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Multi-Isotope Process (MIP) Monitor Deployment at H-Canyon

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ABSTRACT: Material Control & Accounting (MC&A) programs at nuclear processing plants deter and detect theft and diversion of nuclear material by both outside and inside adversaries. Current destructive assay approaches to MC&A can be augmented with nondestructive online monitoring programs to reduce the required resources and improve the timeliness of MC&A at reprocessing facilities. The Multi-Isotope Process (MIP) Monitor is one approach to online process monitoring for MC&A and process control. The MIP Monitor measures gamma-emitting radionuclides present in feed, product, and waste streams of a nuclear reprocessing or separation facility. These radionuclides are monitored online by gamma spectrometry and compared, in near-real-time, to spectral patterns representing "normal" process conditions using multivariate pattern recognition software. By targeting multiple gamma-emitting radionuclides, the MIP Monitor approach is compatible with the use of small, portable, medium resolution gamma detectors that may be easily deployed throughout an existing facility.

The distribution of fission and activation products in a reprocessing stream is influenced by several operational variables, including acid concentration, tributyl phosphate (TBP) concentration, temperature, and used fuel characteristics (e.g. reactor type, initial enrichment, burn-up, cooling time). For a consistent industrial process with reproducible conditions, the distribution of each radionuclide between the organic and aqueous phases should be relatively constant for a given type of used fuel. Consequently, the distribution of radionuclides in both product and waste streams at each stage in the facility should be relatively reproducible (within normal industrial variations). Initial simulations and bench-scale experiments have documented the performance and potential of the MIP Monitor; this approach has yet to be applied to a full-scale facility.

Researchers at the Pacific Northwest National Laboratory in conjunction with Savannah River National Laboratory, University of Tennessee-Knoxville and the University of Texas-Austin are working to deploy the MIP Monitor at the H-Canyon Nuclear Separations Facility at the Savannah River Site. The goal of the deployment project is to continuously gather and process the spectral data in the hot and warm sampling aisles at the H-Canyon facility during actual used nuclear fuel separation campaigns. The information gathered could be used to improve safety, material security, efficiency, material and process controls, and overall plant performance to aid both the regulator and the plant operator. In addition, demonstrating the operation of the MIP Monitor in a full-scale facility will help to advance the technological readiness and realize the benefits associated with the technique. This technology is intended to support the Department of Energy's (DOE) Nuclear Energy's Fuel Cycle Research and Development (FCR&D) and Materials Protection, Accounting and Control Technology (MPACT) program. This paper will present an overview of the MIP Monitor deployment project, detail our experimental deployment plan at H-Canyon, and present preliminary data from current deployment efforts.

KEYWORDS: *Reprocessing, Process Monitoring, Nuclear Safeguards, Gamma-ray Spectroscopy, Multivariate Analysis, Non-Destructive Analysis*

I. INTRODUCTION

Researchers at the Pacific Northwest National Laboratory (PNNL), in conjunction with Savannah River National Laboratory (SRNL) and U.S. universities, are working to deploy the Multi-Isotope Process (MIP) monitor at the H-Canyon Nuclear Separations Facility located at the Savannah River Site (SRS). The MIP monitor is a process monitoring and verification tool for monitoring used nuclear fuel reprocessing in aqueous separation facilities, non-destructively and in near-real-time. The goal of the MIP monitor deployment project is to improve material control and accountancy (MC&A), process control, safety, efficiency, and overall plant performance to aid both the plant operator

and the regulator.

Current conventional nuclear safeguards and process control methods rely, in part, on destructive analyses to ensure MC&A of nuclear material at reprocessing facilities. However, destructive analyses are extremely resource intensive, have limited sampling rates, and require a significant time lag from sampling to reporting the final results. In addition, while the accuracy of these destructive measurements is excellent, error associated with these analyses scales directly with the size of the facility [1]. As such, highly precise destructive measurements of special nuclear material alone at large facilities may not be optimal to meet the goals of MC&A programs.

The MIP monitor takes advantage of the normal distribution of radionuclides that are mixed during fuel recycling and separations. By combining information-rich gamma-ray spectroscopy with pattern recognition software, the MIP monitor can identify normal and off-normal conditions in process streams and even quantify certain plant and fuel conditions. Because it targets gamma-emitting isotopes, the MIP monitor approach is compatible with several separation process methods. Laboratory scale experiments proved successful using small, portable, medium resolution gamma detectors. These same detectors can be deployed throughout the nuclear separations facility at multiple measurement locations. Previous laboratory-scale research has included applying multivariate statistical techniques to simulated and empirical gamma spectra representative of spent fuel separations under various process conditions [2], demonstrating the validity of the MIP monitor. Current efforts include deploying the MIP monitor at H-Canyon over the course of its used nuclear fuel processing and plutonium blending campaigns. A review of the technology, previous research, and current efforts will be presented here.

