A Machine Learning Method for the Forensics Attribution of Separated Plutonium

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Abstract—*A nuclear forensics technique, based on maximum likelihood method, for the attribution of reactor-type, fuel burnup, and time since irradiation (TSI) of separated pure plutonium (Pu) samples was previously developed at Texas A&M University. The method utilized measured values of ten intra-elemental isotope ratios in the Pu sample and a large database consisting of the values for these ratios as a function of the three attributes: reactor-type, fuel burnup, and TSI. However, this method failed for Pu samples with mixed attributes. Hence, a new technique based on machine learning methods was developed that matched the capabilities of the previous maximum likelihood method for pure Pu samples. This new methodology used Support Vector Machines for reactor-type discrimination and Gaussian Process Regression for fuel burnup quantification. The TSI was calculated analytically using the predicted reactor-type and fuel burnup. This new method holds great potential for the attribution of mixed Pu samples.*

Keywords—*Nuclear forensics; Separated plutonium; Machine learning; Reactor-type; Time since irradiation; Fuel burnup.*

I. INTRODUCTION

The specter of nuclear terrorism is an existential threat currently facing the United States.[1] The unpredictable nature of terror strikes—in conjunction with the damage that an improvised nuclear device (IND) could inflict—represent a nightmare scenario for the national security complex. To act proactively, the global community must ensure that special nuclear material (SNM), especially

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plutonium (Pu), is never able to fall into the hands of these rogue nonstate actors. The two acquisition pathways that an adversary of this nature could use to acquire weapons-usable Pu would be the theft of the material from a nuclear facility or a clandestine material hand-off from a state actor that is sponsoring the terror group. In either case, a robust nuclear forensics methodology that is able to find the age and origin of the SNM could be used to deter these activities from taking place.

Should a Pu sample be interdicted, either in the course of crossing an international border or as the result of operation by a law enforcement or intelligence agency, it is of great importance to obtain the most information possible about the Pu sample to best aid the conventional investigation. Knowledge of the origin of a Pu sample or the sample's age could yield useful answers about how the material was procured and who procured it, enabling authorities to punish the organization, or organizations, responsible and also to implement measures to ensure it does not happen again.[2] The capability to connect the material to those responsible and mete out the penalties for such extreme actions could dissuade an organization from even attempting to procure nuclear material. In the case of theft, this could be accomplished by shoring up physical protection systems or identifying any insiders who might have aided the organization that stole the nuclear material. Nuclear forensics can also be a deterrent to a state actor supporting nuclear terrorism. The hand off of nuclear material from a state to a terrorist organization is an unprecedented, but feared scenario. A state might engage in this activity as a means to further its foreign policy goals by enabling a closely allied terrorist group to make attacks that the state would be unable to make under international law. If there was a significant chance that the hand off of nuclear material could be attributed to a state, the state would most likely not engage in that activity due to the severity of the repercussions, whether those are of an economic or military nature. A nuclear forensics

methodology capable of attributing Pu could also be used to verify the declarations of facilities under an International Atomic Energy Agency (IAEA) safeguards agreement. This would deter the misappropriation of Pu, as any attempt to conceal the diversion of Pu from declared activities could be detected. There would also be an incentive to making sure records are as accurate as possible and that all security and material accounting measures are adequate.

A comparison of the concentration of isotopes in different Pu samples acts as a signature that can be used to find different attributes of the sample's history. Many studies have demonstrated that the type of reactor that produced Pu can be discriminated using these signatures[3,4,5]. These methods of differentiation leverage the fact that irradiation in a particular reactor-type (hence, with distinct neutron energy spectra) will lead to differences in the fission product yields and neutron interaction cross sections resulting in the concentrations of isotopes produced during the course of irradiation being unique to those reactor conditions. The areas where the use of isotope concentration comparisons requires some care is when discriminating between reactor-types with similar fuel compositions and spectral attributes, as the difference in concentrations will not be as stark. This can be overcome by selecting the appropriate isotopes for comparison to ensure that there is enough difference between the concentrations. Another aspect where great care must be taken is in the analysis of separated Pu samples due to differences in the separation efficiencies of different elements.[6]. To ensure that the nuclear forensics work is accurate, the comparison of isotope concentrations must either account for how the separative process changes the isotope concentrations relative to each other, or must be agnostic of the separation method, such that the separation efficiencies need not be accounted for the isotope concentrations that are analyzed. Depending on what isotope concentrations are analyzed, this method does not adequately distinguish between samples in the low fuel burnup range, which coincidentally represents the

ideal condition for producing weapons usable Pu. The reason why the differentiation is more difficult for these conditions is that some isotopes need more operational time to build up or reach their equilibrium values. This problem can be avoided by selecting appropriate isotopes for analysis, ones that can be reliable even in low fuel burnup Pu samples.

A nuclear forensics methodology developed by Osborn et al. at Texas A&M University utilizes a set of intra-elemental isotope ratios [$^{137}Cs/^{133}Cs$, $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$, and $^{242}Pu/^{239}Pu$] to attribute separated Pu's production reactor-type, fuel burnup, and TSI (Refs. 3 and 4). Isotope ratios of the same element are used to keep the forensics methodology agnostic of the separation method for the reasons stated below. During the separation of Pu from irradiated nuclear fuel, most commonly performed using solvent extractions like the Plutonium Uranium Redox Extraction (PUREX), some elements will also be separated out with the Pu in trace amounts. The amount of an elemental impurity in the separated Pu compared to the amount that was initially in the spent nuclear fuel is the element's decontamination factor. The decontamination factor for each element separated in a chemical process will depend on many factors; however, for isotopes of the same element, the decontamination factor will be the same, negating the need to know the separation process.[6,7] So, the use of a set of intra-elemental isotope ratios ensures that the nuclear forensics methodology is useful regardless of the chemical separation process used to separate Pu from the irradiated nuclear fuel.

