Multivariate Analysis of Gamma Spectra to Characterize Used Nuclear Fuel

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Abstract

The Multi-Isotope Process (MIP) Monitor provides an efficient means to monitor the process conditions in used nuclear fuel reprocessing facilities to support process verification and validation. The MIP Monitor applies multivariate analysis to gamma spectroscopy of key stages in the reprocessing stream in order to detect small changes in the gamma spectrum, which may indicate changes in process conditions. This research extends the MIP Monitor by characterizing a used fuel sample after initial dissolution according to the type of reactor of origin (pressurized or boiling water reactor; PWR and BWR, respectively), initial enrichment, burn up, and cooling time. Simulated gamma spectra were used to develop and test three fuel characterization algorithms. The classification and estimation models employed are based on the partial least squares regression (PLS) algorithm. A PLS discriminate analysis model was developed which perfectly classified reactor type for the three PWR and three BWR reactor designs studied. Locally weighted PLS models were fitted on-the-fly to estimate the remaining fuel characteristics. For the simulated gamma spectra considered, burn up was predicted with 0.1% root mean squared percent error (RMSPE) and both cooling time and initial enrichment with approximately 2% RMSPE. This approach to automated fuel characterization can be used to independently verify operator declarations of used fuel characteristics and to

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inform the MIP Monitor anomaly detection routines at later stages of the fuel reprocessing stream to improve sensitivity to changes in operational parameters that may indicate issues with operational control or malicious activities.

Keywords: Used Nuclear Fuel, Fuel Characterization, Multivariate Analysis, Gamma Spectroscopy

1. Introduction

Current approaches to nuclear material control and accountancy (MC&A) rely on destructive assay (DA) techniques, such as thermal ionization mass spectrometry, to deter and detect theft of special nuclear materials (SNM) at nuclear reprocessing plants. Although these DA measurements are highly accurate and precise, they require extensive effort by skilled staff. While analytical results are being generated, significant quantities of SNM could be diverted from a large reprocessing facility. Additionally, the errors associated with material balances using these measurements scale with the size of accountancy tank. Without new methods to augment the current destructive assay-based MC&A approaches, these errors may limit the size of a material balance area or of the overall reprocessing facility. Online, near-real-time process monitoring methods can be used in concert with traditional MC&A measures to reduce the resources necessary to detect material diversion, improve the timeliness of detection and MC&A decisions, and support large-scale used nuclear fuel reprocessing facilities.

The Multi-Isotope Process (MIP) Monitor is one approach to online process monitoring in nuclear fuel reprocessing facilities. The monitor uses multivariate analysis of gamma spectra collected at key points in reprocessing streams to detect small changes in the measured spectra that may indicate changes in process conditions. By targeting the full suite of gamma-emitting indicator isotopes, small, inexpensive, portable gamma detectors can be deployed throughout an existing facility to support the MIP Monitor. Because the MIP Monitor focuses on process monitoring, it could be used as an operator support tool for pro-

25 cess control in reprocessing facilities or as a safeguards tool by the regulating
authority; the same multivariate analysis techniques support either capability.

In past research, a database of training spectra covering normal operating conditions across a variety of used nuclear fuel types was used to train these multivariate methods [1, 2]. This approach can identify gross changes in process parameters, but small deviations may be obfuscated by the large number
30 of conditions lumped into a single model. As the range of normal conditions included in training increases (e.g., by including fuel samples from many different reactors operating under different conditions), the sensitivity of the model to small deviations may decrease. This research extends the MIP Monitor by
35 characterizing the used fuel samples at the initial dissolution stage. By characterizing the used fuel, multivariate models of normal behavior at later stages in the reprocessing stream can be trained specific to the conditions expected for the present used nuclear fuel sample. This will improve the sensitivity of the MIP Monitor to very small changes at later stages in the process. Additionally,
40 this automated fuel characterization can be used to verify operator declarations of used fuel inventories unobtrusively and independently, effectively in real time.

