QUANTIFYING THE EFFECT OF MEASUREMENT UNCERTAINTY ON A SEPERATED PLUTONIUM ATTRIBUTION METHODOLOGY

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

Ongoing work at Texas A&M University has produced a nuclear forensics methodology that can attribute a separated plutonium sample's reactor-type, fuel burnup, and time since irradiation (TSI). The attribution of these three parameters is performed using two models trained with machine learning, a classification model for the reactor-type and a regression model for fuel burnup. The TSI is calculated analytically using the predicted reactor-type and fuel burnup. Sets of intra-element isotope ratios are the features used in both models. The training data used in producing the models is sampled from a library of Monte Carlo N-Particle (MCNP) fuel burnup simulations that have been performed for a set of reactors of interest. For the validation of the model performance multiple irradiation campaigns were conducted to produce physical samples that could be attributed. These campaigns involved irradiating uranium samples of varying initial enrichment levels at the High Flux Isotope Reactor (HFIR) and the University of Missouri Research Reactor (MURR). These include a depleted UO_2 sample irradiated at HFIR in a pseudo-fast neutron spectrum, and a natural UO_2 and low enriched UO_2 (3.44 wt-%) sample irradiated at MURR in a thermal neutron spectrum. Subsequently the plutonium produced was separated and the isotopic concentrations determined. The use of simulated data for the model production, and then physical data in the use of the model introduces an unavoidable amount of incongruency, as there will always be differences between these two. Additionally, both sets of data introduce their own sources of uncertainty, and characterizing this uncertainty is important for judging the ultimate capabilities of this attribution methodology. A study was performed to use the validation data's measurement uncertainties to study the effect that variance in the input can have on model predictions. To do this, a set of test data was produced for each validation plutonium sample by sampling each isotope ratio from a normal distribution with the measured mean and variance for that isotope ratio, and then predictions were made with this data set to find how the predictions change with the natural variation in the measured isotope ratios values. By analyzing the extended uncertainty bounds the effect that measurement uncertainty has on the methodology's prediction capability can be visualized.

INTRODUCTION AND METHODOLOGY

A nuclear forensics methodology currently being developed at Texas A&M University uses machine learning models to attribute a separated plutonium sample's reactor-type, fuel burnup, and time since irradiation (TSI) [1]. The forensics signature used in the methodology are a set of seven intra-element isotope ratios: $^{134}Cs/^{137}Cs$, $^{135}Cs/^{137}Cs$, $^{150}Sm/^{149}Sm$, $^{152}Sm/^{149}Sm$, $^{154}Eu/^{153}Eu$, $^{240}Pu/^{239}Pu$, and $^{241}Pu/^{239}Pu$. To train the machine learning models used for the attribution, a dataset is produced by randomly sampling from a library of separated Pu isotopic compositions for various reactor-types. This library is comprised of simulated data produced by performing fuel burnup

simulations of six different reactor-types using MCNP [2]. The parameter space of interest is from 0 to 5 GWd/MTU for fuel burnup and 0 to 5,000 days for TSI, and all training data is sampled within those ranges. The size of the training dataset used is 3,000 isotope ratio sets, approximately 500 sets for each of the reactor-types of interest. The six reactor-types of interest include three generic power reactors, a pressurized water reactor (PWR), a pressurized heavy water reactor (PHWR), and a fast breeder reactor (FBR), as well as irradiation positions at two research reactors, High Flux Isotope Reactor (HFIR) and University of Missouri Research Reactor (MURR). There are two different reactor-type classes included in the reactor classifier training dataset that use MURR, which are differentiated by their starting ^{235}U enrichment. These two classes will be denoted as MURR-natural and MURR-3.44%.