In the Osborn et al. methodology, the discrimination of the three parameters (reactor-type, fuel burnup, and TSI) of interest is performed using a maximum likelihood calculation to compare a set of ten intra-elemental isotope ratios for an unknown sample against a database of isotope ratios created using fuel burnup simulations (Refs. 3 and 4). The set of isotope ratios in the database that

is found to be the most similar to the unknown ratio set indicates the most likely reactor-type, fuel burnup, and TSI of the unknown Pu sample. In this maximum likelihood method, all isotope ratios in the ratio set are used to find all three parameters of interest at the same time. The accuracy of this methodology was validated using measurements of Pu produced using experimental irradiations in two different research reactors and with treatments to produce samples with both thermal and pseudo-fast neutron energy spectra (Refs. 4 and 5). The methodology performed very well and was able to correctly identify the reactor-type where the Pu samples were produced in, as well as estimate the fuel burnup and TSI. It is worthwhile to note that among the ten isotope ratios, some of these ratios are interdependent but the maximum likelihood method treated them as independent features.

While the maximum likelihood methodology performs well for straightforward samples, one potential area of concern for this methodology is its inability to provide meaningful predictions for samples that are mixtures of Pu from different reactor-types or fuel burnups or TSI. This is because the isotope ratio set for a mixed sample is quite dissimilar to the ratio sets within the database. A possible remedy to this would be to include mixed ratio sets within the database. This approach would massively increase the size of the isotope ratio set database, as each additional reactor-type would actually increase the total number of reactor-types enormously. This would add to the computation time required for attribution, as the probability of each entry in the reference database matching the unknown sample must be computed. Further including variations in mixing compositions would increase the size of the database and as a result the computation time for a discrimination even more. One way to reduce the computation time and database size for the maximum likelihood method would be to implement more involved methods, such as the Latin Hypercube sampling of the database. In addition, directed search algorithms could be used to more

quickly narrow down on which areas of the problem domain are most likely to match the experimental sample being tested. These improvements could make the maximum likelihood much more agile. These problems could also be alleviated by methods that use machine learning. Even though the number of training datasets needed for machine learning model increases when additional reactor types or variations (mixed compositions) are added, the size of each training dataset being smaller compared to the maximum likelihood reference database is an advantage for a machine learning methodology. More importantly, the attribution time will not increase in the case of a machine learning methodology because once a machine learning model has been trained, the attribution process consists of solving the functions produced during training, a relatively faster process. Another problem with the maximum likelihood approach is that in certain complex problems, it is likely to yield multiple answers, and the current maximum likelihood method has no way of effectively deciding between multiple maximum logarithmic-likelihood values. Machine leaning methodology is also vulnerable to such a problem of yielding multiple answers. However, there are a few aspects in the machine learning methodology developed that insulates from that possibility and the affect it might have on the prediction. This is achieved first, by the reactor-type classification not being adjudicated by exceeding a threshold for positive classification, but rather the output is the reactor class that has the highest maximum posteriori probability. In this approach, it is also technically possible for there to be multiple predictions, which is made unlikely through careful feature selection to ensure that each class has features that are sufficiently different from each other. Also, by separating the classification of reactor-type and quantification of burnup into two separate steps, the machine learning method developed greatly reduces the chance of experiencing multiple answers or maxima. The maximum likelihood approach used does not separately assess each parameter of interest, which increases the possibility

of multiple maxima. This is because in the maximum likelihood approach developed, the features that do not provide information on certain parameters are included regardless of predicting that parameter because of the convoluted three-parameter-in-one-step approach compared to the deconvoluted three-step approach employed in the machine learning approach developed.

The study presented in this paper seeks to build upon the foundations laid in the maximum likelihood methodology created by Osborn et al.; however, it uses predictive models trained with machine learning techniques in place of the maximum likelihood calculation (Refs. 3 and 4). This will lead to a methodology that can be more easily adapted to accommodate mixed Pu samples. The overall scope of this paper involves matching the performance of the Osborn et al. methodology with pure Pu samples to verify that the machine learning approach is a valid next step (Refs. 3 and 4). Additionally, feature selection techniques will be used to further examine how the different isotope ratios contribute to the prediction of each parameter. The rest of the paper is organized as follows. Description of the new nuclear forensics methodology development is provided in Section II. Results obtained using the new methodology and discussion of results are presented in Section III. Salient conclusions of this study are provided in Section IV.

II. DESCRIPTION OF THE NEW NUCLEAR FORENSICS METHODOLOGY DEVELOPMENT

II.A. Foundational Work from the Maximum Likelihood Methodology

The concept of utilizing intra-elemental isotope ratios for the Pu attribution, developed by Osborn et al. using the maximum likelihood method, laid the foundation for the new method based on the machine learning techniques. A database containing the values of those intra-elemental isotope ratios, in separated Pu, as a function of reactor-type, fuel burnup, and TSI was produced as part of the maximum likelihood method, but a portion of this database could also be used as a machine learning training dataset. The ideal formulation of the Osborn et al. nuclear forensics

methodology used ten intra-elemental isotope ratios: $^{137}Cs/^{133}Cs$, $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$, and $^{242}Pu/^{239}Pu$. These isotopes were selected because they contribute information on at least one of the three parameters of interest, were of a high enough yield to be measurable in separated Pu samples, and some of them had a long enough half-life to be a useful signature for the estimation of TSI.

In the maximum likelihood methodology, an unknown sample's isotope ratio values are compared against a database of reference isotope ratio values, which was produced by modeling various reactors using the Monte Carlo N-Particle (MCNP) transport code and its evaluated nuclear data library (ENDF/B-VIII)[8] and performing fuel burnup calculations for a range of 0 to 5 GWd/MTU. The MCNP simulation results were expanded by using linear interpolation between the fuel burnup time steps to produce isotope ratios at 100 fuel burnup values. The values of the isotope ratios after decay were also calculated for 5,000 days in 1-day time steps. This interpolation and decay scheme lead to a matrix of 500,000 data points for each reactor-type to compare against using the maximum likelihood calculation. Each point in the database had stored values for the aforementioned set of ten intra-elemental ratios for a given reactor-type, fuel burnup, and TSI. Osborn et al. simulated nine different reactor types: a generic pressurized water reactor (PWR) with ^{235}U enrichments of 2.35 at. %, 3.4 at. % and 4.45 at. %; a generic pressurized heavy water reactor (PHWR); the blanket region of a fast breeder reactor (FBR) containing depleted UO_2; a generic UK MAGNOX reactor; a generic Canadian NRX reactor; a simulation of an irradiation conducted at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory, and a simulation of an irradiation at the University of Missouri Research Reactor (MURR) (Refs. 3 and 4). The two simulations of irradiations at HFIR and MURR were included because the validation data was produced by performing experimental irradiations at these two reactors, and this