1.1. Background

The MIP Monitor is designed to track plant conditions indirectly and non-destructively by monitoring multiple gamma-emitting radioactive isotopes through-
45 out an aqueous reprocessing system. The distribution of many fission and activation products is influenced by several operational variables, including acid concentration, TBP concentration, temperature, and used fuel characteristics (e.g. reactor type, burn up, cooling time). For a consistent industrial process with reproducible conditions, the distribution of each isotope between the
50 organic and aqueous phases should be relatively constant. Consequently, the distribution of isotopes in both product and waste streams at each stage in the facility should be relatively reproducible (within normal industrial variations), resulting in a signature of “normal” process conditions. Under abnormal conditions (such as those expected under some protracted diversion scenarios), the

55 distribution of isotopes in the various streams may change measurably [3, 4]. The MIP Monitor approach capitalizes on these expected changes in the distribution of gamma-emitting isotopes in the aqueous and organic phases to serve as an indication of “off-normal” conditions in the process chemistry. The MIP Monitor relies on gamma detectors that can be deployed at key locations
60 throughout an existing facility. Multivariate analysis of gamma spectra is used to automatically and autonomously process the spectra from these detectors in real time, continuously monitoring for indications of changing stream conditions. Initial simulations and bench-scale experiments reported in prior work have documented the performance and potential of the MIP Monitor [1, 2]. This
65 approach can identify gross changes in process parameters, but small deviations may be obfuscated by the range of normal conditions, resulting in part from the variety of used nuclear fuel that can be reprocessed in a given facility. As the range of normal conditions included in training increases, the sensitivity of the model to small deviations and anomalies may decrease. This research
70 extends the MIP Monitor by characterizing the used fuel samples after initial dissolution according to the reactor type (pressurized or boiling water reactor; PWR or BWR, respectively), initial enrichment, burn up, and cooling time. This characterization allows for independent verification of operator-declared fuel inventories. Additionally, multivariate models used to monitor later stages
75 in the reprocessing facility can be trained using only gamma spectra of fuel samples similar to the current one according to the estimated fuel characteristics. This will improve the sensitivity of the MIP Monitor to very small changes that may be indicative of undesirable process changes or diversion of special nuclear material.

80 1.2. Organization of the Article

This article presents a framework for multivariate analysis of gamma spectra for the purpose of characterizing used nuclear fuel. The following section describes the multivariate analysis methodology used: partial least squares regression, as well as two special cases, partial least squares discriminate anal-

85 ysis and locally weighted partial least squares regression. The specific locally
 weighted partial least squares implementation described here differs from other
 approaches in the information used to identify local observations and calculate
 weights. This is followed by a summary of the simulation of used nuclear fuel
 composition and resulting gamma spectra in Section 3. Results of the simula-
 90 tion, modeling, and characterization are given in Section 4. Finally, conclusions
 are drawn and discussion of the expected efficacy of the method on real-world
 data is given.

2. Methodology

Several multivariate analysis methods were investigated for application in
 95 this study, including methods for classification and estimation. Previous work
 with the MIP Monitor has demonstrated the applicability and efficacy of partial
 least squares regression (PLS) to gamma spectra [1, 2]. The analysis models
 employed to characterize simulated used fuel samples are all based on PLS: PLS
 discriminate analysis (PLSDA) for classification, and both PLS and locally-
 100 weighted PLS (LWPLS) for estimation.

PLS is a linear regression model that capitalizes on the colinearity of input
 variables, a feature of most data that makes it unsuitable for traditional linear
 regression. PLSDA adapts PLS for classification by applying a threshold to
 the PLS output to infer whether an observation is in a specific class. LWPLS
 105 extends the inherently linear PLS algorithm to nonlinear data by assuming the
 data is approximately linear in some neighborhood about the query observation.
 LWPLS preserves the globally non-linear information by creating locally linear
 models as new observations are evaluated. As new query observations are pre-
 sented, the LWPLS model trains a new PLS model "on the fly" by using only
 110 training data within some pre-defined neighborhood of the new observation.
 The PLS, PLSDA, and LWPLS algorithms used in this research are described
 in the following sections, preceded by a brief discussion of the preprocessing
 methods used to prepare gamma spectra for modeling.

