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Experimental Validation of Nuclear Forensics Methodologies for Reactor-type Attribution, Burnup Determination, and Time Since Irradiation Estimation.

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ABSTRACT

Nuclear forensics (NF) is a critical field of nuclear security and nonproliferation that must continually grow and improve to combat potential threats. Two NF methodologies developed at Texas A&M University (TAMU), Maximum Likelihood Methodology (MLM) and Machine Learning Technique (MLT), are capable of determining three key parameters of interdicted irradiated nuclear material. The three parameters are reactor-type, fuel burnup, and the time since fuel irradiation was completed (TSI). The current database at TAMU for the NF methodologies contain information from eight reactor types: pressured water reactor (PWR); pressurized heavy water reactor (PHWR); fast breeder reactor (FBR); fast flux test facility (FFTF); Canadian national research experimental (NRX); MAGNOX; high flux isotope research reactor (HFIR); and the University of Missouri Research Reactor (MURR). Additionally, data for each reactor spans burnup values up to 5 GWd/MTU (to simulate weapons-grade plutonium) and radioactive decay calculations up to 5000 days post-irradiation. All the data points for these parameters were generated by creating Monte Carlo N-Particle (MCNP) radiation transport models of these nuclear reactors and by performing fuel burnup simulations. To determine the above three parameters of interest, the MLM and MLT methodologies require intra-element isotopic ratios of plutonium and fission products, namely: $^{137}/^{133}$Cs, $^{134}/^{137}$Cs, $^{135}/^{137}$Cs, $^{154}/^{153}$Eu, $^{150}/^{149}$Sm, $^{152}/^{149}$Sm, $^{240}/^{239}$Pu, and $^{241}/^{239}$Pu. Mass spectrometry is the best tool to measure the required isotopic fractions and, in this study, inductively coupled plasma mass spectrometry (ICP-MS) was used. The NF (MLM and MLT) methodologies had previously been validated using experimental data from post-irradiation examinations (PIE). One PIE was completed for depleted UO_2 (DUO_2) irradiated in HFIR to ~5 GWd/MTU and another for natural UO_2 ($NatUO_2$) irradiated in MURR to ~1 GWd/MTU. A third validation dataset for low enriched uranium dioxide ($LEUO_2$) has been done for the work presented here. The results showed that both methodologies can accurately predict reactor-type and burnup. Additional steps such as adding another fission product ratio was required to improve the accuracy of the TSI calculations. The results support that MLM and MLT methodologies are powerful and beneficial tools in the NF repertoire.

1. INTRODUCTION

The work presented here is a continuation of several other research publications from Texas A&M University (TAMU). The first being Osborn et al. who developed the MLM for reactor-type discrimination, burnup quantification, and TSI prediction (Osborn et al., 2018). The second is O'Neal et al. who developed the MLT which was a successor to MLM (O'Neal et al., 2022). Third, the "unknown" $LEUO_2$ sample which was used for the experimental validation of this work comes from Martinson et al. (Martinson et al., 2023).

The premise of the first two studies was to develop nuclear forensics tools to be used on separated plutonium, which may be interdicted. These tools can determine the three key parameters reactor-type, fuel burnup, and time since fuel irradiation was completed (TSI) by a measurement of intraelements ratios of separated plutonium nuclides and other nuclide contaminants present in plutonium. Both MLM and MLT nuclear forensics (NF) methodologies have been validated against two sets of experimental measurements completed at TAMU. The first measurements came from DUO_2 which had been irradiated in the HFIR to approximately 5 GWd/MTU in a pseudo-fast neutron spectrum (Swinney et al., 2017). The second measurements came from natural UO_2 which had been irradiated in the MURR to approximately 1 GWd/MTU in a thermal neutron spectrum. A logical progression was to make a third measurement would be $LEUO_2$. In 2023, Martinson et al. published a study to test and validate an MCNP model of $LEUO_2$ (3.44% enriched) irradiated in the MURR. The focus of the work presented here is to validate the two nuclear forensics methodologies further by incorporating measurements from the same irradiated $LEUO_2$ used by Martinson et al.

Section 2 covers the methods of the work done in this study. Section 3 discusses and shares the results. Section 4 contains the conclusions of the study.

2. METHODOLOGY

The methodology is described in two Sections 2.1 and 2.2. Section 2.1 briefly discusses the developments of both MLM and MLT methodologies. Section 2.2 covers the development of the MCNP model for the $LEUO_2$ irradiation and the experimental data acquisition.