necessitated including MCNP simulations in the reactor-type database that mirrored the conditions that created those samples. The HFIR experimental irradiation was conducted on depleted UO_2 samples surrounded by a thermal neutron shield (gadolinium) and this was reflected in the MCNP simulations that were incorporated into the database. The use of gadolinium shielding was done to emulate the type of fast neutron flux that might be seen in the blanket region of an FBR. The experimental irradiation at MURR was conducted using natural UO_2 samples in a thermal neutron energy spectrum to mimic the environment and fuel composition that might be similar to that of a PHWR. The real-world measurements of the separated Pu samples that resulted from these two experimental irradiations were used to validate the methodology.

The attribution of an unknown sample's parameters of interest (reactor-type, fuel burnup, and TSI) is conducted using maximum likelihood estimation. This is performed by taking the product of the likelihoods that each of the ten ratios of the experimental sample matches with the corresponding ratio for a point in the reference database. The model, *M*, matching the measured vector, r_{mes}, given by Eq. 1 is used to compute the likelihood and subsequently the log-likelihood values were estimated using Eq. 2 to avoid the computational overflow errors.

$$L(M|r_{mes}) \propto L(r_{mes}|M) = \prod_{j=1}^{n} \frac{1}{\sigma_{j,sim}\sqrt{2\pi}} exp\left\{-\frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right\} \quad (1)$$

$$Log\ L(M|r_{mes}) = \sum_{j=1}^{n}\left[log\left(\frac{1}{\sigma_{j,sim}\sqrt{2\pi}}\right) - \frac{(r_{j,mes}-r_{j,sim})^2}{2\sigma_{j,sim}^2}\right] \quad (2)$$

where

$r_{j,mes}$ = j^{th} intra-element isotope ratio (for example $^{137}Cs/^{133}Cs$) in a vector of *n* measured intra-element ratios (in this paper *n*=10)

$r_{j,sim}$ = j^{th} intra-element isotope ratio in a vector of *n* simulated intra-element ratios from model, *M*, (database)

$\sigma_{j,sim}$ = uncertainty associated with the j^{th} intra-element ratio from vector, r_{sim}.

The calculated likelihood and loglikelihood values obtained are indicative of each reactor model, *M*, whether the measured material is from that reactor, with higher likelihood or loglikelihood values indicating a higher possibility of that reactor being the source of the material.

The maximum likelihood calculations using Eqs. 1 and 2 are conducted for every point in the database. The logarithmic-likelihood values can be mapped, creating a surface for each reactor-type of interest. The highest peak found across all of the surfaces indicates the predicted reactor-type, fuel burnup, and TSI value (Ref. 3).

II.B. Machine Learning Methodology for Nuclear Forensics Attribution of Plutonium

The machine learning-based nuclear forensics methodology presented here utilizes the same foundations as the previous methodology. The isotope ratios used in the machine learning predictive models were selected from the ten previously used ratios, and training data was generated using the simulations that formed the backbone of the maximum likelihood approach's reference database. The continued use of MCNP simulations to populate the reference database was found to be appropriate, because any stochastic error introduced was found to be within acceptable limits for this type of analysis.[9,10,11] There is a large structural difference in how the machine learning and the maximum likelihood approaches utilize the information. The maximum likelihood approach uses all of the isotope ratios to resolve all three parameters of interest simultaneously. With the machine learning approach used in this work, only one parameter of interest (reactor-type, fuel burnup, TSI) is resolved at a time, and only the isotope ratios that contribute to the discrimination of that parameter are used. The machine learning approach breaks the Pu attribution into the reactor-type determination using a classifier, followed by the fuel burnup quantification using a regression model, and then the TSI is solved for analytically using the

predicted reactor-type and fuel burnup. Out of the 10 isotope ratios used in the maximum likelihood method, only eight were selected for use in the machine learning methodology. The $^{242}Pu/^{239}Pu$ ratio was discounted because in our datasets ^{242}Pu does not appear in measurable quantities until relatively late in the fuel burnup range, meaning it offered little value when compared to the other isotope ratios. The $^{137}Cs/^{133}Cs$ ratio was also discounted because only one ratio is required for the TSI calculation, rendering it redundant to the $^{134}Cs/^{137}Cs$ ratio. In some cases, the $^{137}Cs/^{133}Cs$ ratio could provide value in analyzing plutonium samples that are fairly old. Of the eight ratios used in the machine learning method, the reactor-type determination utilized the following seven ratios: $^{135}Cs/^{137}Cs$; $^{136}Ba/^{138}Ba$; $^{150}Sm/^{149}Sm$; $^{152}Sm/^{149}Sm$; $^{154}Eu/^{153}Eu$; $^{240}Pu/^{239}Pu$; and $^{241}Pu/^{239}Pu$ and the fuel burnup quantification utilized $^{134}Cs/^{137}Cs$; $^{136}Ba/^{138}Ba$; $^{150}Sm/^{149}Sm$; $^{152}Sm/^{149}Sm$; $^{154}Eu/^{153}Eu$; $^{240}Pu/^{239}Pu$; and $^{241}Pu/^{239}Pu$. Six of the isotopic ratios used were common to both the reactor-type classification and the fuel burnup quantification. The $^{135}Cs/^{137}Cs$ ratio was used only for reactor-type classification and not for the fuel burnup quantification, whereas the $^{134}Cs/^{137}Cs$ ratio was used in the fuel burnup quantification but not in the reactor-type classification. The $^{134}Cs/^{137}Cs$ ratio was also the sole ratio used for the TSI calculation. The machine learning models that produced the results found in Section III.A were trained using these aforementioned eight intra-element isotope ratios. It is important to note for the case of two validation datasets using measured samples of Pu, presented in Section III.B, some isotope ratios were not available for use, this is true for both the maximum likelihood methodology as well as machine learning methodology. In both the HFIR and MURR experimental Pu samples, the $^{136}Ba/^{138}Ba$ ratio of the sample was unable to be accurately quantified due to Ba contamination from the glass vials that held the samples. Additionally, for the MURR Pu sample, the $^{154}Eu/^{153}Eu$ ratio was below the minimum detection threshold. As a consequence, the results presented in