The attribution of an unknown sample using our methodology takes part in three steps: first a classifier trained with ^{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 ratios is used to find the reactor-type of the sample. The classifier is a support vector machine (SVM) with a cubic kernel. Using the predicted reactor-type the fuel burnup is quantified using a gaussian process regression (GPR) model trained with an exponential kernel. A GPR model is trained for each reactor-type class and the ratios used are ^{134}Cs/^{137}Cs, ^{150}Sm/^{149}Sm, ^{152}Sm/^{149}Sm, ^{154}Eu/^{153}Eu, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu. Last the predicted reactor-type and fuel burnup are used along with the ^{134}Cs/^{137}Cs ratio to perform an analytic calculation to determine TSI.

The nuclear forensics methodology has been validated using data from plutonium samples produced by irradiating uranium samples in research reactors. The Pu was separated and the isotopic composition was quantified using gamma, alpha, and mass spectroscopy. The first validation sample produced was from an irradiation campaign at HFIR [3] [4]. The sample irradiated was depleted UO_2 sheathed in gadolinium, to emulate a fast neutron flux spectrum. During the course of irradiation, the gadolinium was depleted, and the sample was exposed to the thermal neutron flux spectrum. The sample reached approximately 4 GWd/MTU fuel burnup. The second validation sample produced was a natural uranium sample irradiated in a thermal neutron flux spectrum at MURR [5]. The sample reached approximately 1 GWd/MTU fuel burnup. During quantification, the ^{154}Eu/^{155}Eu was unable to be determined. The last validation sample was produced by irradiating $LEUO_2$ with an enrichment of 3.44 wt-% in a thermal neutron flux spectrum at MURR [6]. The sample reached a fuel burnup of approximately 1 GWd/MTU. During quantification of the sample, it was found that there were isobaric interference that was leading to difficulties measuring the ^{134}Cs concentration in the sample. This interference was due to Xe contamination of the Ar carrier gas used in the mass spectrometer. The ^{134}Cs/^{137}Cs ratio is typically used for both fuel burnup quantification and TSI, where it is the sole ratio used. A substitute ratio, ^{144}Ce/^{140}Ce, was identified and measured in the sample. In the results for the MURR-3.44% sample, the fuel burnup model is trained with ^{150}Sm/^{149}Sm, ^{152}Sm/^{149}Sm, ^{240}Pu/^{239}Pu, and ^{241}Pu/^{239}Pu, and the ^{144}Ce/^{140}Ce was used for the TSI calculation.

To study the effect of measurement uncertainty on the model predictions, a new test dataset was produced for each validation sample. Each testing dataset included 10,000 isotope ratio sets. The isotope ratios in this dataset were produced by independently sampling each isotope ratio from a normal distribution defined by a mean corresponding to the measured isotope ratio value of the validation sample being investigated and a standard deviation corresponding to the measurement uncertainty. This means that each testing datasets includes 10,000 isotope ratio sets that could have been feasibly measured from the validation sample of interest. These three testing datasets were then used to make reactor-type, fuel burnup, and TSI predictions, which produces a distribution of

the possible predicted values. Each predicted fuel burnup and TSI also had their respective prediction standard deviation added and subtracted to produce an uncertainty band for each prediction. The minimum and maximum fuel burnup value in all the predictions' uncertainty bands were used to produce an extended uncertainty band that is a result of the measurement uncertainty.

RESULTS AND DISCUSSION

The MURR-natural sample has a measured fuel burnup of 0.97 ± 0.03 GWd/MTU and a TSI of 318 d. The original predicted fuel burnup and TSI values using our machine learning method for the MURR-natural sample using the measured isotope ratio values were 1.06 ± 0.06 GWd/MTU and 312 ± 55 d. The MURR-natural class was predicted correctly for 93.5% of the samples in the dataset that includes measurement uncertainty, the other 6.5% of the samples were predicted to be MURR-3.44%. The distribution of fuel burnup predictions and TSI predictions using the dataset that includes measurement uncertainty can be seen in Figure 1 and Figure 2, respectively. It should be noted that these distributions only include fuel burnup and TSI predictions for samples that were correctly identified as MURR-natural. The expanded uncertainty bounds can be seen in the dashed lines. For fuel burnup, the extended uncertainty ranged from 0.87 to 1.25 GWd/MTU, respectively, or each were approximately 18% difference from the originally predicted fuel burnup value. The expanded TSI uncertainty range was 0 to 827 d, which is due to a combination of sensitivity to the predicted fuel burnup value as well as to the fact that TSI is predicted using only one ratio. If the one ratio used for the TSI calculation deviates too far from the simulated database values for that ratio, the prediction will suffer as a result.