2.1. Multivariate Analysis Algorithms

115 Preprocessing input data before performing multivariate analysis can simplify calculations by removing systematic deviations, such as noise, mean signal values, or signal background, that do not provide useful information about the process or system. Data preprocessing can be performed within the variables or within the observations. Observations can be normalized to reduce the effect
120 of relative intensity across multiple observations. In gamma spectra analysis, this would be necessary if counting time or detector geometry is allowed to vary between observations; however, in this study the counting time and geometry are kept constant, and so observation intensity is retained because it is expected to contain important information for the used fuel characterization. In the analyses presented here, the spectra channels are mean-centered; that is, the mean
125 of each variable (energy channel) in the training data is subtracted from each observation of that variable (energy channel) to give a post-processed variable mean of zero in the training data. This mean centering removes the large, but uninteresting, offset of the channel average value from zero while maintaining
130 the relative deviation of a particular observation of that channel from the mean. This is particularly important for PCA and PLS analysis, because these methods identify underlying components that capture data variation from the origin. If the mean values of each variable are not removed from the training data, the fitted components will attempt to capture that variation; typically the first
135 principal component (PCA) or latent variable (PLS) will point to the mean of the training data. Other data preprocessing methods can be applied to reduce noise and maximize the signal-to-noise ratio (SNR). The simulated spectra used in this work are noise-free, so no noise analysis is investigated here. In a deployed system, noise analysis and reduction, as well as background subtraction,
140 may become important.

2.1.1. Partial Least Squares Regression

Partial least squares regression (PLS) was originally developed in the field of econometrics, but has found more ubiquitous application in chemometrics.

Practically, its application is not limited to chemical analyses; PLS is a supervised transformation technique that captures colinearity between input variables as well as correlation of inputs to the model output by identifying the underlying latent variables in the input and output space. A fully motivated derivation of PLS is given in [5]; an abbreviated treatment is given here.

PLS combines the attractive features of principal component analysis (PCA) with multiple linear regression. PCA is an unsupervised method for identifying the underlying uncorrelated components in a set of correlated variables [6, 7]. The first principal component (PC) lies in the direction of greatest variance of the original data set. Subsequent PCs are orthogonal and lie in the direction of the greatest remaining variation. Each subsequent PC contains less information (variance) from the original data than the PCs before it. Typically, small PCs contain primarily noise and are excluded from the PC space, so that PCA can also be used as a dimension reduction technique. PCA decomposes input variables into a set of independent components that explain the variance in the input space. PLS extends PCA by identifying components (here called latent variables) that capture both the correlations in the input variables and the covariance between the input and output variables. That is, the latent variables (LVs) are preferentially chosen to both explain variance in the input space and to maximize their predictive potential for the dependent variable.

There are several iterative algorithms for fitting the PLS model; one such algorithm is shown in Algorithm 1 [8]. PLS training begins by identifying a vector, u_1 in the input space that has large correlation with the output. The input variables are projected on this direction to compute the corresponding regression coefficient, β_1 . Further directions in the input space and regression coefficients are identified by repeating this process with the residual signal (that is, the portion of the signal that has not been captured in previous latent variables) in the input and output space. Here, X is the training input data, and y is the training output data. Because the rank of y remains unchanged through PLS model development, the maximum number of LVs that can be identified is equal to the rank of X . Typically, however, fewer LVs are retained in the model

Algorithm 1: PLS Training and Prediction, from [8]

Training:
 $X_1 = X$
 $y_1 = y$
for $i = 1$ *to* k **do**
 $\mathbf{u}_i = X_i^T y_i$
 $\mathbf{s}_i = X_i \mathbf{u}_i$
 $\beta_i = \mathbf{s}_i^T y_i / (\mathbf{s}_i^T \mathbf{s}_i)$
 $y_{i+1} = y_i - \beta_i \mathbf{s}_i$
 $\mathbf{p}_i = X_i^T \mathbf{s}_i / (\mathbf{s}_i^T \mathbf{s}_i)$
 $X_{i+1} = X_i - \mathbf{s}_i \mathbf{p}_i^T$
end