Section III.B were produced using models that were trained to accommodate the reality of the samples. A reactor-type classifier and set of burnup quantification models were trained for the HFIR validation by omitting the $^{136}Ba/^{138}Ba$ ratio as a model feature. Similarly, a reactor-type classifier and set of burnup quantification models were trained for the MURR validation by omitting the $^{136}Ba/^{138}Ba$ and $^{154}Eu/^{153}Eu$ ratios as model features.

The training data set produced for the machine learning approach covered the same fuel burnup range (0-5 GWd/MTU) and TSI range (0-5,000 days) that the maximum likelihood approach used. Presently the machine learning approach is only capable of differentiating between five reactor types: a generic PWR with fuel enrichment of 2.35 at. %; a generic PHWR; the blanket region of an FBR containing depleted UO_2; and the two experimental irradiations at HFIR and MURR. The machine learning methodology should be able to accommodate all of the reactor-types investigated in the maximum likelihood methodology, but the five reactors listed were used to simplify the initial stage of methodology development. The training data set was produced by randomly sampling a reactor-type, fuel burnup value, and TSI value. An interpolation and decay calculation were then conducted to find the isotope ratio set that corresponded to those three parameters of interest. This process was used to produce a training data set of 12,500 isotope ratio sets (roughly 2,500 per reactor type). At the same time that the training data set was created, a testing data set was also created by randomly sampling an additional 12,500 isotope ratios, ensuring that the values of the testing data set were unique from those of the training data set. The isotope ratios of the training dataset were preprocessed by taking the common logarithm of each value to minimize the effects that the vast differences in quantity magnitude can have. Additionally, a Z-score standardization was performed on the training data set to further scale the data set to ensure that features that happen to have higher magnitudes will not dominate the predictions. The sample

means and standard deviations of the training data sets were saved for use in standardizing the testing and validation data.

II.B.1 Reactor-type determination using a Support Vector Machine (SVM) based classifier

The classification portion of the machine learning approach uses these seven isotope ratios: $^{135}Cs/^{137}Cs$; $^{136}Ba/^{138}Ba$; $^{150}Sm/^{149}Sm$; $^{152}Sm/^{149}Sm$; $^{154}Eu/^{153}Eu$; $^{240}Pu/^{239}Pu$; and $^{241}Pu/^{239}Pu$. Due to either their sensitivity to a reactor's neutron energy spectrum or to a reactor's neutron flux magnitude, these seven ratios offer insight into what reactor-type might have produced an unknown sample. Many different machine learning algorithms were tested, including decision trees, discriminant analysis, logistic regression, naïve Bayes classifiers, nearest neighbor classifiers, and ensemble classifiers.[12] Ultimately, it was determined that a Support Vector Machine (SVM) based classifier would be the most applicable, since the preliminary testing results had the highest accuracy amongst all the methods tested. The SVM classifiers create hyperplanes to partition the data set according to what class the data belongs to, which is done to maximize the margin around the separating hyperplane.[13,14]. The use of hyperplane boundaries minimizes the chance of a misclassification. The advantage of using SVM for classification is that these boundaries can be found even if the data set is not linearly separable, by using a kernel trick to transform the input space into a higher dimension that is linearly separable. In the SVM used for this nuclear forensics methodology development, the best kernel was found to be cubic, which takes the form in Eq. 3, where x_1 represents an observation and x_2 is a training data value that is compared to x_1 (Ref. 12).

$$K(x_1, x_2) = (x_1 \cdot x_2 + 1)^3, \qquad (3)$$

This specific classification technique and kernel were found to be optimal for this problem and the data set. As greater complexity is added to the problem domain, other classification techniques

and kernels should be tested to see if better performance can be achieved. The same is true for the choice of regression algorithm and kernel discussed in the next section.

II.B.2 Fuel burnup quantification using Gaussian Process Regression (GPR) algorithm

The fuel burnup quantification in the machine learning approach uses these seven isotope ratios: $^{134}Cs/^{137}Cs$; $^{136}Ba/^{138}Ba$; $^{150}Sm/^{149}Sm$; $^{152}Sm/^{149}Sm$; $^{154}Eu/^{153}Eu$; $^{240}Pu/^{239}Pu$; and $^{241}Pu/^{239}Pu$. The algorithm selected was Gaussian Process Regression (GPR), which performed better in initial testing than various other model types, including linear regression, regression tree, SVM, and ensembles of tree-type models (Ref. 12). A GPR is a nonparametric probabilistic model that uses a distribution of all admissible functions that fit the data to create the best predictive function for the response variable given the input. A Gaussian process is a set of random variables, such that any finite number of them have a joint Gaussian distribution (Ref. 14). By applying a kernel, or covariance, function, with the parameters determined by the multivariate Gaussian, a smooth function that accurately predicts a response variable can be created. The use of covariance function narrows the group of all possible functions that fit the data by determining the most similar observations. In this study, the kernel used was exponential (Eq. 4), where σ_l is the characteristic length scale, which designates how input values have to be apart before they are considered uncorrelated, σ_f is the signal standard deviation, x_i and x_j are two input values whose responses are being compared, θ represents the set of hyperparameters including σ_l and σ_f, and d is defined by Eq. 5 (Ref. 12).

$$k(x_i, x_j|\theta) = \sigma_f^2 e^{-\frac{d}{\sigma_l}} \tag{4}$$

$$d = \sqrt{(x_i - x_j)^T (x_i - x_j)} \tag{5}$$

II.B.3 Analytic estimation of time since irradiation (TSI)

