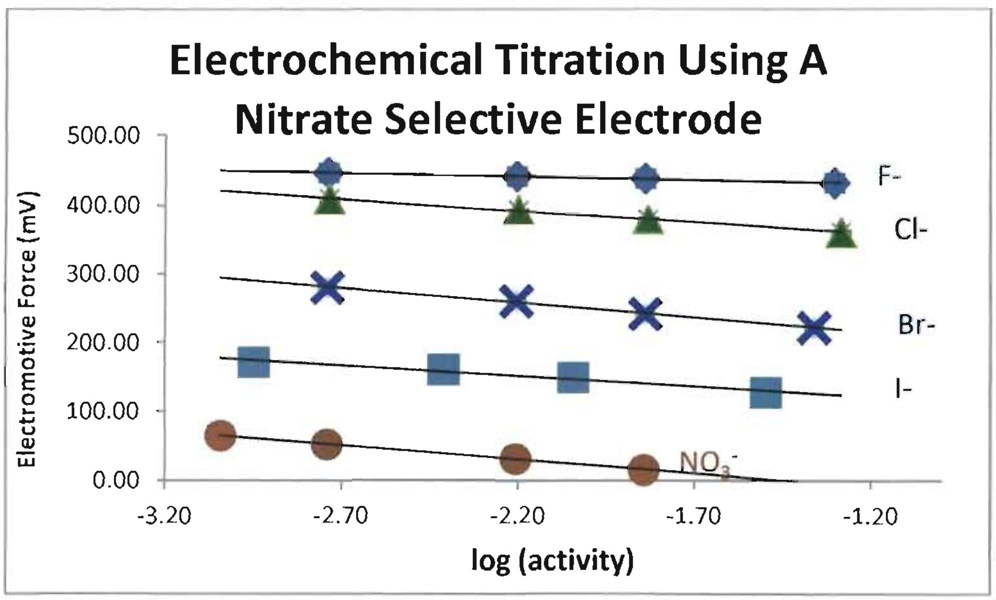

Figure 8. Selective affinity that electrodes can have to ions of interest.

Simplified Example of Combined NMA and PM Detection

One approach developed to determine the added value for PM in addition to NMA alone is shown below by Equation 1, where the overall probability of failing to detect material loss for a specific diversion is the combination of failing to detect loss with both NMA and PM. This approach is an example of putting NMA and PM on "equal footing" and this particular approach requires independence of NMA and PM measurements.

(1) $P(failure_{combined}) = P(failure_{NMA})P(failure_{PM})$

The NGSI PM project has focused on developing algorithms for detecting loss with PM, and then combining with existing approaches for detecting loss with NMA. To illustrate the use of Equation 1, Diversion Scenario (2) will be evaluated for combined NMA and PM, as shown by Figure 9. This scenario is described in the earlier section "Simulated Operating Plant Data with Diversions".

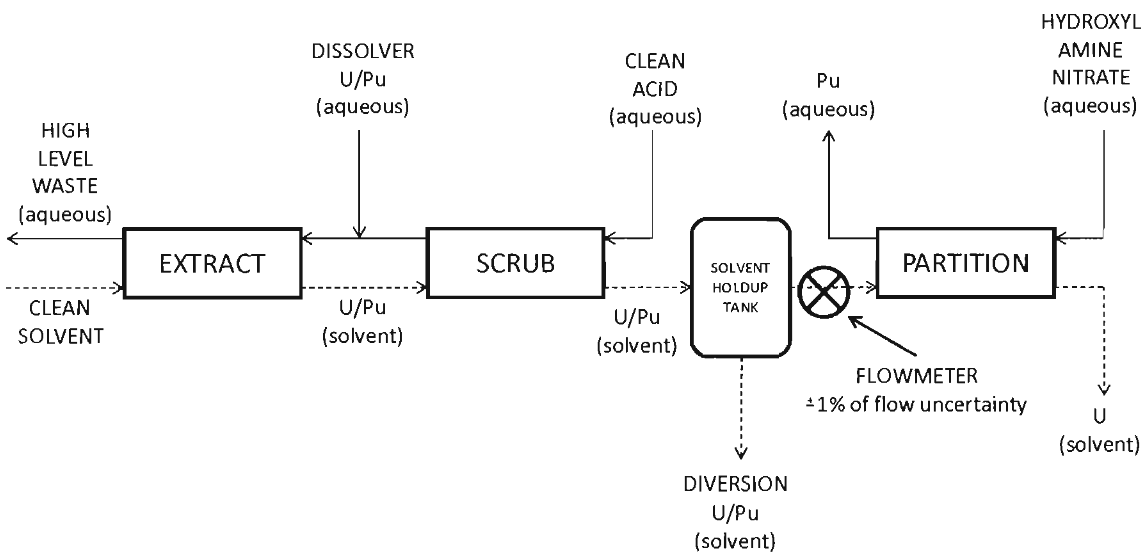

Figure 9. Diversion Scenario (2)