Prediction:
 $x_1 = x$
 $y_1 = 0$
for $i = 1$ *to* k **do**
 $\mathbf{s}_i = x_i \mathbf{u}_i$
 $y_{i+1} = y_i + \beta_i \mathbf{s}_i$
 $x_{i+1} = x_i - \mathbf{s}_i \mathbf{p}_i^T$
end
 $\hat{y} = y_{k+1}$

175 because, as in PCA, the later LVs likely contain primarily noise and information
 180 unuseful for predicting y . The algorithm shown selects some preselected number
 of LVs, k , **based on prior knowledge of the true dimensionality of the system**.
 Another approach is to select LVs until the output residual (the unexplained
 information in y , given by y_{i+1}) is smaller than some specified threshold.

180 In addition to the traditional PLS algorithm, two variations are used in this
 research: PLS discriminate analysis for classification and locally weighted PLS
 for estimation. These variations are described in the following subsections.

2.1.2. PLS Discriminate Analysis

The PLS algorithm given in Algorithm 1 can be easily adapted for classifica-
 185 tion purposes, commonly known as PLS discriminate analysis (PLSDA) [9]. In
 PLSDA, the output is a vector of binary dummy variables indicating whether
 the observation belongs to a specific class according to equation 1.

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

With this encoding, the PLS algorithm will preferentially select between-

group variance that discriminates between classes. In the prediction step, the
 190 membership of an observation in each class is estimated; these continuous mea-
 sures of membership are converted to binary numbers by either applying the
 threshold or a winner-take-all approach.

2.1.3. Locally Weighted PLS

Conventional PLS is an inherently linear modeling technique, which can limit
 195 its applicability to nonlinear datasets. However, most nonlinear surfaces can be
 considered approximately linear in some small neighborhood about a point. Lo-
 cally weighted models are often used to apply linear methods to nonlinear data
 by building local models around each new prediction point. Locally weighted
 PLS (LWPLS) can preserve the globally nonlinear information by creating lo-
 200 cally linear PLS models as new observations are made [10, 11, 12].

LWPLS differs from conventional PLS by the introduction of a weighting
 matrix, W , that determines the relative importance of each training observa-
 tion in determining LVs and regression parameters. In the LWPLS training
 algorithm, $\mathbf{u}_i$, β_i , and $\mathbf{p}_i$ are calculated according to the formulae in equation
 205 2.

$$\begin{aligned}\mathbf{u}_i &= X^T W y_i, \\ \beta_i &= \mathbf{s}_i^T W y_i / (\mathbf{s}_i^T W \mathbf{s}_i), \text{ and} \\ \mathbf{p}_i &= X_i^T W \mathbf{s}_i / (\mathbf{s}_i^T W \mathbf{s}_i),\end{aligned}\tag{2}$$

where W is a diagonal matrix that contains the weight for each of the exemplar
 input/output pairs in X and y . The weights can be defined in any way that
 indicates the similarity of a new observation to the historic (exemplar) data.
 A Gaussian kernel is commonly used to convert the distance between the new
 210 query input and an exemplar input to a similarity or weight [10, 11]. In this
 approach, the weight for an exemplar vector with respect to a new query vector

is given by equation 3:

$$w_i = e^{-d_i^2/h^2}, \quad (3)$$

where h is the kernel bandwidth and the distance, d , is typically given by the Euclidean distance (equation 4):