With the reactor-type and fuel burnup predicted, the TSI can be calculated analytically. For this methodology the $^{134}Cs/^{137}Cs$ ratio was used as the primary signature, although other isotope ratios could be used, provided they have appropriately long half-lives. The first step in the discrimination of TSI was to determine the value of the $^{134}Cs/^{137}Cs$ ratio at the time of discharge by interpolating from the reference isotope library using the predicted reactor-type and burnup. With knowledge of the values of the ratio at fuel discharge, C_0, and the experimental ratio, $C(t)$, as well as the ^{134}Cs and ^{137}Cs half-lives ($t_{½,134}$ and $t_{1/2,137}$), then the time since irradiation (TSI), t, can be solved using Eq. 6. A visual depiction of how the overall Pu attribution methodology proceeds, as well as which inputs are required at each step, can be found in Fig. 1.

$$C(t) = C_0 e^{-t\left(t_{1/2,134} - t_{1/2,137}\right)} \qquad (6)$$

A few feature selection techniques were also used to determine whether the machine learning approach can be further improved. The first method used was a Minimum Redundancy Maximum Relevance (MRMR) algorithm on the reactor classifier training data set. An MRMR algorithm finds which features (isotope ratios) most effectively predict the response variable while being the most dissimilar to other features (Ref. 15). This analysis indicates the most impactful features for use in classification. The other feature selection technique used was a Neighborhood Component Analysis (NCA) for each of the fuel burnup quantification training datasets (Ref. 16). This finds which features have the best effect on prediction accuracy by quantifying the regression accuracy loss when a feature is left out. The MRMR and NCA are useful indicators, but true performance loss is not known until various machine learning models are trained and tested using different subsets of the features available, which were also performed as part of the nuclear forensics methodology development. It should be noted that isotope ratios should not be eliminated on the

basis of poor performance in the feature selection techniques; as greater complexity is added in the problem domain, a previously unattractive isotope ratio might prove useful.

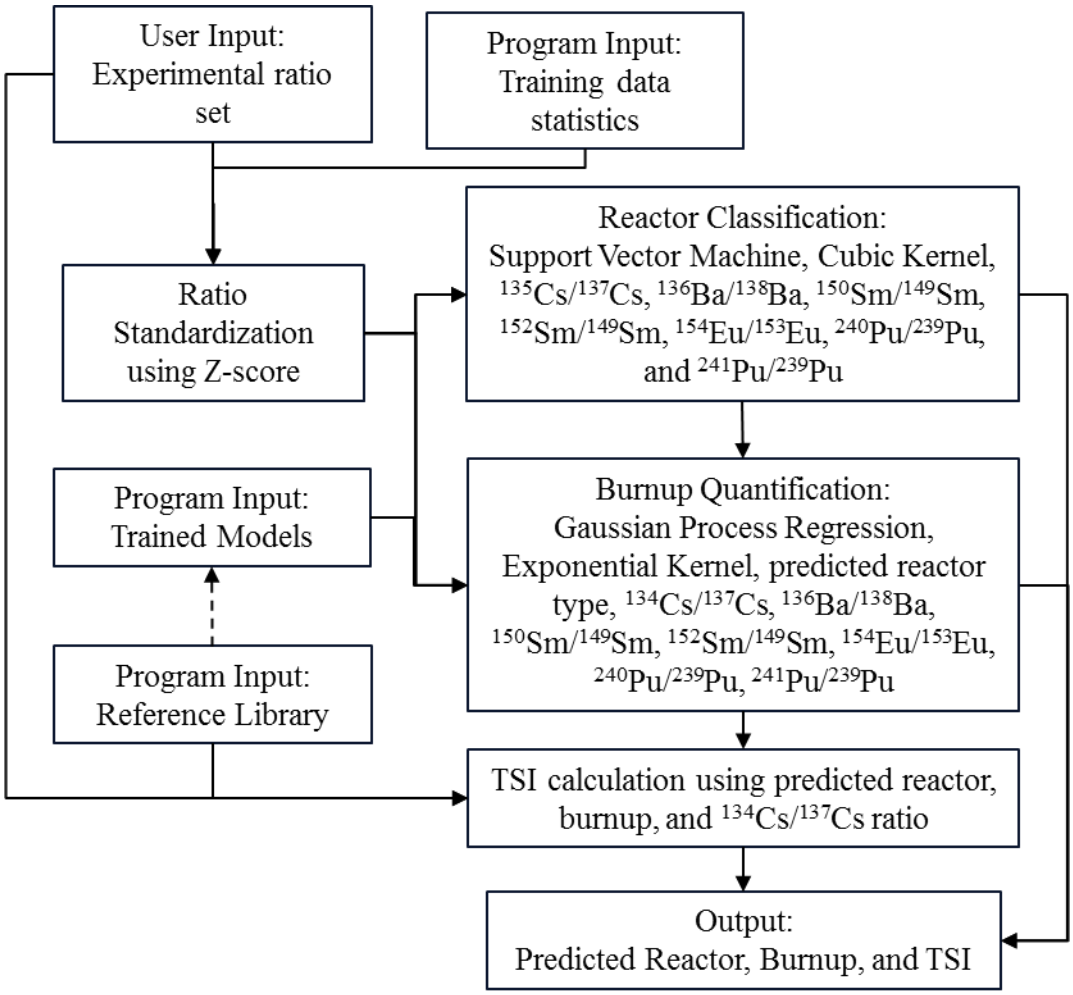

Fig. 1. Flow chart detailing the machine learning method of Pu attribution.

