Proceedings of the INMM & ESARDA Joint Annual Meeting
August 23-26 & August 30-September 1 2021, Virtual Meeting

ATTRIBUTION OF SEPERATED PLUTONIUM USING MACHINE LEARNING TECHNIQUES

Patrick J. O'Neal[1] and Sunil S. Chirayath[1,2]

Department of Nuclear Engineering, Texas A&M University, College Station, TX 77843-3133Center for Nuclear Security Science & Policy Initiatives, Texas A&M University, College Station, TX 77843-3473 [1] patrick.oneal@tamu.edu [2] sunilsc@tamu.edu

ABSTRACT

The ability to find the conditions in which a sample of plutonium was produced would be a powerful tool to support nuclear nonproliferation efforts. This capability would act as a deterrent to smuggling efforts, and also help regulatory agencies verify declared nuclear activities. Work at Texas A&M University yielded a nuclear forensics methodology, which is capable of determining separated plutonium's reactor-type of origin, burnup, and the time since irradiation (TSI)—three parameters of interest. The methodology used a set of ten intra-element isotopic ratios found in separated plutonium, which was compared to a library of isotopic ratio values produced using neutronics simulations for reactors of interest. By calculating the probability that the isotopic ratio set matched a set in the library, the methodology could predict the three parameters of interest of the sample. One shortcoming of this methodology was an inability to correctly attributing spoofed plutonium, where plutonium sourced from two different reactors or two different fuel burnup levels are mixed. A new methodology to rectify this vulnerability using machine learning (ML) technique is under development, instead of the maximum likelihood calculation previously used. The new methodology attributes in three steps, one for each parameter, rather than resolving all of the three parameters simultaneously like the previous maximum likelihood approach. First, a support vector machine classifier with a set of seven isotopic ratios finds the reactor of origin and a set of regression models trained using gaussian process regression predicts the burnup with a different set of seven isotopes. Last, TSI is calculated analytically using decay equations. Thus far, the new methodology is capable of attributing pure plutonium samples and has been validated using experimental data. The next step will to be augment the classifier training data set with spoofed plutonium data.

INTRODUCTION

Nuclear forensics capabilities to enable the attribution of illegal special nuclear material (SNM) could deter the adversary from attempting malicious acts. Another use for a nuclear forensics methodology capable of attributing the production conditions of SNM is the verification of declared activities at facilities under nuclear safeguards agreements with the International Atomic Energy Agency (IAEA). Hence, forensics methodology use-case could help deter diversion of SNM at the state level or the theft of SNM by facility insiders. Previous work at Texas A&M University produced a nuclear forensics methodology capable of finding a separated plutonium sample's reactor-type of origin, burnup, and time since irradiation (TSI) [1] [2]. This methodology utilized a set of ten intra-element isotope ratios as the main forensics signature and compared the isotope ratio values against a reference library of data in performing a maximum likelihood calculation to determine the most probable reactor-type, burnup, and TSI of the plutonium sample. This methodology proved very successful in the attribution of pure plutonium samples, those where all the plutonium was produced with the same reactor-type, burnup, and TSI, but it was

unable to attribute the samples effectively if there was any mixing. A new methodology that built upon the strengths of the previous work has been under development in hopes it can attribute these mixed or "spoofed" plutonium samples. The main change is that instead of attribution using a maximum likelihood calculation, the attribution is performed using models trained with machine learning techniques.

PREVIOUS METHODOLOGY

The main forensics signatures used in both methodologies is a set of intra-element isotope ratios. In the previous methodology $^{137}Cs/^{133}Cs$, $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, $^{241}Pu/^{239}Pu$, and $^{242}Pu/^{239}Pu$ were used [1]. The reason that the isotope ratios must not feature isotopes of different elements is to ensure that the methodology can be agnostic of the separation technique used to produce plutonium, as different proportions of each element will be separated depending on the details of the chemical separation process. A reference library was produced using a set of fuel depletion calculations for various reactor types for a range of burnup (0 to 5 GWd/MTU) and TSI (0 to 5000 days) using the Monte Carlo N-Particle Transport code (MCNP) [3]. The collection of burnup steps and times steps formed a reference matrix for each of the isotope ratios of interest. Some reactors modelled for inclusion in the methodology included a generic pressurized water reactor (PWR), a generic pressurized heavy water reactor (PHWR), and a generic fast breeder reactor (FBR), as well as experimental irradiation positions for the High Flux Isotope Reactor (HFIR) and Missouri University Research Reactor (MURR). The two experimental irradiation positions were used to produce validation data that was used to test the forensics methodology's performance [4] [5].

The actual attribution was performed by comparing the ten isotope ratios found using a collection of destructive and non-destructive assay techniques to each value in every reference matrix set. For each burnup and time point a product of probability values that an isotope ratio from the experimental sample matches the isotope ratio corresponding to that burnup and TSI value. The largest value for this product indicates which burnup and TSI value for each reactor would be the most likely to produce the set of isotope ratios found within the experimental ratio. Similarly, the reactor-type with the greatest maximum likelihood value is the most likely to have produced the experimental sample. A validation of this method was conducted by irradiating natural uranium at HFIR and MURR, and then performing separations and assays at Texas A&M University [4] [5]. In the validation, the correct reactor-type of origin, burnup, and TSI were found using the reference data library and the maximum likelihood calculation.