$$d_i = \sqrt{\sum_{k=1}^N (x_{ik} - q_k)^2}, \quad (4)$$

215 where x_{ik} is the value of the k^{th} variable in the i^{th} exemplar observation and q_k is the k^{th} variable in the query vector. The kernel bandwidth, h , **controls the relationship between the distance and the resulting weight**. The MIP Monitor characterization algorithm takes a similar approach to determining the weights of each exemplar vector; however, the similarity is calculated based on the
220 similarity of the fuel characteristics that have been estimated in previous stages of the characterization for a new query (i.e., reactor type, initial enrichment, burn up, or cooling time) to the known values of these characteristics for the exemplar data. This weighting scheme does not explicitly evaluate the similarity of a query spectrum to the exemplar spectra, but instead relies on the similarity
225 of quantified fuel characteristics. **This novel weighting approach differs from previously developed LWPLS methods that use the data observations themselves to evaluate similarity [10, 11]**. The weight of the i^{th} exemplar observation for a new prediction is given by the product of the available estimated characteristic similarities (equation 5):

$$w_i = \prod_k w(\hat{c}_k, c_{i,k}), \quad (5)$$

230 where k is the set of characteristics that have been estimated in previous stages of the query characterization, $\hat{c}_k$ is the estimated value of the k^{th} characteristic for the query observation, and $c_{i,k}$ is the known value of the k^{th} characteristic for the i^{th} exemplar. The similarity for binary characteristics (i.e., reactor type,

Rx) is calculated as in equation 6.

$$w(\hat{c}_{Rx}, c_{i,Rx}) = \begin{cases} 1 & \hat{c}_{Rx} = c_{i,Rx} \\ 0 & \hat{c}_{Rx} \neq c_{i,Rx} \end{cases} \quad (6)$$

such that the resulting w_i will be zero if the predicted reactor type does not
 235 match the exemplar reactor type. The similarity for continuous characteristics
 (initial enrichment, burn up, cooling time) is calculated using the Gaussian ker-
 nel in equation 3. The kernel bandwidth, h , is optimized for each characteristic
 individually, h_k , based on the range of values of the characteristic and the ad-
 240 ditional predictive power gained by including that information in the model.
 As h_k approaches zero, equation 3 approaches the binary weighting in equation
 5, effectively producing a nearest-neighbor matching; conversely, as h_k becomes
 large relative to the expected distance between query and exemplar features, the
 weights all asymptotically approach one, and the LWPLS algorithm converges
 245 to the traditional PLS model.

2.2. Fuel Characterization Algorithms

Three characterization algorithms were compared. The first algorithm per-
 formed all characterizations in parallel, meaning that fuel type, burn up, en-
 richment, and cooling time were characterized using only the measured gamma
 250 spectrum (Figure 1). The second algorithm, referred to as Algorithm A and
 shown in Figure 2, first classified the fuel type (PWR or BWR), and then used
 reactor type-specific PLS models to predict burn up, enrichment, and cooling
 time. Finally, the third algorithm (Algorithm B, Figure 3) is fully serial, where
 the fuel type is first classified; then the burn up is estimated using a reactor
 255 type-specific PLS model; next the predicted reactor type and burn up are used
 to train a LWPLS to predict cooling time; and initial enrichment is finally es-
 timated using an LWPLS model weighted according to the estimated reactor
 type, burn up, and cooling time. **Because the weights in each subsequent model
 depend on the fuel characteristics previously estimated, the order of evaluation
 260 affects the overall algorithm performance.** The evaluation order for the final

Figure 1: Flow Diagram for Parallel Analysis

Figure 2: Flow Diagram for Algorithm A

Figure 3: Flow Diagram for Algorithm B

algorithm was selected through trial-and-error by testing each permutation of evaluation order on a set of test data to optimize the overall performance across all four fuel characteristics, represented by the average root mean squared percent error across the four characteristics.