III. RESULTS AND DISCUSSION

III.A. Performance of Reactor-type Attribution and Fuel Burnup Quantification Models

After attributing every value in the testing data set of 12,500 points, the accuracy of the SVM-based reactor-type classifier was found to be 99.9%. The high accuracy value indicates that the model was properly fitted. The accuracy value also suggests that the features (isotope ratios) used

in the methodology have distinct features for each reactor-type that give adequate information to make the determination. The few misclassifications that did occur were in an extremely low fuel burnup range (<0.01 GWd/MTU). This is expected because at such a low burnup many of the isotope ratios used would not have had the time to properly develop the features that allow for the reactor-type identification. The Root Mean Square Error (RMSE) values for each burnup quantification model corresponding to each reactor-type were calculated using Eq. 7, where $\hat{y}_t$ represents the predicted burnup value and y_t represents the actual burnup value.

$$RMSE = \sqrt{\frac{\sum_{t=1}^{T}(\hat{y}_t - y_t)^2}{T}} \tag{7}$$

The RMSE values for each fuel burnup quantification model are provided in Table I. It is important to note that a testing point only contributed to a reactor's RMSE if the classifier correctly identified the reactor-type. One interesting feature is that the RMSE values for the fuel burnup quantification model tended to inversely correlate to the amount of fuel burnup time steps in the MCNP simulations, with the exception of the RMSE value for HFIR. The PHWR fuel depletion simulation had the fewest fuel burnup time steps followed by HFIR, PWR and FBR, and lastly MURR with the most fuel burnup time steps in their MCNP simulations.

TABLE I
RMSE Values for the Fuel Burnup Quantification Models

Reactor Type	RMSE (GWd/MTU)
PWR	0.149
PHWR	0.202
FBR	0.116
MURR	0.034
HFIR	0.054

Estimation of the uncertainty associated with the predictions made by the fuel burnup quantification models was done by determining the standard deviation of the predicted value. It can be obtained from the predicted posterior distribution of the GPR. The variation in standard deviation for the PWR, PHWR, and FBR models are listed in Table II.

TABLE II

Uncertainty of PWR, PHWR, and FBR Fuel Burnup Quantification Model

Burnup (GWd/ MTU)	PWR			PHWR			FBR		
	Prediction (GWd/ MTU)	σ (GWd/ MTU)	Error (%)	Prediction (GWd/ MTU)	σ (GWd/ MTU)	Error (%)	Prediction (GWd/ MTU)	σ (GWd/ MTU)	Error (%)
0.5	0.485	0.0277	5.71	0.52	0.0243	4.66	0.539	0.0268	4.97
1	0.984	0.0219	2.23	1.02	0.0189	1.85	1.042	0.025	2.40
1.5	1.492	0.0203	1.36	1.52	0.0221	1.46	1.539	0.0215	1.40
2	2.003	0.0181	0.90	2.01	0.0178	0.89	2.025	0.0236	1.17
2.5	2.518	0.0194	0.77	2.50	0.0185	0.74	2.509	0.0245	0.98
3	3.036	0.0192	0.63	2.99	0.0188	0.63	2.990	0.0259	0.87
3.5	3.557	0.0206	0.58	3.47	0.0197	0.57	3.468	0.0277	0.80
4	4.079	0.0205	0.50	3.96	0.0203	0.51	3.945	0.0284	0.72
4.5	4.600	0.0205	0.45	4.28	0.0217	0.51	4.413	0.0293	0.66

Table II shows that the percent error of all three models decreased as a function of burnup, which can be explained by the fact that as samples reach higher burnup values the behaviors of the isotope ratios become more resolved from each other, enabling more accurate quantification. A similar method of quantifying the uncertainty of the reactor-type classifier can be conducted utilizing the classification scores and posterior probability values produced by the model. A thorough examination of these indicators over the problem space could be an interesting avenue for future studies.

III.B. Experimental Validation of Pu Attribution Using the New Machine Learning Methodology

The reactor-type classifier and fuel burnup quantification models were validated using the chemically separated Pu data obtained from the experimental uranium samples irradiated at HFIR and MURR. The performance of the machine learning methodology was compared with that of the maximum likelihood methodology. The machine learning models used in this comparison were trained with the isotope ratios that were able to be experimentally measured in the HFIR and MURR Pu samples, rather than the full isotope ratio set. This means that the models used in this section to attribute the HFIR Pu sample did not include the $^{136}Ba/^{138}Ba$ ratio in training and models used in this section to attribute the MURR sample did not include the $^{136}Ba/^{138}Ba$ or $^{154}Eu/^{153}Eu$ ratios in training. This is in contrast to the previous performance assessment using the testing data set generated from MCNP simulation values, which utilized models that contained all the eight isotope ratios of interest. The machine learning models' performance attributing the experimental Pu samples can be found in Tables III and IV respectively for HFIR and MURR experimental irradiations. From Tables III and IV, it can be inferred that the machine learning models were able to correctly classify both reactor-types and also accurately quantify the fuel burnup and TSI for the samples, the performance of which is comparable to that of the maximum likelihood method.

TABLE III

Performance of Attributing the HFIR Plutonium Validation Sample

	Burnup (GWd/MTU)	TSI (days)
Experimental	4.36 ± 0.28	1601
Maximum Likelihood	4.29 ± 0.01	1834 ± 54
Machine Learning (SVM-GPR)	4.20 ± 0.059	1820 ± 6

TABLE IV

Performance Attributing the MURR Plutonium Validation Sample

	Burnup (GWd/MTU)	TSI (days)
Experimental	0.973 ± 0.032	318
Maximum Likelihood	1.01 ± 0.02	305 ± 83
Machine Learning (SVM-GPR)	1.045 ± 0.0336	315 ± 38

III.C. Investigation of Isotope Ratio Importance using Feature Selection Algorithms

The results of the MRMR analysis for the reactor-type classifier can be found in Fig. 2. The analysis indicated that the isotope ratios that give the most unique information to the classifier were the $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, and $^{240}Pu/^{239}Pu$ ratios.

