Advancing Methods for Providing Attribution for the HEU Used in a Terrorist Nuclear Weapon

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Abstract:

An algorithm was developed that uses measured isotopic ratios from fission product residue following the detonation of a high-enriched uranium nuclear weapon to compute the original attributes of the nuclear material used in the device. The specific attributes assessed are the uranium isotopics (considering ^{234}U, ^{235}U, ^{236}U, and ^{238}U) and the type of enrichment process used to create the material (e.g., gaseous diffusion, gas centrifuge, etc.). Using the original material attributes of the weapon significantly increases the probability of identifying the perpetrator of the attack. In this study, research was conducted to perform sensitivity analysis of the calculated values, analyze alternate methods of enrichment, determine the source (uranium mine) from which the feed material was taken and assess potential "spoofing" techniques. The purpose of this research was to verify that the analytical method developed would remain valid for a multitude of conceivable variations that could potentially be used to disguise the origin of the original nuclear material used in the device. It is envisioned that this methodology could serve as a pre-processing step to a more computationally intensive and more accurate system in the event of a nuclear terrorist attack.

Keywords: highly enriched uranium; material attribution; terrorism; post-detonation analysis;

1. Introduction

The development and implementation of safeguards to prevent a nuclear terrorist attack is crucial to improving security throughout the global community. If a terrorist nuclear device was detonated, how quickly could the site be assessed to determine what type of device was detonated, how powerful the device was and where it came from? The detonation of an HEU weapon would cause catastrophic damage and mass casualties. Due to the severity of such an attack, it is critical to be able to compute the original material attributes of a weapon because it significantly increases the probability of identifying perpetrators of the attack.

The objective of the algorithm developed was to utilize post-detonation measured isotopic ratios in order to determine the pre-detonation material attributes within reasonable accuracy. More computationally intensive methods are being developed elsewhere; however, these methods require extensive computational times in order to produce acceptable results. In effort to reduce the computational time required to compute the original material attributes, the method developed here uses an analytical approach which consisted inversions of the burnup and decay equations (all first-order ordinary differential equations). It is envisioned that this methodology could serve as a pre-processor step to a more computationally intensive and more accurate system.

This work is focused on the post-detonation attribution of a Highly Enriched Uranium (HEU) terrorist nuclear weapon. Terrorist devices may differ from military nuclear weapons mainly in the sophistication

applied when constructed (e.g. type and grade of material used and quality of tamper/reflector). Since a gun-type weapon is considerably less complex than an implosion weapon (generally, gun-type weapons are not tested), this is considered to be a likely scenario for a nuclear terrorist attack [1].

Given a measurement of the post-detonation isotopics from fission product residue, the interest in this work was to attempt to determine the following characteristics (in this order of importance): (1) pre-detonation ^{235}U enrichment, (2) pre-detonation $^{234}U/^{238}U$ isotopic ratio, (3) pre-detonation $^{236}U/^{238}U$ isotopic ratio, (4) enrichment method used to produce material, (5) pre-enrichment $^{234}U/^{238}U$ isotopic ratio, (6) pre-enrichment $^{236}U/^{238}U$ isotopic ratio, and (7) source (mine or otherwise) from which feed uranium was taken. It was acknowledged immediately that steps (1)-(3) would have a likely chance of success and the steps (4)-(7) would be significantly more difficult.

2. Methodology

The algorithm developed here consists of two main parts: a forward model and an inverse model. The forward model consisted of simulations to predict post-detonation (actually post-irradiation) isotopics given the original isotopics of the material and the number of fission (or yield) of the device. The data from the forward model was mainly used to test the viability of the inverse model. The inverse model predicted pre-detonation isotopics using analytical inversions of the buildup and decay equations and post-detonation isotopic measurements. The inverse model also included error propagations to allow for prediction of uncertainties in the attributes as well as to determine the sensitivity of the results to the input data.

2.1. Forward model

The forward model simulations were performed using the ORIGEN2 computer code [2]. ORIGEN2 calculates the buildup and depletion of isotopics from irradiation and decay. The code possesses a large set of libraries (each library corresponds to a specific type of reactor) with cross-section, decay, and fission product yield data. ORIGEN2 uses the matrix exponential method to solve a large system of coupled, linear, first-order ordinary differential equations. While not a weapons burn code, ORIGEN contains sufficient capability to allow for analysis of the feasibility of the method developed here.