2.1. Maximum Likelihood Methodology (MLM) and Machine Learning Technique (MLT) Development.

The first NF methodology developed at TAMU was based on maximum likelihood probability densities and was developed by Osborn et al. The method could take intra-element ratios of specific nuclides of fission products and plutonium to predict three key parameters of irradiated nuclear material all at once. Namely, reactor-type, burnup, and TSI. These ratios are $^{137}/^{133}$Cs, $^{134}/^{137}$Cs, $^{135}/^{137}$Cs, $^{154}/^{153}$Eu, $^{136}/^{138}$Ba, $^{150}/^{149}$Sm, $^{152}/^{149}$Sm, $^{240}/^{239}$Pu, $^{241}/^{239}$Pu, and $^{242}/^{239}$Pu. These isotopes were chosen for the following reasons: long-lived half-lives or stable, measurable through gamma spectroscopy, can be produced in significant concentrations at low-burnup, and can ratios differ from one reactor to another. The MLM essentially will predict the values from its database which most likely match the unknown sample. Therefore, this method requires a large volume of data to cover many cases. In fact, the database was created by simulating fuel irradiations with MCNP (Werner, 2017). The parameters of the dataset ranged from fuel burnup ranging from 0 to 5 GWd/MTU and TSI ranging from 0 to 5000 days for the following reactor-types: Pressurized Water Reactor (PWR), Pressurized Heavy Water Reactor (PHWR), Fast Breeder Reactor (FBR), Fast Flux Test Facility (FFTF), Canadian National Research eXperimental (NRX); MAGNOX; high flux isotope research reactor (HFIR); and the University of Missouri Research Reactor (MURR). Each reactor-type has a matrix of isotope ratio sets corresponding to 100 burnup values x 5000 TSI values, which equates to 500,000 isotope ratio sets per reactor type. In total, over 3 million data points make up the MLM prediction space.

The second NF methodology, MLT diverges from the first by using supervised machine learning. Supervised machine learning is a subset of data science which uses labeled data and algorithms to train models for future predictions. A major advantage of using machine learning models over a maximum likelihood calculation is that once a model is trained, predictions can be made rapidly. Therefore, the time-consuming portion of machine learning is training and fine-tuning models through testing. Additionally, in this application, machine learning has another advantage over maximum likelihood in that machine learning is predicting or calculating the three key parameters separately, rather than simultaneously. The MLT uses a Support Vector Machines (SVM) classifier for reactor-type discrimination, and a Gaussian Process Regression model to calculate burnup. TSI is calculated analytically once reactor-type and burnup have been determined. O'Neal et al. also found that some of the nuclides used by Osborn et al. could be removed from training sets while retaining accuracy leading to the following six ratios $^{135}Cs/^{137}Cs$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, and $^{241}Pu/^{239}Pu$ (O'Neal et al, 2022). Finally, both methods have been experimentally validated using previous irradiated nuclear material measurements from Swinney et al. and Osborn et al. (Swinney et al., 2017; Osborn et al., 2019).

2.2. MCNP Model Development and Experimental Data Acquisition

A full description of the sample history and preparation can be found in Martinson et al. An abridged description is presented in this work (Martinson et al., 2023). In 2021, TAMU prepared three $LEUO_2$ samples between 13.7-18.1 g and 3.44% enrichment. One sample remained at TAMU while the others were sent to the MURR to be irradiated to approximately 1 GWd/MTU in a thermal neutron flux. This corresponds to about 25 days of full-power effective days within the reactor. The samples then cooled for three months before being sent back to TAMU in November 2021. Once a full irradiation history was provided from MURR, an MCNP model of the irradiation was generated. The isotope production and burnup calculations from the MCNP model were then used for a validation study using experimentally measured values. Figure 1 shows a visualization of the MCNP model of the MURR core with the location of the irradiated samples with respect to the center. The MCNP model was 1/8 section of the annular MURR core to conserve computational resources.

The database that was used in the original MLM implementation was generated by MCNP core fuel depletion models; however, the MURR simulation contained natural uranium. The experimental data from Martinson et al. used irradiated $LEUO_2$ at 3.44%. This means that the database had to be expanded before predictions could be done with the $LEUO_2$ measurements. This new MCNP model extended the burnup of the experimental irradiation of $LEUO_2$ fuel material described above to 5 GWd/MTU. Decay calculations were done analytically outside of MCNP.

One of the two irradiated samples was stored while the other was used for measurements and analysis. The first analysis was gamma spectroscopy using a high-purity germanium detector (HPGe). Afterwards, the sample was dissolved in 8M nitric acid and then an additional gamma spectrum was taken. Next the Pu was separated from the fission products and U using PUREX. Eventually, mass spectrometry samples were prepared and analyzed; however, the results showed that the samples were too diluted to be accurately measured. Another set of mass spectrometry samples were prepared in February 2022 (TSI = 139 d) and measured. An inductively coupled plasma mass spectrometry (ICP-MS) was used. ICP-MS measurements can be used to calculate burnup and TSI, but can also predict reactor-type when coupled with MLM or MLT.