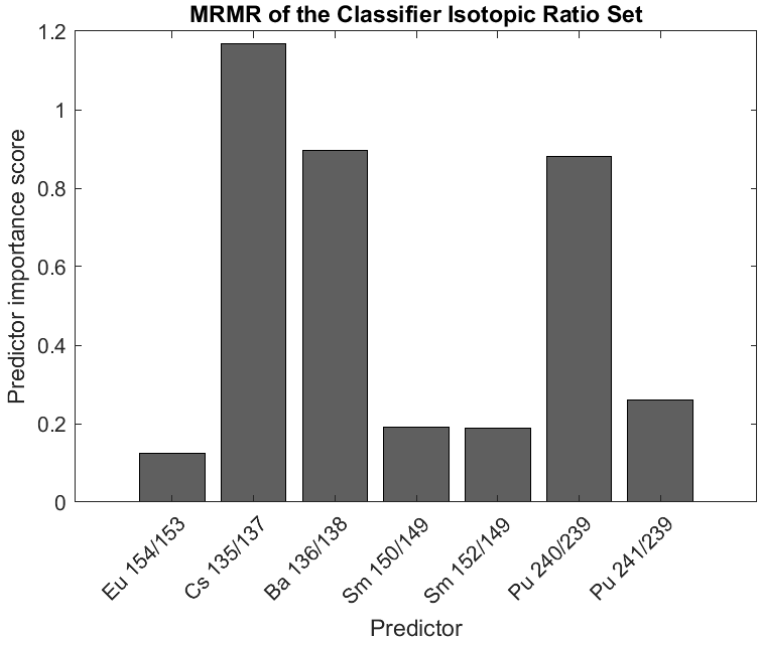

Fig. 2. MRMR analysis for the isotope ratio set of the reactor-type classifier.

The results of the NCA analysis for each burnup quantification model are shown in Table V. For the thermal neutron energy spectra reactors (PWR, PHWR, and MURR experimental irradiation), $^{136}Ba/^{138}Ba$ proved to be the most important. These reactors all had similar NCA

results as a result of the similarities between their neutron energy spectra. The most impactful ratio for quantifying FBR fuel burnup was found to be the $^{152}Sm/^{149}Sm$ ratio. The $^{240}Pu/^{239}Pu$ ratio was the most impactful for the burnup of HFIR experimental irradiation which was carried out in a pseudo-fast neutron energy spectrum, although both Sm ratios also had high feature weights. The ratios, $^{134}Cs/^{137}Cs$, $^{154}Eu/^{153}Eu$, and $^{241}Pu/^{239}Pu$ were not highly weighted for any reactor-type. The poor performance of the $^{154}Eu/^{153}Eu$ in both the MRMR and NCA analysis indicates that it is the least valuable feature for the machine learning methodology to use.

TABLE V

Results of the NCA Analysis for Each Reactor-type*

	$^{154/155}Eu$	$^{134/137}Cs$	$^{136/138}Ba$	$^{150/149}Sm$	$^{152/149}Sm$	$^{240/239}Pu$	$^{241/239}Pu$
PWR	1.68×10^{-3}	1.04×10^{-4}	*7.49*	0.772	1.74	1.50	1.09×10^{-3}
PHWR	1.74×10^{-3}	3.34×10^{-5}	*7.82*	0.476	0.266	0.900	2.28×10^{-4}
FBR	2.18×10^{-3}	1.44×10^{-5}	0.427	0.650	*7.96*	1.17	1.92×10^{-4}
HFIR	1.70×10^{-5}	4.07×10^{-5}	2.69×10^{-4}	2.07	4.26	*7.05*	5.07×10^{-5}
MURR	3.79×10^{-5}	2.67×10^{-4}	*8.38*	1.05	2.83	1.69	1.32×10^{-4}

*Each reactor-type's highest weighted isotope ratio is indicated by italic typeface

III.D. Optimization of Number of Isotope Ratios for the New Machine Learning Methodology

The final step of this study was to determine the minimum number of features (isotope ratios) required to ensure a high degree of accuracy. Seven different sets of classifiers and regression models were trained using subsets of the isotope ratios used. These groupings were informed by the MRMR and NCA analyses. In an ideal world, as few isotope ratios as possible would be used. This would offer the maximum savings in model training time as well as the chemical separation laboratory work. However, if this methodology is to be useful for nuclear forensics, there need to be enough number of isotopes used so that concealing a Pu sample's attributes could not be done

easily done by tampering with only one or two isotope ratios. The various subsets of isotope ratios used for the optimization study are listed in Table VI. The selection of the isotope ratios subsets was done in a way to test not only different combinations of the isotope ratios, but also to provide some variation in the total number of isotope ratios used. The $^{154}Eu/^{153}Eu$ ratio was not included in many of the isotope ratio subsets, because it was uniformly one of the worst performing isotopes in the MRMR and NCA analyses. The $^{136}Ba/^{138}Ba$ was another isotope ratio that was underrepresented in the isotope ratio subsets. This is because, despite having highest importance in the MRMR and several of the NCA analyses, $^{136}Ba/^{138}Ba$ is a difficult isotope ratio to quantify in the laboratory, which makes it an unattractive candidate to use in a reduced isotope ratio set.

TABLE VI

Isotope Ratio Sets Investigated for Performance Testing of the New Nuclear Forensics Method Based on Machine Learning Technique

Case Number	Isotope Ratios Used
Base	$^{134}Cs/^{137}Cs$[a], $^{135}Cs/^{137}Cs$[b], $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$
1	$^{154/155}Eu$, $^{134/137}Cs^{*}$, $^{135/137}Cs^{\dagger}$, $^{240/239}Pu$, $^{241/239}Pu$
2	$^{150/149}Sm$, $^{152/149}Sm$, $^{134/137}Cs^{*}$, $^{135/137}Cs^{\dagger}$, $^{240/239}Pu$, $^{241/239}Pu$
3	$^{150/149}Sm$, $^{152/149}Sm$, $^{240/239}Pu$, $^{241/239}Pu$
4	$^{240/239}Pu$, $^{241/239}Pu$
5	$^{134/137}Cs^{*}$, $^{135/137}Cs^{\dagger}$, $^{150/149}Sm$, $^{152/149}Sm$
6	$^{134/137}Cs^{*}$, $^{135/137}Cs^{\dagger}$, $^{136/138}Ba$, $^{240/239}Pu$
7	$^{134/137}Cs^{*}$, $^{135/137}Cs^{\dagger}$, $^{240/239}Pu$, $^{241/239}Pu$

[a]Only used in burnup and TSI quantification
[b]Only used for reactor-type classification

An important consideration, when potentially using less features, is about the speed and ease of obtaining those features. The fastest way to quantify isotope ratios is with gamma or alpha

spectroscopy. This quantification can take place on the order of minutes. The isotope ratios that can be quantified entirely with these techniques are $^{134}Cs/^{137}Cs$ (gamma spectroscopy) and $^{240}Pu/^{239}Pu$ (alpha spectroscopy). ^{154}Eu can also be quantified using gamma spectroscopy, however ^{153}Eu is stable meaning that mass spectroscopy is required to fully quantify that isotope ratio. A feature that would require a moderate amount of time to quantify would be $^{241}Pu/^{239}Pu$, which can be quantified with gamma spectroscopy, but only after being chemically separated from other isotopes that drown out some of the lower magnitude gamma peaks produced in Pu samples. This quantification can take place on the order of hours. All the isotope ratios can be quantified using mass spectroscopy, although this is the slowest technique, taking on the order of days. Additionally, special care must be taken to ensure that any isobaric interference is corrected for when using this method, which adds a degree of difficulty that is not found when using gamma or alpha spectroscopy.