265 3. Application to Simulated Spectra

The data used to develop and test the analysis methodology were generated using ORIGEN-ARP version 5.1.01 and ENDF/B-VI nuclear data [13]. The development data consisted of three PWR and three BWR reactor designs, six initial enrichment values evenly distributed among typical commercial values, 270 four cooling times, and nine discharge burn up values (Table 1). Each simulation used three cycles of equal length (18 months) with 100% availability. The

first two cycles are followed by a 30-day outage, and the third cycle is followed
 the relevant fuel cooling time. The cooling times were chosen based on the stor-
 age capacity at Rokkasho Reprocessing Plant, a commercial fuel reprocessing
 facility in Japan [14]. A consistent reactor operating history was maintained
 in this study to evaluate the ability to estimate fuel characteristics alone; the
 effects of operating history on the efficacy and accuracy of this approach should
 be investigated in future work. The 200 nuclides with the highest activity in
 the ORIGEN-ARP output of each fuel sample were retained and used to simu-
 late the gamma spectra of the fuels [15]. This resulted in 218 unique nuclides,
 although any single simulation had only 200 non-zero nuclide concentrations.
 Benchmark studies of ORIGEN-ARP/ORIGEN-S [16, 17, 18] were reviewed to
 gauge the accuracy of the nuclide simulations, which would in turn indicate the
 accuracy of the simulated gamma spectra. The results of this benchmarking
 evaluation are available in [19]. Only benchmarking studies evaluating used
 fuel similar to those simulated here were considered. Of the fourteen nuclides
 evaluated for accuracy, most were accurate to 10%, although ^{126}Sn and ^{79}Se
 have reported error of 202.0 and 980.0%, respectively. These large errors are
 typically attributed to inaccurate cross section data used for simulation. De-
 spite the large errors in these and potentially other key nuclides, the overall
 trends in between-sample spectra variations are thought to be representative of
 the variations expected in practice. Artifacts in the simulations caused by the
 miscalculation of key nuclides limit our ability to directly apply the developed
 models to measured UNF spectra. Because the MIP Monitor analysis capitalizes
 on these between-sample variations for fuel characterization instead of the in-
 tensity of any specific peak or set of peaks, the proof-of-principle demonstration
 with these data suggest that this approach is viable for real-world deployment
 if appropriate measured spectra are available to train the classification and es-
 timation models.

The 200 most abundant nuclides for each of the simulated fuel samples are
 input to SYNTH, a program developed at Pacific Northwest National Labora-
 tory (PNNL) to simulate the measured gamma spectrum for a given source and

Table 1: Used Fuel Characteristics for Data Simulation. Fuel and gamma spectra are simulated for each combination of reactor design, BU, IE, and CT.

Reactor Design	Burn Up (MWd/kgU)		Initial Enrichment (%)	Cooling Time (years)
CE14x14 (PWR)	20	36	2.5	2
S14x14 (PWR)	25	40	3.0	3
W17x17(PWR)	30	45	3.5	4
Atrium9x9 (BWR)	34	50	4.0	5
GE7x7 (BWR)	35		4.5	
SVEA64 (BWR)			5.0	

Table 2: HPGe Detector and Electronics Characteristics in SYNTH Simulation of Gamma Spectra

End Cap	Thickness	0.5 mm
	Material	Aluminum
	Spacing	0.67 mm
Dead Layer	Thickness	0.67 mm
Sensor	Diameter	6.67 cm
	Length	6.67 cm
	Efficiency	66.60 %
	Resolution @ 1332 keV	1.67 keV
Counting Parameters	Zero	0 keV
	Gain	1 keV/ch
	Quad	0 keV/ch ²
ADC	Channels	4096
	Live Time	666 sec
Full Scale Energy		4096 keV

detector configuration [20]. For this analysis, the gamma source was treated as a point source 25 cm from a coaxial high-purity germanium (HPGe) detector; pertinent characteristics of the detector and electronics are listed in Table 2.

The 1296 simulated gamma spectra were divided into three independent sets for model development and testing. The training data set, which is used to train the various PLS-based characterization models, includes 722 spectra that cover the range of simulated values in each channel and the range of fuel characteristics. This coverage is important to limit the extrapolation that the model must perform when presented with new spectra; empirical models generally should not be relied on to extrapolate outside of their training region. The test data contains 324 spectra, which are used to optimize the trained PLS-based models for the number of latent variables and the bandwidth used to determine weights in LWPLS models. The remaining 324 spectra are used to characterize the performance of the fuel characterization algorithm, the so-called validation data. The characterization results presented in the next section are evaluated on the validation data.