II. Overview of MIP Monitor

Modern industrial reprocessing techniques are typically based on solvent extraction between opposing organic and aqueous phases. In these bi-phase systems, product (actinide) and contaminant (fission and activation products) elements are preferentially driven (thermodynamically) to opposite phases, with small amounts of each remaining in the other phase [3]. The distribution of each element, between the organic and aqueous phases, is determined by major process variables such as acid concentration, organic ligand concentration, reduction potential, and temperature. Hence, for a consistent industrial separation process, the distribution of each element between the organic and aqueous phases is expected to be relatively predictable.

The MIP monitor is designed to track changes in the distribution of gamma-emitting radioisotopes indicating unexpected changes in the process conditions . In-process surveillance by the MIP monitor is accomplished by coupling the gamma spectra recorded from constituent streams with multivariate analysis techniques such as principal component analysis (PCA) or partial least squares regression (PLS). The MIP Monitor technique evaluates time-varying spectral patterns of the gamma-emitting contaminants in near-real-time to detect relevant signs of potential changes to the process chemistry or to classify characteristics of the fuel or the separations process.

a. Gamma Detection

The MIP Monitor analyzes process streams online in near-real-time, which requires gamma detectors placed next to a grab sample or directly connected to a processing pipe in key locations throughout the facility. Given the large amount of high-energy gamma rays, a significant percentage of the gamma rays should penetrate pipe walls to provide detector signal while removing the necessity of placing a sensor inline. Depending on the deployment locations at the facility, this penetrating gamma flux may require gamma detectors to be placed behind dose mitigating biological shields or other attenuating materials at the reprocessing facility, requiring that the detectors be robust, easily collimated, and require low maintenance. Given these operating conditions, the use of medium resolution, room temperature detectors has been explored for use with the MIP Monitor analysis techniques in addition to proof-of-concept simulations and experiments performed using high-resolution detectors. Experimental, bench-scale results to date suggest that medium-resolution detectors are sufficient for fuel characterization and anomaly detection [4].

b. PCA

PCA is an unsupervised technique that linearly transforms correlated data into an uncorrelated PC-space. This process is illustrated in Figure 1 with notional data. Here, data are collected for three correlated variables. The first PC is found in the direction of the greatest variance of the original data. The second PC is found such that it (1) is orthogonal to the first PC and (2) lies in the direction of the greatest remaining variance. The third PC is then orthogonal to both the first and second PCs and lies in the direction of the greatest remaining variance. For the example data, two PCs are sufficient to describe the majority of variance in the data; the third PC likely captures noise in the system. In this way, PCA can be considered a dimension reduction technique: data with large dimensionality can be reduced to fewer PCs without significant loss of information.

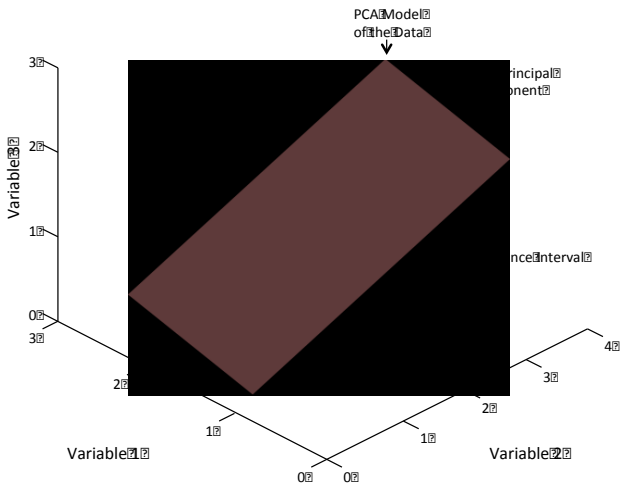

Figure 1: Notional illustration of principal component analysis

In the MIP Monitor approach, gamma spectra from simulations and experiments are analyzed using PCA to identify trends in the spectral patterns and clusters of normal data in the PC-space. These clusters may be useful to classify the spectra according to the physical differences such as acid concentration or fuel composition over time. A successful application of PCA may be able to cluster the samples into distinct groups in PC-space based on these physical variations. When applying PCA to gamma spectra, each channel in a spectrum can be treated as a variable. PCA reduces the large number of channels (typically 1024 for a medium-resolution detector) into a more manageable number of PCs. PCA is ideally suited for compressing correlated or redundant data while retaining the essential information found in the data. This compression step not only simplifies the original data but it has the benefits of signal averaging and capturing information that depends on how the variables (e.g., energy channel intensities) change with respect to one another. A full explanation of PCA and how it has been used as part of the MIP monitor can be found in Reference [5].