Additional information to estimate detection probability can be found in Suzuki and DeMuth, 2011. Figure 10 is taken from Suzuki and DeMuth, and shows an estimated NMA sigma-MUF for a "model" 800 MTHM/yr PUREX facility operated 200-days/yr[1]. This sigma-MUF is based on simulated plant data. As shown in Figure 10,

[1] Suzuki, M. and DeMuth, S., Proliferation Risk Assessment for Large Reprocessing Facilities with Simulation and Modeling, Proceedings of Global 2011, Paper 468544, Chiba, Japan, December 2011.

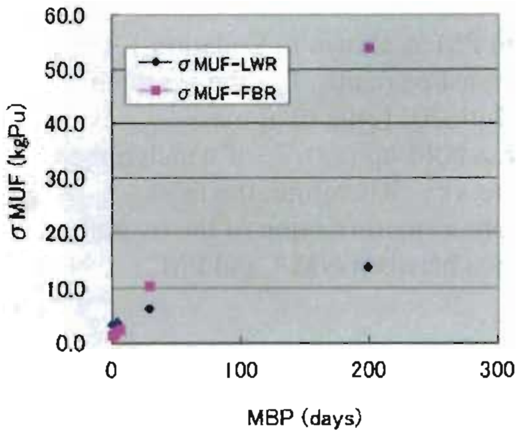

Figure 10. sigma-MUF for 800 MTHM/yr PUREX facility

sigma-MUF at 30-days is approximately 8 kg-Pu For this NMA example we calculate the probability of detecting an 8 kg-Pu loss given that sigma-MUF equals 8 kg-Pu. Following is the NMA probability of detection at 30-days for an 800 MTHM/yr PUREX facility described by Suzuki and DeMuth. Thirty days inventory was selected for this example as it is the IAEA requirement for interim inventory. The probability of detection for a sigma-MUF of 8 kg-Pu is shown to be 0.258 with a false alarm probability (FAP) of 0.05, which is quite low compared to the IAEA goal for detection of 0.95 with a FAP of 0.05. This low probability of detection is consistent with the claim made in the Executive Summary of this report "For large throughput nuclear facilities such as commercial spent fuel reprocessing plants, it is difficult to satisfy the IAEA's goal for detection probability using NMA alone."

$$P_{detection,NMA}(x = 8\ kgPu, \sigma = 8\ kgPu, FAP = 0.05) = 0.258$$

For Diversion Scenario (2) the PM flow meter measurement uncertainty is $\pm$ 1% of flow at one standard deviation, which for this example is assumed to be equal to a single measurement of total flow at 30-days. While this is a simplification of perhaps one or more measurements per day for 30-days, the approximation is valid if the systematic error (as opposed to random) is a significant portion of the $\pm$ 1%. This is perhaps an acceptable assumption given that flow meter calibration would likely be done at longer intervals than 30-days given the typically difficult access to process cells. Assuming $\pm$1% error standard deviation on the total flow into the solvent tank, an 800 MTHM/yr PUREX facility operating 200 days/yr with $\pm$1% Pu in the spent fuel will process 1200 kg-Pu/30-days, which translates to a standard deviation in measurement error for PM of 12 kg-Pu. For this PM example we want to calculate the probability of detecting 8 kg-Pu loss given that sigma-PM equals 12 kg-Pu. The following is the PM probability of detection at 30-days, based on a FAP of 0.05, for an 800 MTHM PUREX facility as described by Suzuki and DeMuth for Diversion Scenario (2). It is shown to be less than that of NMA as would be expected, because the measurement standard deviation for NMA is 8 kg-Pu and for PM is 12 kg-Pu.

$$P_{detection,PM}(x = 8\ kgPu, \sigma = 12\ kgPu, FAP = 0.05) = 0.165$$

To combine the probabilities of detection for NMA and PM as shown in Equation 1, the case of independence of NMA and PM measurements must be made. For the example of Diversion Scenario (2), as reported in Suzuki and DeMuth 2011, the total mass contribution to sigma-MUF at 30-days for the in-process hold-up is 16%, of which only a fraction would exist in the solvent holdup tank of Figure xxx. Therefore, the mass measurements associated with Diversion Scenario (2) are a small fraction of the overall NMA sigma-MUF, leading to approximate independence between NMA and PM measurements.