The reactor-type classifier and fuel burnup quantifiers trained for each case were tested with a set of 2,000 test points. Any misclassifications were tallied, and the RMSE was calculated for each case. The results of this testing and a comparison to the base case containing all isotope ratios are shown in Table VII. The results in Table VII indicate that high accuracy from the classifier can be maintained as long as four or more isotopes ratios are used in training. When dropping the number of isotope ratios (see Table VI) used in classifier training, the number of misclassifications begins to rise (Table VII), even if the three isotope ratios used are the three most important ratios indicated by MRMR, which can be seen in Case 6. The fuel burnup quantification RMSE values showed that the uncertainty in the fuel burnup quantification remains level across all cases, indicating that even if only two isotope ratios were used, accurate fuel burnup quantification can be expected,

provided the correct reactor-type is predicted in the previous step of this machine learning methodology.

TABLE VII

Reactor-type Classification and Fuel Burnup Quantification Performance with Different Isotopic Ratio Groupings in Machine Learning Technique

Case	Classifier Errors	Burnup RMSE		
		PWR	PHWR	FBR
Base	1	0.130	0.212	0.110
1	2	0.131	0.205	0.107
2	1	0.134	0.218	0.110
3	2	0.130	0.217	0.106
4	156	0.133	0.204	0.106
5	99	0.130	0.204	0.108
6	19	0.132	0.208	0.111
7	11	0.135	0.211	0.111

Another method to test the performance of each case's fuel burnup quantification models is to use the two experimental Pu samples produced at HFIR and MURR. The values of the predicted fuel burnup values for both the HFIR and MURR samples are shown in Table VIII, as well as the measured fuel burnups of the samples. One aspect to note is that Case 6 was unable to be used because it utilized $^{136/138}$Ba, which was not able to be measured for those two samples. Similarly, Case 1 with the MURR Pu sample was unable to be conducted due to the inability to measure the ^{154}Eu/^{153}Eu ratio for that sample. With regards to the HFIR sample, the fuel burnup quantification models for Case 2 and Case 3 were able to more closely predict the sample's fuel burnup than the base case, although they are all the within the error of the measured values. The only groupings that were not within the error of the HFIR sample's measured fuel burnup were Case 1, Case 5, and Case 7. When evaluating the performance in quantifying the fuel burnup of the MURR, the only subset of isotope ratios that improved upon the performance of the base case was Case 5,

although only marginally. Case 4 and Case 7 deviated the furthest from the fuel burnup of the MURR sample, but not to an alarming degree. A version of the maximum likelihood method that only used the isotope ratios of Case 3 (with only Pu and Sm ratios) was tested with the two experimental validation points for these ratios. For the HFIR case, the maximum likelihood method predicted the sample was from HFIR with a burnup of 4.37 ± 0.516 GWd/MTU. For the MURR case, the maximum likelihood method predicted that the sample was produced in MURR with a burnup of 1.02 ± 0.516 GWd/MTU. For both of these cases, the maximum likelihood method showed strong predictive performance although the uncertainty is quite high, which was expected because the Sm and Pu ratios are very strong indicators for reactor-type and burnup predictions.

Table VIII

Burnup Quantification Performance with Experimental Samples

Case	Burnup (GWd/MTU)	
	HFIR	MURR
Experimental	4.36 ± 0.28	0.973 ± 0.032
Base	4.20 ± 0.059	1.045 ± 0.0336
1	3.91 ± 0.047	Not Applicable
2	4.22 ± 0.059	1.045 ± 0.035
3	4.36 ± 0.047	1.052 ± 0.034
4	4.09 ± 0.021	1.091 ± 0.023
5	5.06 ± 0.070	1.032 ± 0.037
6	Not Applicable	Not Applicable
7	3.90 ± 0.047	1.059 ± 0.034

The TSI values were not calculated for the isotope optimization study because those calculations use the predicted reactor-type and fuel burnup to select the appropriate value of $^{134/137}$Cs ratio from the database. This means that any change in the predicted fuel burnup will change the TSI prediction, but in a predictable manner.

IV. CONCLUSIONS

The nuclear forensics methodology using machine learning techniques was able to perform at the same level as that of the maximum likelihood estimation based on the results shown in Tables III and IV to attribute the history of two Pu experimental samples produced at HFIR and MURR. Additionally, feature selection techniques were used to find which isotope ratios were the most impactful in each parameter's discrimination, and it was found that this attribution methodology still achieved high accuracy when limited to only using four isotope ratios. Machine learning algorithms can leverage relationships in data that many other approaches might miss and are capable of being very efficient in learning what information is required without challenging feature engineering. In this study, the number of isotope ratios used was reduced from ten to eight: one isotope ratio was eliminated due to poor quality of data over the data range of interest, while the other was eliminated because the machine learning approach only requires one isotope ratio for TSI calculation. As evidenced in Section III.D, there exists potential for further reduction in the number of isotope ratios required. Additionally, the machine learning approach ensures that the only isotope ratios used in the discrimination of an individual parameter of interest are those that contribute information. The machine learning approach outlined here offers a promising platform for continued methodology development, with the ultimate goal of including more complex samples, e.g. mixed Pu samples, and resolving additional parameters of interest like mixing composition. It is also planned to incorporate in the machine learning method the uncertainty in the measured values of 14 isotopes used as features (10 ratios).

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