c. PLS

Partial least squares regression (PLS) is a supervised prediction technique similar to PCA: PLS applies a linear transformation to a set of correlated input data to identify the underlying latent variables (LVs). In PCA, the PC loadings (the linear transformation) are chosen to maximize the variance explained by subsequent PCs; in PLS, the LV loadings are chosen to maximize the variance explained in the input data as well as the covariance between input and output data in subsequent LVs. Because LVs are selected to preferentially predict a specific output, PLS is a supervised analysis method.

PLS is a multivariate calibration method that can make quantitative predictions of the physical attributes mentioned in the previous section (e.g. burnup, acid concentration, cooling time, etc.) using the measured spectrum of a sample. The PLS method identifies LVs with maximum correlation to a desired output. A more detailed review of PLS and its use in the MIP monitor analysis can be found in Reference [5].

A specific formulation of PLS, PLS-discriminant analysis (PLS-DA) can be used for classification purposes. In this case, the desired output is the observation class (e.g., fuel type or sampler box). PLS-DA may outperform standard PCA for classification because the LVs are chosen to maximize their predictive power. For PLS-DA, the model output is a set of binary variables indicating whether or not an observation, *i*, belongs to a specific class, *j*:

$$y_{ij} = \begin{cases} 1 & \text{if obs } i \in \text{class j} \\ 0 & \text{otherwise} \end{cases}$$

This output encoding encourages the PLS-DA to identify between-group variance that is useful to discriminate between classes.

III. Proposed Research and Preliminary Results

The MIP monitor was deployed at H-Canyon to capture the changing radiological conditions within a nuclear separations plant in near real time while the facility conducts normal processing, cycling, and transfer operations. The primary objective of this report was to capture and analyze the blending of two different mixed oxide (MOX) fuels as two separate tanks containing MOX dissolved fuel of different purities were combined and blended into a third tank. Tanks Alpha and Charlie were the source tanks and Delta was the combination tank. Table 1 compares the plutonium isotopics of each tank.

MOX Metadata		
	Alpha	Charlie
Pu-238	0.01%	0.02%
Pu-239	93.94%	93.09%
Pu-240	5.91%	6.55%
Pu-241	0.10%	0.13%
Pu-242	0.04%	0.21%
[Pu]	4.42 g/L	3.00 g/L
[U]	0.00467 g/L	0.00337 g/L
Density	1.321 g/mL	1.244 g/mL
SRNL-L4600-2015-00021		

Table 1 - Chemical analyses of MOX solutions in Alpha and Charlie

The deployment locations were metal sampling boxes inside the hot sampling aisle at the separation facility that provided direct access to the dissolution and mixing tanks located deep inside the lower levels of H-Canyon. Each sampling box is roughly $1m^3$ in dimension with a sidewall thickness of approximately 25 mm. Attached to each of these boxes was an air pneumatic system that allows for the sample transfer of material from the large storage tanks located elsewhere in the facility into the hot sampling aisle. The tank's solution is circulated through the sampling box during the cycling of the tanks providing a constant stream of solution into the sampling box. Inside each of these boxes was a 10-mL

collection vial that was used by facility personnel as a means of capturing grab samples of the tank's contents for chemical and radiological analysis. The project deployment team used the same grab vials as the primary radiological source for gathering gamma-ray spectra from the material in the tanks. Given appropriate training at the H-Canyon and SRNL facilities, deployment team members could be positioned a few feet away from the sampling box while it was opened and cycling eliminating the need for collimation and expensive stand-off facility modifications to the sampling boxes for this effort.

Two tanks containing dissolved plutonium solutions of different isotopics (Table 1) were selected for blending in the facility. Solid state scintillation detectors were placed on a deployment rig that can be positioned within 25 cm of the grab vial (FIGURE 2). The data was gathered to resolve and identify subtle changes with the spectra of the two different MOX solutions.

Figure 2 - Detectors placed within 25cm of source term in H-Canyon.

The blending plan at H-Canyon was to transfer 25% of solution in Charlie into Delta, and upon completion of that transfer, 100 % of Alpha solution would be transferred into Delta at a constant rate.