Using a false alarm probability of 0.05 for NMA and PM each, yields an overall combined probability of 0.38 shown by Equation 2.

(2) $P(success_{combined}) = 1 - (1 - 0.258)(1 - 0.165) = 0.380$

For this case the combined NMA and PM overall false alarm probability is 0.0975, which is almost twice as high as the IAEA requirement for NMA testing alone.

$$P_{combined}(false\ alarm) = 1 - [1 - P_{NMA}(false\ alarm)][1 - P_{PM}(false\ alarm)]$$

However, another approach is to use a combined NMA and PM overall false alarm probability equal to the IAEA requirement for NMA testing alone, which is 0.05. This yields an individual NMA and PM false alarm probability of 0.0253. The individual detection probabilities for NMA and PM based on an individual false alarm probability of 0.0253 each, are then reduced from 0.258 to 0.169 and 0.165 to 0.102 respectively.

$$P_{detection,NMA}(x = 8\ kgPu, \sigma = 8\ kgPu, FAP = 0.0253) = 0.169$$
$$P_{detection,PM}(x = 8\ kgPu, \sigma = 12\ kgPu, FAP = 0.0253) = 0.102$$

Using the revised detection probabilities for NMA and PM based on a false alarm probability of 0.0253, the overall combined NMA and PM detection probability then becomes 0.254 as shown by Equation 3.

(3) $P(success_{combined}) = 1 - (1 - 0.169)(1 - 0.102) = 0.254$

Notice that when assuming the overall false alarm probability per test of 0.05, Equation 3 implies there is actually a disadvantage to combining PM with NMA due to the reduced individual false alarm probability of 0.0253. If the flow meter measurement uncertainty is reduced to $\pm$0.5% or even $\pm$0.25%, combining PM with NMA significantly improves the detection probability even with the reduced individual false alarm probability of 0.0253. For a flow meter uncertainty of $\pm$0.5% the combined NMA and PM probability of detection is 0.39, and for $\pm$0.25% it is 0.80, as shown in Table 2.

Table 2. Probability of detection for diversion scenario shown in Figure 9.

Flow meter uncertainty ($\pm$%)	P(NMA alone)	P(NMA + PM)
1.00	0.258	0.254
0.50	0.258	0.39
0.25	0.258	0.80

Combined NMA and PM Detection for Safeguards by Design

The approach demonstrated with the "Simplified Example" for estimating the probability of detection using combined NMA and PM can be used to evaluate early safeguards options during a plant design. It can be seen from Equation (3) that improving the detection for either NMA or PM will improve the overall detection probability for this one particular scenario, of a diversion from the solvent tank All approaches that combine NMA with PM will need to pay careful attention to the overall false alarm probability, which in this example was simple because only two independent tests were performed.

The low probability of detection for NMA is due primarily to the large throughput for an 800 MTHM/yr PUREX facility. The NMA probability of detection can be improved by reducing the inventory time, which was assumed to be 30-days for this example based on the maximum allowable for an IAEA interim inventory. This is the basis for current efforts at near-real-time accountancy (NRTA) at existing facilities such as Rokkasho; however, there is a cost associated with each inventory. *Additionally, reducing the interim inventory time, as in NRTA, reduces the ability to detect protracted diversion.*

The low probability of detection for PM is due to the large throughput for an 800 MTHM/yr PUREX facility and the relatively high flow meter uncertainty of 1%. The PM probability of detection can be improved in ways that are analogous to improved NMA, by reducing the inventory time; however, it can also be improved by the use of flow instrumentation with reduced measurement uncertainty. Improved instrumentation has an additional cost as does increasing the number of inventory measurements. It is the ability to quantify the contribution from PM to the overall detection probability that provides an opportunity for cost-benefit trade studies comparing new instrumentation against additional inventories. *The approach developed here provides a new opportunity for Safeguards by Design.*

Actual Operating Plant Data

We briefly describe currently-available PM data from 4 real facilities which are or were operating. The PM project has considered both currently-available PM data (such as in-tank volumes and masses) and potentially-available PM data such as material flows (which can be difficult to measure in high-radiation environments) and data from the MIPM. The last U.S. facility closed in 1996 and data from foreign facilities is often proprietary and closely held; however, progress has been made as described next.