Four different uranium signatures from gaseous centrifuge and gaseous diffusion enriched uranium, both with and without ^{236}U present in the original material, were simulated. In order to simulate the detonation of a 20 kT HEU weapon in ORIGEN2, the mass of ^{234}U, ^{235}U, ^{236}U (if applicable), and ^{238}U were calculated. Assuming the total mass of uranium equalled one metric ton and was enriched to 95 a/o ^{235}U, values for the enrichment of ^{234}U and ^{236}U were calculated*. Natural uranium contains essentially no ^{236}U (though small quantities are found in natural material due to the activation of ^{235}U from neutron background); however, enriched uranium of U.S. or Russian origin includes a significantly higher abundance of ^{236}U due to the re-enrichment of naval fuel. Thus, the presence of ^{236}U in the original material provides a unique signature indicating the geographic origin of where the uranium was enriched.

Then, the burnup of the initial material in the weapon given a 20 kT yield was simulated using ORIGEN2. Generally, a 2 kT yield is associated with terrorist weapons; however, this value was not used because only 2% of the original material fissions. The task of determining the original material used in the weapon becomes much simpler for low yields because there is only a slight difference between the pre-detonation composition and the post-detonation composition of the weapon. The resultant isotopics produced from this burnup were then decayed for 1.0 day (assumes that it will take approximately 1 day or more to acquire measurements from the post-detonation fission product residue). Assuming that the weapon was detonated on the ground or at a relatively low altitude, ^{89}Sr and ^{95}Zr (characterized by long half-lives, low absorption cross-sections, and the ability to be measured in the environment) were the two fission products used to calculate the total number of fission from the device in the inverse model.

* Equations used in algorithm may be obtained from the original paper (reference 3).

2.2. Inverse model

The inverse model equations are all expressed in terms of atom ratios relative to ^{238}U (the ^{238}U concentration in the device is roughly constant during irradiation). The algorithm [3] implemented in the inverse model uses an iterative procedure to calculate the original material attributes* which consists of the following steps:

1. The pre-detonation $^{235}U/^{238}U$ ratio is set to an initial guess input by the user.
2. A guess was made for the method of enrichment used and whether or not ^{236}U was present in the initial material and the corresponding pre-detonation $^{234}U/^{238}U$ and $^{236}U/^{238}U$ (if applicable) ratios were calculated using eqs. (2.1) - (2.3).
3. The pre-detonation $^{234}U/^{238}U$ and $^{236}U/^{238}U$ (if applicable) ratios were combined with the initial guess for $^{235}U/^{238}U$ to calculate the ^{235}U enrichment of the original material.
4. The number of fissions in the device per unit mass was calculated using the measurement of two fission products: ^{95}Zr and ^{89}Sr.
   - A single fission product could have been used but by using two fission products, iteration between the two yielded a better prediction of the number of fissions.
   - The equation derived for the total number of fissions assumed that the recoverable energy per fission from ^{235}U equalled 200 MeV and that all fissions were from ^{235}U.
5. An updated $^{234}U/^{238}U$ value was calculated using measurements of $^{232}U/^{238}U$ in the residue.
   - It was assumed that no ^{232}U existed in the original material and the measured ^{232}U concentration was produced only from the $^{234}U(n,3n)^{232}U$ reaction.
6. An updated $^{235}U/^{238}U$ value was then calculated using measurements of $^{235}U/^{238}U$ in the residue.
   - It was assumed that the change in ^{235}U was equal to its loss rate from absorption.
7. Then, an updated $^{236}U/^{238}U$ value was calculated using measurements of $^{236}U/^{238}U$ in the residue.
   - This derivation assumed that the change in ^{236}U was equal to its production rate from radiative capture in ^{235}U minus the loss rate from the absorption of ^{236}U.
   - The final equation for the updated $^{236}U/^{238}U$ value was obtained by assuming that the ratio of $^{236}U/^{235}U$ as a function of irradiation time was linear and therefore was easily integrated.
8. A new value for the ^{235}U enrichment was then calculated and steps (4) – (7) were repeated iteratively until the pre-detonation $^{235}U/^{238}U$ ratio converged to a value within a specified tolerance.