Both Alpha and Charlie have been characterized using NaI (Tl) and $LaBr_3$(Ce) detectors. Two different NaI detectors have been used to gather preliminary data including a 7.5cm x 2.5cm and a 2.5cm x 2.5cm detector. A 5cm x 5cm $LaBr_3$ detector was also used for this effort. The deployment of multiple detectors was designed to gather enough data at both high and low gamma-ray energies to aide in detector optimization and process signature understanding. Although the plutonium tanks should not have high energy gammas associated with it, other tanks within the facility certainly will. The data were collected using Maestro software operated in List Mode. This mode puts a time stamp on each count collected by the software allowing for the near-real-time data analysis of Alpha and Charlie at the exact moment they are blended into the Delta tank.

Preliminary gamma spectroscopy data using a 1"x1" NaI detector is shown in **Error! Reference source not found.**.

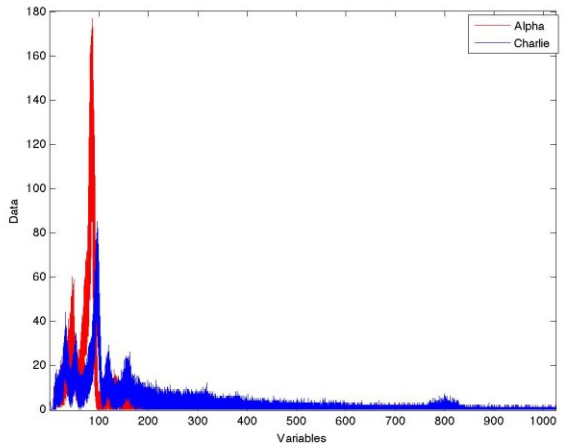

Figure 3 - 5-second gamma spectra measured at Alpha and Charlie locations (1x1 detector)

Figure 4 shows the results of a PCA model calibrated with 5-second spectra from both Alpha and Charlie tanks. In this case, a single PC is sufficient to separate the two data sets. A plot of the first LV is very similar. In this case, discrimination between classes is straightforward: a threshold can be applied to the first PC or LV score and the class assigned accordingly. As the Delta sampler is filled with a mixture of Alpha and Charlie, it is expected to lie between the two clusters in the PC- or LV-space. The exact PC or LV value may be sufficient to estimate the proportions of Alpha and Charlie material in the mixture.

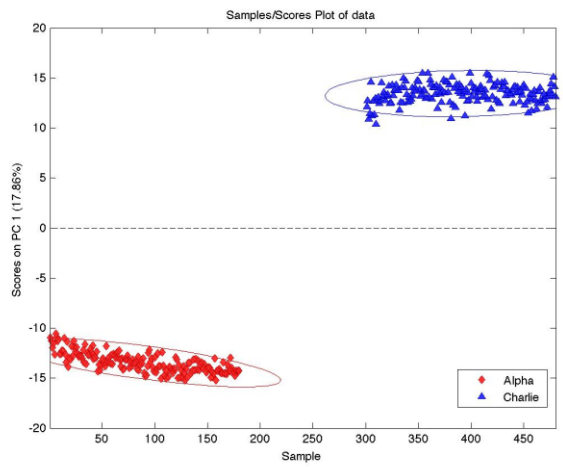

Figure 4: PCA model of Alpha and Charlie (1x1 detector, 5 second spectra)

IV. Future Research

Deployment efforts will continue at H-Canyon, capturing the processing of used nuclear fuel, plutonium blending efforts, as well as other targets of opportunity that occur at H-Canyon. Different and more complex multivariate approaches are also being explored and developed to optimize the analysis methods for spent nuclear fuel, including accounting for all of

the variations associated with it, as well as the various reprocessing techniques and parts therein. Efforts to improve material control and accountancy (MC&A), process control, safety, efficiency, and overall plant performance will continue to be optimized as well.

IV. CONCLUSIONS

The MIP monitor approach has been shown to be able to distinguish different fuel characteristics and process conditions using the gamma-ray spectra collected from various process streams by several different detector types. Further research and development is necessary to advance the MIP monitor to a deployable systems, but the technique holds promise as an online, non-destructive, near real-time, process monitor capable of assisting the regulatory and operator with material control and accounting.

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H Canyon, which is located at the Savannah River Site, is the only hardened nuclear chemical separations plant still in operation in the United States. In 2013, the Department of Energy issued a Record of Decision (ROD) enabling H-Canyon to process used nuclear fuel from domestic and foreign research reactors through 2018. The ROD will be instrumental in furthering research efforts in Safeguards and Fuel Cycle Research and Development by providing a facility that can be used as a live testing environment

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