Barnwell

Data was collected at the Barnwell Nuclear Fuels Plant (BNFP) in the early 1980s[34]. The BNFP was a commercial-scale facility with a computerized process data collection system used as a safeguards testbed. BNFP data provides a rare feature in that it *includes diversions of known volumes at know locations and times.* To our knowledge, no other nonproprietary data sets with these attributes exist. However, recoverable electronic versions of this data are not available; it now exists only as graphs in reference[36] (see Figure B.1).

The PM team was charged with determining if a digital form of the data could be re-created, and, if so, how useful would it would be. A sub-goal was to discover any potential diversion indicators and incorporate them into the IKE (Integrated Knowledge Engine) Diversion Detection Model at LANL.

The team selected 1 of 5 mini runs (approximately 1 week of data per mini run) documented in and digitized the graphs[34]. This process started by creating jpeg screen captures of each graph from a pdf version[36]. There are frequently two traces per graph, they often intersect at multiple locations, and one of the traces is a dashed line (i.e. fainter than the other trace). Given these conditions, the digitization software could not automatically extract the traces without significant efforts to correct mistakes. Instead, the user manually placed points on the trace and the software recorded the X-Y coordinates of each point. The digitization process was labor intensive. Following digitization and interpolation of the data, a complete digital dataset was created. An important advantage of digitizing the graphs is we can now perform automated analyses on the data rather than just relying on visual inspection. Once the complete data set for the run was assembled the team conducted analyses to determine how consistent the data were and looked for indications of the documented diversions.

Analyses indicated a number of potential inconsistencies such as: difficulty locating the diversion events on the time axis of the graphs, time discrepancies between batch shipment and receipt in tanks (both in times and volumes), and an apparent lack of process equilibrium in the column portion of the process. The only available expert on this data, Mike Ehinger of ORNL was involved in these safeguards tests at BNFP. After examining the data and process diagrams, Mr. Ehinger was unable to explain the inconsistencies. Unfortunately, the team cannot go back to the original level, density, temperature data collected by the PM system, nor can it recreate the analyses presented in the report in an effort to resolve the questions.

In light of these facts, the conclusion is that the digitized portion of the Barnwell data is of less use to the safeguards community than first hoped. Also, inspection of the graphs from the other four mini runs does not indicate they would lead to significantly different results. Nevertheless, we believe that this effort at least provided a data source for increased understanding of some aspects of solution monitoring data, such as measurement results during non-transfer periods in some tanks. However, the effects of the known diversions are difficult to analyze in meaningful ways because of the inconsistencies described above. With this finding we believe that simulation with realistic effects learned from actual facility data is a good next step.

Idaho Chemical processing Plant (ICPP)

As described in the INL (Idaho National Laboratory) FY09 report (which includes limited updates through FY10), the Idaho Chemical Processing Plant (ICPP) data has been largely recovered and finalized in both an Official Use Only and open format for release to national laboratories and universities. Additionally, INL provided continued support for data users, clarifying questions about the data as needed. The INL has also begun linking metadata with specific tanks for easier analysis. This metadata linkage will decrease the learning curve required for use of the data, and is intended to allow for faster validation of diversion-detection events. Artifacts in the data which occur when a sensor is saturated are being removed through an analysis of variance. These highly variant signals are being tested against physical reality (e.g. a tank cannot hold more than 100%). This data improvement will mark, but not delete, the highly variant areas, meaning that they can also provide validation advantages. In addition, INEL began an effort with SRS to recover data from their separations canyon and with ORNL to recover the Barnwell data described above. To date, only the ICPP data has been recovered.

Savannah River Site (SRS) Uranium Tank Data

Hourly tank solution level (*L*) and density (*D*) (but not temperature *T*) data from 9 Uranium storage tanks is available from May 1, 2005 through April 30, 2007, which is 17,516 hourly snapshots of each tank. References [7,17] report initial evaluation of this data.