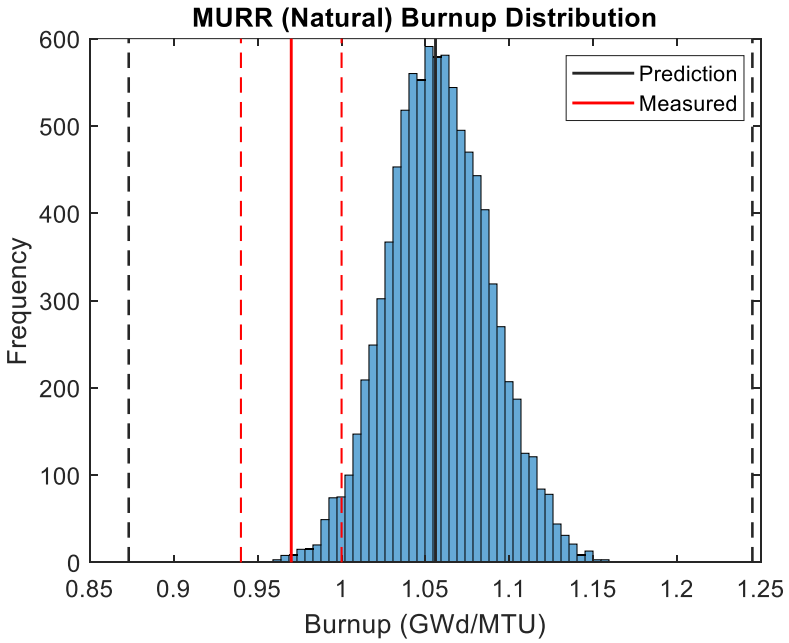

Figure 1. The distribution of fuel burnup predictions and expanded uncertainty (black dashed lines) for the MURR-Natural sample, as well as measured (red) with uncertainty (red dashed) and original predicted (black) values highlighted.

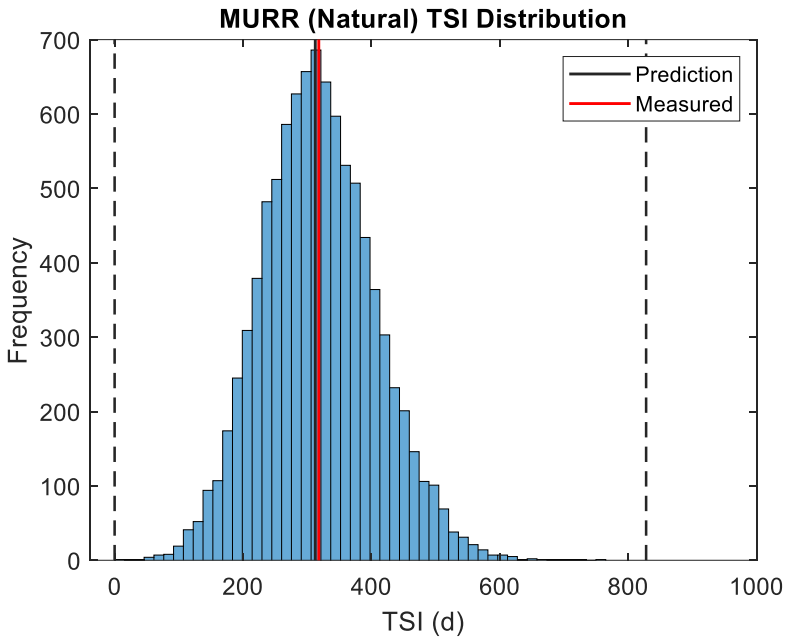

Figure 2. The distribution of TSI predictions and expanded uncertainty (dashed lines) for the MURR-Natural sample, as well as measured (red) and original predicted (black) values highlighted.