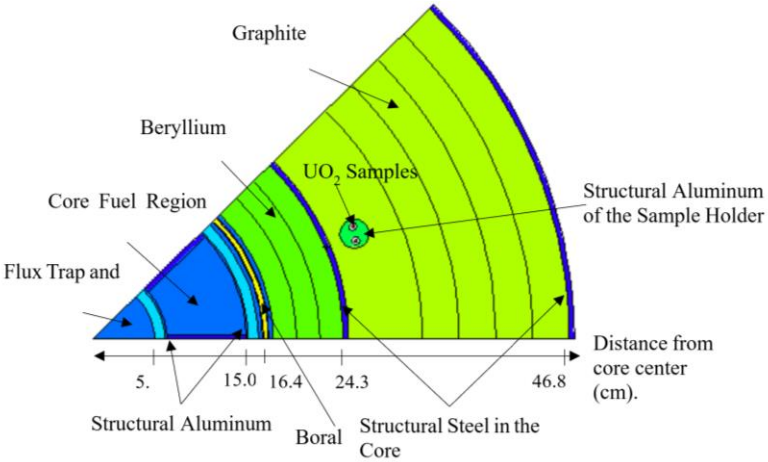

Figure 1. 1/8th model of MURR core with $LEUO_2$ sample and distances from the core center are labelled.[1]

There were high uncertainties for the following nuclides: ^{134}Cs, ^{136}Ba, and ^{242}Pu. For ^{242}Pu the low concentration was the reason for the poor uncertainty. On the other hand, it was not clear for nearly a year for the reasons for the uncertainties in ^{134}Cs and ^{136}Ba. The culprit was discovered to be Xe gas mixed with the Ar carrier gas. It was assumed that the Ar gas was the sole constituent of the gas, and therefore Xe contamination was never considered. ^{134}Cs plays a vital role in TSI predictions, and therefore the large discrepancy led to incorrect predictions. Xe has two stable isotopes (134 and 136) which are isobars to ^{134}Cs and ^{136}Ba. Therefore, these values cannot be accurately measured without using ultrapure Ar gas free of Xe. In December 2022 (TSI = 449 d), a third round of ICP-MS measurements was completed. To address the TSI issue another ratio, ^{144}Ce/^{140}Ce, was measured. Table 1 shows the ICP-MS measurements for the two TSI dates. ^{140}Ce is a stable fission product, while ^{144}Ce has a half-life of 285 days which means that this ratio can be used for TSI predictions up to seven years.

Table 1. Intra-element ratios of $LEUO_2$ material irradiated in the MURR.

Isotope Ratio	TSI = 139 d Measurement		TSI = 449 d Measurement	
	Measured Ratio	Error (%)	Measured Ratio	Error (%)
$^{137/133}$Cs	0.944	3.0	0.890	3.5
$^{134/137}$Cs	1.39×10^{-4}	5.5	5.42×10^{-2}	3.8
$^{135/137}$Cs	0.285	3.5	0.327	6.4
$^{154/155}$Eu	8.80×10^{-3}	11.8	7.79×10^{-3}	0.6
$^{150/149}$Sm	1.78	5.6	3.82	0.3
$^{152/149}$Sm	1.03	6.6	1.01	0.2
$^{240/239}$Pu	6.60×10^{-3}	9.4	6.92×10^{-3}	31.5
$^{241/239}$Pu	1.90×10^{-4}	33.9	2.31×10^{-4}	22.1
$^{242/239}$Pu	8.27×10^{-6}	163.7	1.47×10^{-5}	21.6
$^{144/140}$Ce	Not measured		0.292	1.4

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[1] Reprinted with permission from Martinson et al. "Nondestructive and destructive assay for forensics characterization of weapons-grade plutonium produced in LEU irradiated in a thermal neutron spectrum." *Annals of Nuclear Energy* 183 (2023): 109645

3. RESULTS AND DISCUSSION

The results have been split into the following groups: MLT reactor classification, MLM reactor classification, MLT burnup prediction, and MLM burnup and TSI prediction. Only the TSI=449 d predictions for MLM and MLT are included. Table 2 shows the MLT reactor classification scores for TSI=449. The highest score (bolded) indicates the reactor-type that the model predicts. There were three models used with different isotopic ratios. First, all six isotope ratios (^{135}Cs/^{137}Cs, ^{150}Sm/^{149}Sm, ^{152}Sm/^{149}Sm, ^{154}Eu/^{153}Eu, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu). Second, excluding ^{154}Eu/^{153}Eu. And third, excluding ^{241}Pu/^{239}Pu. Results show that MLT accurately predicts MURR with 3.44% enriched material as the reactor-type of the validation data.