Tokai Reprocessing Plant Plutonium Storage Tank Data

Tank solution *L*, *D*, and *T* data every 5 minutes from each of 7 Pu storage tanks, 1 process input tank and 1 process output tank is available on 348 consecutive days from Aug. 2006 into July 2007. The process output tank ships to the storage tanks and the 7 storage tanks ship among themselves. Of the seven storage tanks, four have *L*, *D*, and *T* readings, but only the first 3 receive from the output tank, and these have *L* readings only. Mass tracking is still feasible using L readings, but volume tracking is most effective if a D reading is available; for the 3 product tanks without inline D, one can assume fairly constant D or estimate D by sampling. References [7,17] report initial evaluation of this data.

Real Data Summary

In summary, real data from Barnwell, INL (ICPP), SRS, and TRP have been made available under several programs and sponsors at the unrestricted or OUO level. These real data sets are modest size and have not yet been fully analyzed. The Barnwell data seemed to be very promising because of the known diversions in the mini-runs, but data inconsistencies introduced either in the data recovery process described, or in the original data, or both, have made data analysis efforts to date problematic. Nevertheless, some aspects of the Barnwell data are useful for understanding tank data during non-diversions.

CONCLUSIONS

*The **Concepts and Approaches** supported by the PM project tasks reported here are consistent with the NGSI goal of "Implementation of safeguards at declared facilities can be made more efficient and effective by incorporating advances in automation, measurement, and information technology" and "promoting Safeguards by Design as an international standard.", and future goals for the IAEA identified at the "Consultancy Meeting on Proliferation Resistance Aspects of Process Management and Process Monitoring/Operating Data" held in Vienna, 28-30 Sept 2011. These IAEA goals included a "proof-of-principle study on a well-known facility, demonstrating impact on efficiency and effectiveness" and a "proof-of-principle study on an advanced (future) facility, demonstrating that safeguards goals could be met using extended PM."*

These Concepts and Approaches can best be described as methodologies to determine the added value for PM in addition to NMA alone, and its use for cost/benefit trade studies in support of a Safeguards by Design (SBD) approach to international safeguards. While multiple methodologies were identified, that of "Equal Footing" was most fully demonstrated through its application to a PUREX reprocessing facility, and more specifically diversion of Pu from a solvent tank between the scrub and partition process operations. It was demonstrated that the diversion detection probability of 8 kg-Pu within 30-days can be measured and improved significantly with the addition of PM flow meter instrumentation, and as expected, the degree of detection improvement is dependent on the flow meter measurement uncertainty. The Equal Footing methodology developed here was used to demonstrate that the contribution of PM to NMA for safeguards can be measured and used for early design of safeguards systems (i.e. SBD) for select diversions. While the methodologies for measuring the contribution of PM reported here represent an improvement in the state-of-the-art, and not about a complete tool for assessing all potential risks, it is important to note that safeguards is concerned with "defense-in-depth" implying small contributions can be significant.

Consistent with NGSI program goals and IAEA objectives the following tasks were undertaken in support of the PM project reported here: (1) develop a partial reprocessing plant model to simulate operating data representative of specific diversions, (2) identify diversion paths that can be used to demonstrate combined PM and NMA detection algorithms, (3) design and prototype PM detection algorithms using currently-available or potentially-available PM data, (4) develop advanced instrumentation for PM such as the Raman/UV-vis-NIR spectroscopic monitor and the Multi-Isotope Process (MIP) monitor with related authentication assessment, and (5) review actual facility data that included deliberate diversions and recover modest amounts of historic real facility data. Regarding (4), an initial effort was made regarding data authentication, as reported in reference [9].

RECOMMENDATIONS FOR FUTURE WORK

The NGSI PM project reported here was intended to further NGSI and IAEA objectives. Ancillary to these objectives, the NGSI PM project team is hopeful that their efforts will open new international dialogue on advanced and/or enhanced PM applications for the 21st century. ***This ancillary objective could be accomplished through efforts such as (1) benchmarking methodologies with foreign collaborators and partnering with the IAEA to better understand their need and (2) identifying and promoting new PM instrument designs that are easily authenticated, unattended, and enable remote signal transmission***. Additionally, without industry participation and feedback regarding proprietary and intellectual property concerns, implementation of advanced PM will lack the needed support. Examples exist where industry supports the use of advanced safeguards technology when it can be leveraged with their needs, such as advanced process control.