The HFIR sample has a measured fuel burnup of 4.36 ± 0.28 GWd/MTU and a TSI of 1601 d. The original predicted fuel burnup and TSI values for the HFIR sample using the measured isotope ratio values were 4.25 ± 0.10 GWd/MTU and 1822 ± 10 d. The HFIR class was predicted correctly for 95.9% of the samples in the dataset that includes measurement uncertainty, the other 4.1% of the samples were predicted to be from the PWR class. The distribution of fuel burnup predictions and TSI predictions using the dataset that includes measurement uncertainty can be seen in Figure 3 and Figure 4, respectively. It should be noted that these distributions only include fuel burnup and TSI predictions for samples that were correctly identified as HFIR. The expanded uncertainty bounds can be seen in the dashed lines. For fuel burnup, the extended uncertainty range ranged from 4.07 to 4.45 GWd/MTU, respectively, or an approximate 4.5% difference from the originally predicted fuel burnup value. The expanded TSI uncertainty range was from 1614 to 2068 d, a 11% and 14% difference, respectively. This TSI uncertainty range for HFIR being much smaller than that of the MURR-natural can most likely be attributed to the lower measurement uncertainty in the $^{134}Cs/^{137}Cs$ ratio for the HFIR data compared to the MURR data. There are other possible explanations for this behavior, including the sample being at a higher TSI, which could lessen the effect the small variation in predicted fuel burnup can have on the TSI prediction; another possible explanation is just better agreement with the simulated values that are used in the TSI calculation.

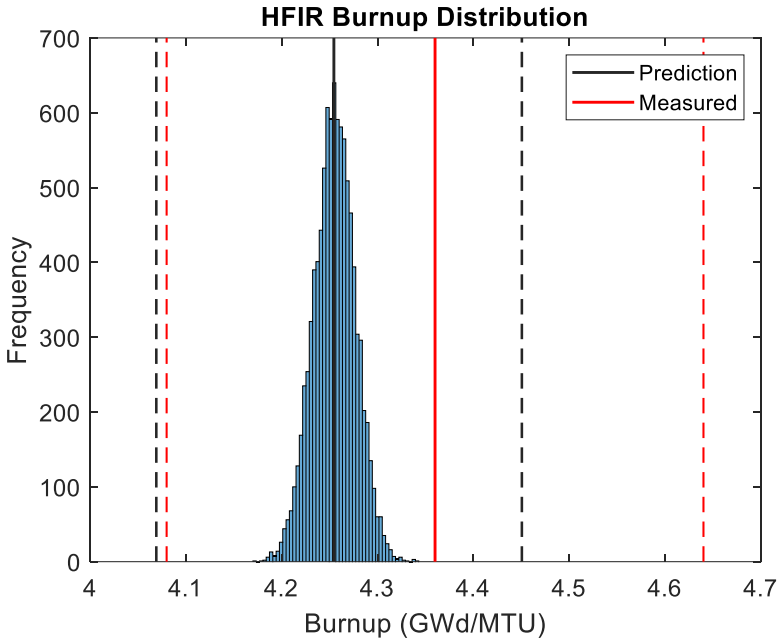

Figure 3. The distribution of fuel burnup predictions and expanded uncertainty (black dashed lines) for the HFIR sample, as well as measured (red) with uncertainty (red dashed) and original predicted (black) values highlighted.