4. Results

Three primary algorithms were evaluated for characterization of used nuclear fuel according to reactor type, initial enrichment, burn up, and cooling time, as described in section 2.2. The results of each of these algorithms are evaluated on accuracy and runtime. The MIP Monitor must be able to make accurate characterization in near-real-time, in order to improve MC&A and process monitoring at reprocessing facilities. Essentially, the MIP Monitor must evaluate key fuel characteristics correctly and before the next spectrum is available.

The PLS algorithms used here are data compression techniques that combine related and redundant information across multiple spectrum channels into underlying LVs. The simulated HPGe gamma spectra include 4096 energy channels, effectively 4096 input variables for the multivariate analysis. These channels are compressed into a smaller subset of LVs, each of which contains some portion of each energy channel. The PLSDA model developed for reactor type classification uses thirty-six latent variables. The PLS and LWPLS models developed for estimating the IE, BU, and CT include from sixteen to fifty LVs.

335 All of these models provide a significant reduction of dimensionality from the original 4096 energy channels. The accuracy results presented below suggest that the true dimensionality of these data is significantly lower than the number of energy channels available; these extra dimensions can confound other multivariate regression techniques that lack the data compression of PLS.

340 4.1. Characterization Accuracy

Three algorithms are evaluated for characterizing used nuclear fuel: parallel analysis (Figure 1), Algorithm A (Figure 2), and Algorithm B (Figure 3). In the parallel analysis, each of the four primary characteristics is evaluated in parallel using only the measured gamma spectrum. In Algorithm A, the reactor type is first classified, and the remaining three characteristics are estimated using reactor type-specific PLS models. In the final approach, Algorithm B, the classification is fully serial, where the classification results from the previous stage are used to develop a new LWPLS model on the fly to evaluate the fuel characteristic at the current stage. The accuracy of each of these algorithms is compared according to the root mean squared percent error (RMSPE), which is calculated according to equation 7:

$$RMSPE = \frac{\sqrt{E[(\hat{y} - y)^2]}}{E[y]} \times 100\% \quad (7)$$

where $\hat{y}$ is the prediction of a specific fuel characteristic whose true value is y and $E[\cdot]$ is the expected value.

The accuracy of each of the three fuel characterization algorithms for predicting the four key fuel characteristics is summarized in Table 3. Here, the results are presented for each algorithm predicting the characteristic for all of the validation data and for the PWR and BWR fuels separately, as well as the overall results combining both PWR and BWR fuels. For all evaluated characterization algorithms, a PLSDA model was trained that perfectly classifies used fuel according to reactor type (PWR or BWR); the same model is used in each algorithm. Figure 4 shows the scores of the test data on latent variables 1 and 2 of the PLSDA classification model. As the figure shows, there is significant

overlap between the two reactor types in two dimensions; the discrimination is not visually obvious in two or three dimensions. As discussed previously, thirty-

365 six latent variables are necessary to accurately discriminate the reactor type. Algorithm A shows a significant improvement over the parallel analysis for the data evaluated, with improvements in accuracy of a factor of two to four for prediction of initial enrichment, burn up, and cooling time. Algorithm B shows further improvement over Algorithm A for prediction of initial enrichment and

370 cooling time. Due to the weighting scheme used, the LWPLS model used to predict the burn up in Algorithm B is effectively identical to the reactor type-specific model used in Algorithm A. The LWPLS models used in later stages of Algorithm B (i.e., prediction of initial enrichment and cooling time) make use of the additional fuel characteristics predicted in earlier stages of the algorithm.

375 The accuracy results achieved in this evaluation are unrealistically low, because the simulated data do not contain measurement noise or random, normal perturbations in the fuel isotopics. However, these results suggest that fuel characterization of dissolved used nuclear fuel samples through multivariate analysis of gamma spectra can be effective.