NEW MACHINE LEARNING METHODOLOGY

The primary functional difference between the previous methodology and the new machine learning methodology is the deconvolution of the resolution of the three parameters of interest as well as a reduction in intra-element isotope ratios required. The previous methodology resolved all three parameters at once, while the machine learning methodology divides the attribution into three steps, first a classification model is used to find the reactor-type, then a regression model is used to find the burnup value, and last an analytic calculation is to find TSI. In the machine learning methodology only eight isotope ratios were used: $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$. The first step of the machine learning methodology was to produce a training dataset. This was accomplished by randomly sampling a reactor-type, burnup, and TSI value from the available reactor types, 0-5 GWd/MTU, and 0-5,000

day respectively, and then using the simulated data from the previous methodology's MCNP simulations the burnup interpolation and time decay calculations were performed to produce the isotope ratio set corresponding to those parameters of interest. The optimal training data set size was found to be 12,500 data points, which corresponds to 2,500 data points per reactor type. A logarithm transformation and a Z score standardization was used on the training data set. At the time of generating the training data, a test data set of the same size was also produced, with great care taken to ensure that no point was common to both sets.

MATLAB Statistics and Machine Learning Toolbox [6] was used to train the reactor-type classifier and burnup quantification models. The reactor-type classification model was trained using Support Vector Machines (SVM). The SVM classifier used a cubic kernel and the $^{135}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$ isotope ratios. A Gaussian Process Regression (GPR) was used to train the burnup quantification regression models. One model was trained per reactor-type of interest. The GPR kernel was exponential and the $^{134}Cs/^{137}Cs$, $^{136}Ba/^{138}Ba$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$ isotope ratios were used. The TSI quantification was performed using the $^{134}Cs/^{137}Cs$ ratio by interpolating what the $^{134}Cs/^{137}Cs$ ratio would have been at discharge from the reactor (TSI=0) using the predicted reactor-type, burnup, and the reference data library. With knowledge of the current $^{134}Cs/^{137}Cs$ ratio value and the discharge $^{134}Cs/^{137}Cs$ ratio the TSI can be solved for using a radioactive decay equation.

RESULTS

After the appropriate models were trained, they were tested for their accuracy using the testing data set. The reactor-type classifier achieved a very high accuracy (>95%). This confirms that the seven isotope ratios used have sufficiently distinct features for each reactor type. The only source of misclassifications were test data points that had very low burnup values (<10 MWd/MTU). This can be explained by the fact that at such low burnup values the isotope ratios have not had the opportunity to reach the trends required for reactor-type discrimination. The performance of the regression models for burnup quantification was evaluated by finding the Root Mean Square Error (RMSE) for each GPR model. These values can be seen in Table 1. All models were found to perform with sufficient accuracy.

Table 1. RMSE values for each burnup quantification model

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

A final test was performed to find how the machine learning methodology fared when tasked with attributing the validation samples produced using real irradiations. The performance and a comparison with the maximum likelihood method can be found in Tables 2 and 3. The ML methodology was able to correctly identify the reactor-type for both the HFIR and MURR plutonium samples. The burnup and TSI were quantified with similar success to the maximum likelihood methodology, which was a very positive indication for the ML methodology.

Table 2. Performance comparison for attribution of the HFIR sample

	Burnup Prediction	TSI Prediction
Maximum Likelihood/Measured	0.984	1.146
Machine Learning/Measured	0.963	1.137

Table 3. Performance comparison for attribution of the MURR sample

	Burnup Prediction	TSI Prediction
Maximum Likelihood/Measured	1.038	0.959
Machine Learning/Measured	1.074	0.991

CONCLUSION

The machine learning methodology performed up to the standard set by the maximum likelihood methodology in the attribution of pure samples. The immense potential of machine techniques to determine very difficult to identify relationships within a data set offers a great promise in attributing more complex plutonium samples in the future. The ability of the machine learning methodology to deconvolute the attribution process and focus on each parameter of interest separately allows the data to be used in a more efficient manner and also opens the possibility for the addition of additional parameters for attribution. The next step in this research effort is to produce a new classifier trained with a data set that has been augmented with "spoofed" plutonium samples. Ultimately, this will be the true test of whether the machine learning methodology offers a significant advantage over the maximum likelihood methodology. Another continuation of this work is the production of additional validation samples, via irradiation and separations.

REFERENCES

[1] Chirayath, S.S., Osborn J.M., and Coles, T.M., 2015. Trace Fission Product Ratios for Nuclear Forensics Attribution of Weapons-Grade Plutonium from Fast and Thermal Reactors. Science & Global Security, 23:1, 48-67.

[2] Osborn, J.M., Kitcher, E.D., Burns, J.D., Folden III, C.M., and Chirayath, S.S., 2018. "Nuclear Forensics Methodology for Reactor-Type Attribution of Chemically Separated Plutonium, Nuclear Technology," 201:1, 1-10.

[3] Werner, C.J. (editor), 2017. MCNP® User's Manual: Code Version 6.2, Los Alamos National Laboratory Report No. LA-UR-17-29981.

[4] Osborn, J.M., Glennon, K. J., Kitcher, E.D., Burns, J.D., Folden III, C.M., and Chirayath, S.S., 2019. Experimental validation of a nuclear forensics methodology for source reactor-type discrimination of chemically separated plutonium, Nuclear Engineering & Technology," 51:2, 384-393.

[5] Osborn, J.M., Glennon, K. J., Kitcher, E.D., Burns, J.D., Folden III, C.M., and Chirayath, S.S., 2019. Computational and experimental forensics characterization of weapons-grade plutonium produced in a thermal neutron environment, Nuclear Engineering & Technology," 50:6, 820-828.

[6] Mathworks, 2019. Statistics and Machine Learning Toolbox 2019b.