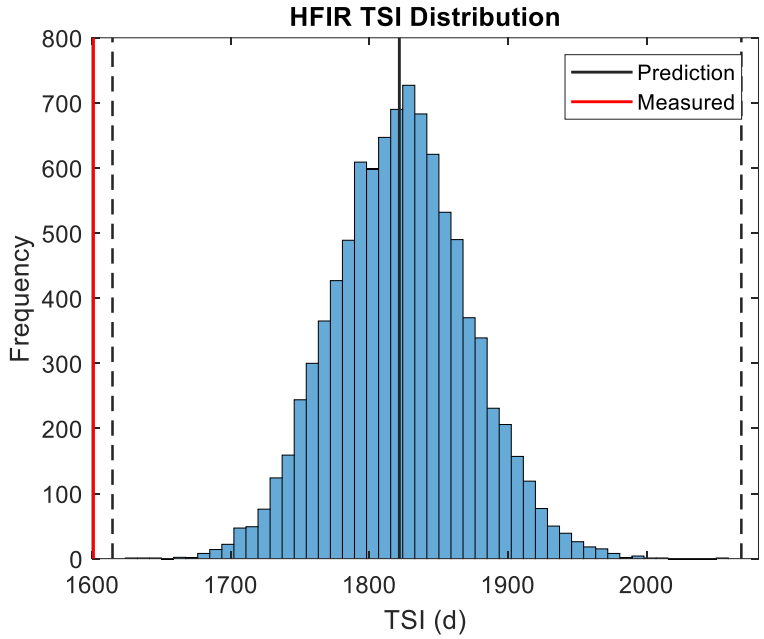

Figure 4. The distribution of TSI predictions and expanded uncertainty (dashed lines) for the HFIR sample, as well as measured (red) and original predicted (black) values highlighted.

The MURR-3.44% sample has a measured fuel burnup of 0.944 ± 0.02 GWd/MTU and a TSI of 449 d. The original predicted fuel burnup and TSI values for the MURR-3.44% sample using the measured isotope ratio values were 1.02 ± 0.08 GWd/MTU and 451 ± 1 d. The MURR-3.44% class was predicted correctly for 81.7% of the samples in the dataset that includes measurement

uncertainty, the other 18.3% of the samples were predicted to be from the MURR-natural class. This higher number of misclassified reactor-type can most likely be attributed to the high uncertainty of the $^{241}Pu/^{239}Pu$ ratio (33.9%), which is a key isotope for distinguishing between the two MURR classes, which only differ in the uranium sample's starting enrichment. This indicates that 33.9% error in a measurement might be high enough to have an adverse effect on model capabilities. The distribution of fuel burnup predictions and TSI predictions using the dataset that includes measurement uncertainty can be seen in Figure 5 and Figure 6, respectively. It should be noted that these distributions only include fuel burnup and TSI predictions for samples that were correctly identified as MURR-3.44%. The expanded uncertainty bounds can be seen in the dashed lines. For fuel burnup this extended uncertainty ranged from 0.79 and 1.3 GWd/MTU, respectively, or a 22.5% and 27.5% difference from the originally predicted fuel burnup value. This higher amount of uncertainty relative to the original prediction is most likely due to the high $^{241}Pu/^{239}Pu$ ratio measurement uncertainty. The expanded TSI uncertainty range was from 430 to 478 d, a 6% and 4.7% difference from the original prediction, respectively. The $^{144}Ce/^{140}Ce$ ratio used for the TSI calculation for this sample is the reason for the relatively small uncertainty range.