380 4.2. Runtime Results

The MIP Monitor is intended to work in real-time (or near-real-time) to monitor reprocessing streams. Generally, the monitor is expected to use 30-second spectra (or longer); therefore, the multivariate analysis methods employed should complete predictions in under 30 seconds to provide information

385 in effectively real-time. This implementation was evaluated on a 3.40 GHz Intel Pentium 4 processor. Execution speed for a single sample was evaluated by averaging the elapsed time for 324 validation samples. The MATLAB implementation averaged 1.85 seconds per evaluation; the model evaluation can be performed faster than the expected sampling time, providing real time fuel

390 characterization.

5. Conclusion

Table 3: Fuel Characterization Error for Parallel Analysis, Algorithm A, and Algorithm B. Error is given as RMSPE (% , top) and 95-percentile ranges of error in engineering units (bottom).

		Initial			Cooling
		Reactor	Enrichment	Burn Up	Time
		Type	(%)	(MWd/kgU)	(years)
Parallel Analysis	Overall	0.00	10.94%	0.36%	6.62%
			[-0.64 0.92]	[-217.1 360.5]	[-0.45 0.40]
	PWR	0.00	10.54%	0.42%	8.65%
			[-0.39 1.09]	[-179.8 332.0]	[-0.36 0.46]
	BWR	0.00	11.32%	0.29%	3.81%
			[-0.68 0.68]	[-221.0 429.9]	[-0.52 0.33]
Alg. A	Overall	0.00	2.71%	0.11%	3.39%
			[-0.20 0.18]	[-77.6 96.3]	[-0.28 0.22]
	PWR	0.00	3.13%	0.09%	2.96%
			[-0.24 0.18]	[-55.3 71.2]	[-0.21 0.23]
	BWR	0.00	2.23%	0.13%	3.79%
			[-0.17 0.18]	[-97.5 105.3]	[-0.31 0.23]
Alg. B	Overall	0.00	1.78%	0.11%	2.13%
			[-0.14 0.11]	[-77.6 96.3]	[-0.08 0.13]
	PWR	0.00	1.69%	0.09%	1.26%
			[-0.16 0.12]	[-55.3 71.2]	[-0.07 0.12]
	BWR	0.00	1.87%	0.13%	2.80%
			[-0.12 0.10]	[-97.5 105.3]	[-0.09 0.13]

This investigation extends the previously developed MIP Monitor approach for process monitoring and MC&A at used nuclear fuel reprocessing facilities to support fuel characterization at early stages in the reprocessing stream through multivariate analysis of gamma spectra. This approach to used fuel characterization provides an independent verification of operator declarations and can improve the ability of the MIP Monitor to detect anomalous conditions at later

Figure 4: PLSDA Scores on Latent Variable 1 and 2 for the Test Data

stages in the fuel reprocessing stream. This initial proof-of-concept study used simulated data from ORIGEN-ARP and SYNTH to develop models and algorithms for fuel characterization. The performance of the developed characterization algorithms, both in terms of accuracy and computational speed, suggests that similar analyses could be successfully developed for *in situ* fuel characterization at an operating aqueous reprocessing facility. The proposed framework for fuel characterization is expected to be generally applicable, but new models and characterization algorithms will need to be developed using experimental or operational spectra.

This initial proof-of-principle study investigates a "best case" scenario for the proposed fuel characterization and declaration verification framework. Spectra are simulated for high-resolution gamma detectors (HPGe), which are unlikely to be deployed in operating fuel reprocessing facilities. Shielding, containers, and conduits that may attenuate gamma radiation are not included in the spectra simulation. Further, background contributions to the spectra are neglected. Initial investigations at the H-Canyon facility at Savannah River Site suggest

that background correction and spectrum alignment will be necessary prepro-
cessing steps for measurements made at multiple locations within the facility
[21]. These same data collection campaigns have successfully deployed NaI(Tl)
detectors at various stages throughout the facility, suggesting that cryogenically-
cooled, high-resolution detectors will not be necessary in a deployment scenario.
A targeted campaign to collect both gamma spectra and meta-data on a variety
of typical used nuclear fuel samples at initial dissolution will be necessary to
develop and fully test the proposed characterization framework.

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