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APPENDICES

Appendix A. Systems-Centric Approach to combine PM with NMA data

Appendix A provides more detail regarding the systems-centric approach to combine PM with NMA data. As shown in Fig. A.1, DEI algorithms for PM utilize knowledge regarding plant operations and characteristics of deployed sensors. Plant operations may be modeled, for example, by stochastic automata (a collection of possible system states and transitions probabilities between the states). Characteristics of deployed sensors, may be modeled, for example, by probabilities of their misdetection, correct detection, and misclassification. With this knowledge, DEI algorithms are able to logically infer occurrences of anomalies without having to directly observe all events that constitute the anomalies. For example, consider a monitored facility consisting of two UO, UO_1 and UO_2, and suppose that high temperature in UO_2 is one of the events that constitute an anomaly of interest. Moreover, suppose the facility operates in a way such that high temperature in UO_2 occurs with a known probability if the concentration of a particular material is too high in UO_1. Consequently, if a sensor with known characteristics is deployed in UO_1 to measure concentration of this particular material, the probability of high temperature in UO_2 can then be inferred without directly observing the temperature in UO_2.
In particular, the developed DEI algorithms for PM update the estimated number of anomaly occurrences as sensor observations become sequentially available. The operations of a DEI algorithm can be written as

$$M_{n+1} = D(M_n, o_{n+1}, A, S)$$
$$(c_n, i_n) = h(M_n)$$

where A and S represent knowledge regarding plant operations and sensor characteristics, respectively, o_n is the nth observation, and M_n represents internal information, updated after o_n becomes available, and is used by the DEI algorithm for necessary calculations (such as calculating the estimated number of anomaly occurrences). Moreover, c_n is the estimated number of anomaly occurrences after o_n is reported, and i_n denotes information on the confidence of c_n. An example of i_n may be an estimated variance of c_n. The function D denotes the process of updating the internal information based on A, S, the current internal information, and the most recent reported observation. Similarly, function h denotes the process of calculating c_n and i_n based on the internal information.

An additional design problem for the observation platform is deciding how much data should be collected (i.e., selection of deployed sensors) and how to collect and evaluate collected data (i.e., selection of the DEI algorithm) so that the observation platform satisfies design requirements (e.g., probabilities of detection and false alarm and cost constraints). In the following, the set of deployed sensors is referred to as sensor configuration. The problem of finding optimal observation platforms entails balancing cost and performance. The performance of an observation platform may be quantified, for example, by the estimated variance of the computed number of anomaly occurrences, while its cost may essentially depend on the number of sensors deployed, their quality (i.e., reliability), their impact on operation, their tampering characteristics, and difficulties of installation and transmission, for example. In general, sensor configurations that cost more may often include more sensors and/or sensors with better quality, which typically leads to better anomaly detection performance. Thus, an optimization algorithm may utilize search algorithms to find sensor configurations that minimize a loss index, L, which is a weighted sum of observation platform performance and cost. Given a DEI algorithm and monitored facility (with defined anomalies of interest for detection), the optimization algorithm can be written as

$$S^* = \arg\min_{S \subseteq U} \{L : C_P, C_C \text{ satisfied}\}$$

where S^* is the optimized sensor configuration, U is the set of possible observables, and C_P and C_C are user-defined constraints on performance and costs, respectively, for the observation platform at hand. For example, C_P may indicate the worst acceptable observation platform performance, while C_C may indicate the maximum cost tolerable. Note that, since L is defined as a weighted sum of observation platform performance and cost, the minimization of L indicates a balance between them. Figure 2 illustrates the observation platform optimization algorithm developed for PM. Note that the options for DEI algorithm and available sensor configurations depend on the monitoring task at hand and operational particulars of the monitored facility. The loss index is user-defined and represents the user's priorities regarding cost and performance. The observation platform performance constraints (C_P) are also user-defined.