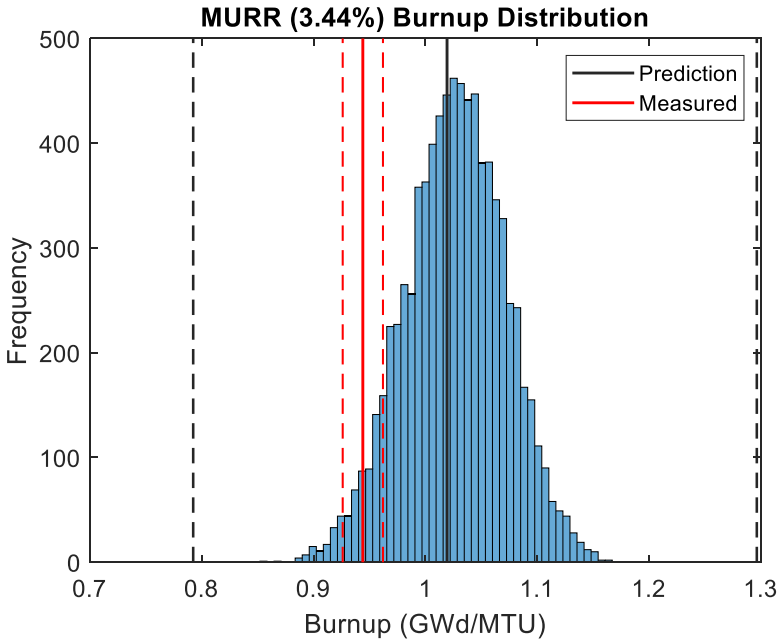

Figure 5. The distribution of fuel burnup predictions and expanded uncertainty (black dashed lines) for the MURR-3.44% sample, as well as measured (red) with uncertainty (red dashed) and original predicted (black) values highlighted.

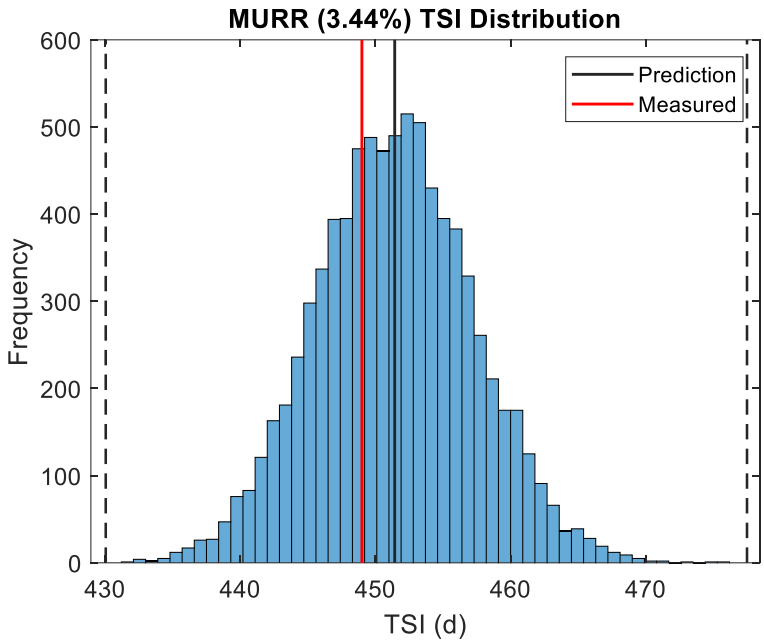

Figure 6. The distribution of TSI predictions and expanded uncertainty (dashed lines) for the MURR-3.44% sample, as well as measured (red) and original predicted (black) values highlighted.

4. CONCLUSIONS

Data from physical measurements will always have some level of uncertainty. It is imperative that those who create methods understand the effects the uncertainties involved will have on their method's capabilities and find potential limits that are dictated by the physical reality of measurement capabilities. It has been found that for the nuclear forensics methodology in question, the measurement uncertainties in the validation sample data understandably impact the model's ability to make accurate predictions, although this deviation was found to be at an acceptable level. It was also found that even when taking a conservative approach to represent the additional uncertainty in the prediction, as a result of the measurement uncertainty in the underlying data, the model can make predictions with sufficient accuracy to be a useful tool in nuclear forensics investigations. The study also gave good indication of the level of fidelity required in measurements that will be used by the nuclear forensics methodology.

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