Table 2. Reactor-type classification with the TSI=449 d measurement data using the MLT.

Reactor-Type	PWR	PHWR	FBR	HFIR	MURR (Natural)	MURR (3.44%)
Full Ratio Set	-0.143	-0.296	-0.342	-0.276	-0.758	**0**
Excluding $^{154/155}$Eu	-0.164	-0.336	-0.344	-0.291	-0484	**0**
Excluding $^{241/239}$Pu	-0.418	-0.316	-0.703	-0.321	-0.635	**0**

As mentioned in Section 2.1, MLM predicts all three parameters simultaneously which means that some ratios that differ from the source database can lead to significantly large deviations. Therefore, different combinations that exclude ratios can improve MLM predictions. Table 3 has log-likelihood values of MLM reactor-type predictions for predictions of TSI=449 d measurements. It is evident that ^{242}Pu/^{239}Pu and ^{134}Cs/^{137}Cs lead to poor predictions likely due to the low concentration of ^{242}Pu and high uncertainty of ^{134}Cs/^{137}Cs. Excluding the ratios was sufficient to correctly predict MURR-3.44.The final combination in the inclusion of ^{144}Ce/^{140}Ce. In this model, the ^{134}Cs/^{137}Cs and ^{242}Pu/^{239}Pu ratios are also excluded. The table shows that the inclusion of ^{144}Ce/^{140}Ce did not affect reactor-type prediction as its major role is for TSI quantification.

Table 3. MLM reactor-type predictions using the TSI=449 d measurement data.

Reactor-Type	PWR	PHWR	FBR	HFIR	MURR (Natural)	MURR (3.44%)
Full Ratio Set	-239.9	-231	-21391	**-175**	-177	-1321
Excluding $^{134/137}$Cs, $^{242/239}$Pu	-6.40	-16.3	-21336	-55.9	11.7	**13.3**
Including $^{144/140}$Ce	-19.3	-38.0	-21344		9.85	**11.6**

Similar to reactor-type prediction, several models were trained for burnup prediction using combinations of ratios. Table 4 shows burnup predictions and uncertainties for TSI=449 d measurements. The experimentally determined burnup from Martinson et al. was 0.944 GWd/MTU. There three combinations are: first, full set; second, excluding $^{134/137}$Cs; and third, excluding $^{134/137}$Cs, but including $^{144/140}$Ce. The best burnup prediction came from the third model which was about two standard deviations from the expected value. The ^{144}Ce/^{140}Ce proved effective in estimating TSI where $^{134/137}$Cs could not be used. The TSI result of using the burnup of 1.215 GWd/MTU was accurately predicted to be 448 d. Therefore, this indicates that ^{144}Ce/^{140}Ce is a strong candidate to be included in the machine learning methodology, given additional testing.

Table 4. MLT quantification of the fuel burnup for the TSI=449 d MURR-3.44 measurements.

	TSI = 449 d Measurement	
	Burnup (GWd/MTU)	σ (GWd/MTU)
Full Ratio Set	1.801	0.204
Excluding $^{134/137}$Cs	1.247	0.106
Including $^{144/140}$Ce	1.215	0.111

Table 5 contains the MLM burnup and TSI predictions for TSI=449 d measurements. The predictions correspond to the reactors with the highest likelihood values for the given model. Again, for the results, there are several models with varying ratios. The results show that all of most of the burnup predictions deviate by more than 20%. However, the TSI predictions improved greatly when $^{144/140}$Ce was included.

Table 5. Fuel Burnup and TSI predictions of the MLM for the TSI=449 d MURR-3.44 measurements.

	TSI = 449 d Measurement		
	Predicted Reactor	Burnup (GWd/MTU)	TSI (days)
Full Ratio Set	HFIR	4.04	0
Excluding $^{134/137}$Cs, $^{242/239}$Pu	MURR (3.44%)	1.35	2379
Including $^{144/140}$Ce	MURR (3.44%)	1.24	468

4. CONCLUSIONS

NF is a critical field that demands innovation. This work demonstrated the benefits and shortcomings of two methodologies, MLM and MLT. The experimental results simulated real world irradiated nuclear fuel that could be interdicted and analyzed. This study showed that both of these methodologies can predict the source reactor of a LEUO$_2$ irradiated material. Furthermore, burnup was also predicted within error for both methodologies. One major improvement was the addition of $^{144/140}$Ce as a chronometer for irradiated material to as a backup if ^{134}Cs cannot be accurately measured. Another finding is that when measurements are known to be poor or inaccurate, both methodologies have shown their ability to predict with less data still correctly.

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