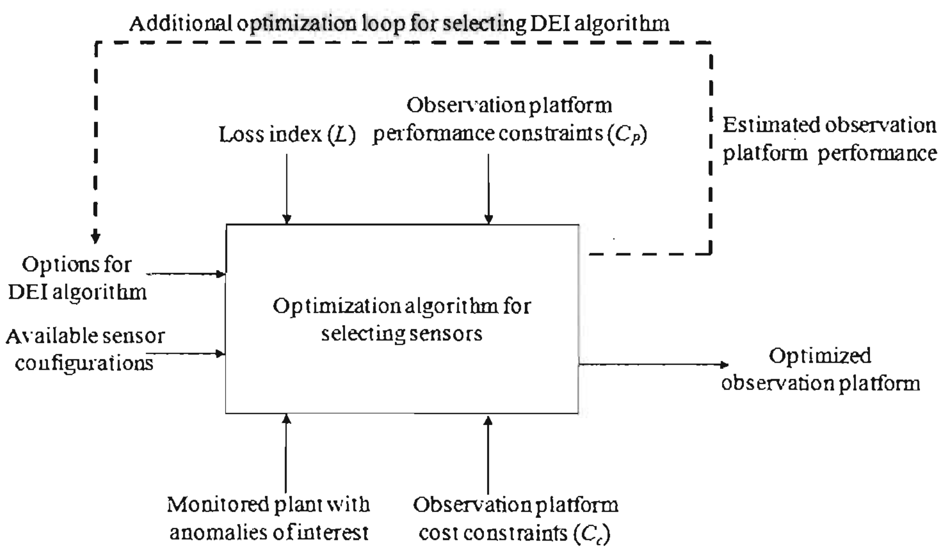

Figure A.1. Observation platform optimization algorithm.

__Appendix B. Barnwell data__

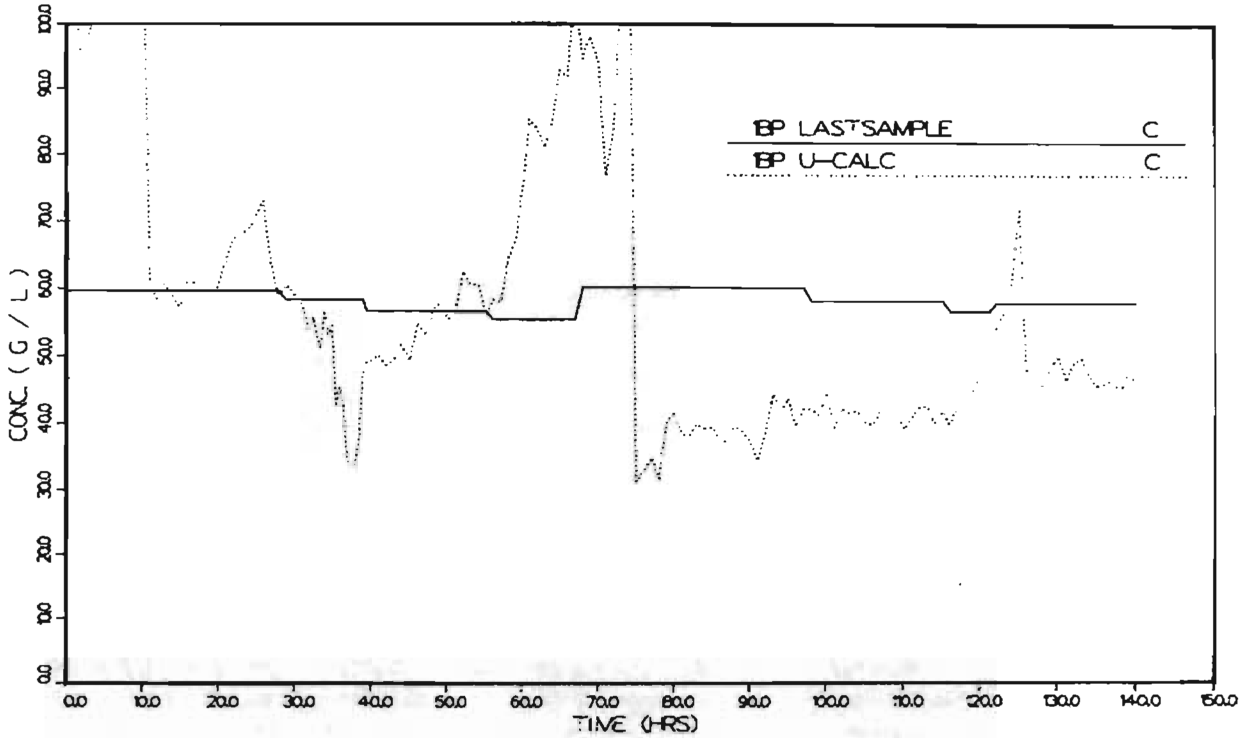

Figure B.1. Example Barnwell Graph[36]. BP denotes a surge tank and "last sample" and U-calc both refer to the particular method used to estimate U concentration.