3. Uranium Signatures

3.1. Enrichment processes

Weapons-grade HEU is typically enriched to 90 a/o ^{235}U or greater. The method of enrichment provides a useful signature that may indicate where the uranium was enriched. Methods used to enrich uranium include: gaseous centrifuge, gaseous diffusion, electromagnetic isotope separation (EMIS), and atomic vapor laser isotope separation (AVLIS) [1]. The two most common enrichment processes used throughout the world are gaseous centrifuge and gaseous diffusion both of which separate the uranium isotopes in a gaseous compound called uranium hexafluoride.

In this study, the algorithm developed analyzed only gaseous centrifuge and gaseous diffusion enrichment methods. These methods are hard to distinguish because they both rely on the differences in mass between ^{235}U containing molecules and ^{238}U containing molecules, though they are based on different physical processes. This results in small separation factors of 1.162 and 1.00429 for gaseous centrifuge and gaseous diffusion, respectively, due to the higher concentration of ^{234}U contamination in the enriched product [4]. It is expected that distinguishing most other methods (such as AVLIS or EMIS) would be much simpler. For instance, the AVLIS process enriches uranium using lasers tuned to a precise frequency so that only the ^{235}U atoms absorb the light. The resulting separation factor is nearly infinite and yields almost no ^{234}U in the enriched product.

In order to determine valid signatures indicating the method of enrichment, the values calculated in the inverse model for post-detonation ^{234}U concentrations were compared. For 95 a/o ^{235}U centrifuge enriched

fuel, the calculated $^{234}U/^{238}U$ ratio was approximately 5.0 times greater than the calculated $^{234}U/^{238}U$ ratio for 95 a/o ^{235}U diffusion enriched fuel. These significant variations in ^{234}U are presented in Table 1 and were used as signatures indicating the enrichment process used.

Enrichment Process		Pre-detonation Value $(N^{234}/N^{238})_0$	Inverse Model $(N^{234}/N^{238})_0$	Percent Difference
Centrifuge	with ^{236}U	1.04	1.06 ± 0.015	1.66%
	without ^{236}U	0.869	0.883 ± 0.013	1.69%
Diffusion	with ^{236}U	0.200	0.204 ± 0.003	2.20%
	without ^{236}U	0.179	0.182 ± 0.005	1.31%

Table 1: Comparison of calculated $^{234}U/^{238}U$ ratios to distinguish centrifuge enriched fuel from diffusion enriched fuel.

3.2. Presence of ^{236}U

After the enrichment process has been determined, whether or not ^{236}U existed in original weapons material must be established. The $^{236}U/^{238}U$ inverse model values computed for gaseous diffusion and gaseous centrifuge enriched uranium, both with and without ^{236}U present, are presented in Table 2. For enriched fuel with ^{236}U present in the original material, the calculated $^{236}U/^{238}U$ value was approximately 4.5 times greater than the $^{236}U/^{238}U$ value for enriched fuel without ^{236}U present in original material.

Enrichment Process		Pre-detonation Value $(N^{236}/N^{238})_0$	Inverse Model $(N^{236}/N^{238})_0$	Percent Difference
Centrifuge	with ^{236}U	0.195	0.204 ± 0.011	4.58%
	without ^{236}U	0.0	0.005 ± 0.008	-
Diffusion	with ^{236}U	0.115	0.121 ± 0.007	5.57%
	without ^{236}U	0.0	0.027 ± 0.003	-

Table 2: Comparison of calculated $^{236}U/^{238}U$ values to determine whether or not ^{236}U was present in original material.

4. Sensitivity Analysis

The methodology developed was tested for a 20 kT detonation of a 95 a/o ^{235}U enriched HEU device. The "measured values" were produced from ORIGEN simulations for four different uranium signatures from gaseous centrifuge and gaseous diffusion enriched uranium, both with and without ^{236}U present in the original material. Error propagations were done by hand to predict uncertainties in the attributes as well as to determine the sensitivity of these results to errors in the input data.

4.1. Sensitivity of initial guess for ^{235}U concentration

The algorithm was insensitive to the initial guess for ^{235}U concentration. In all cases less than 10 iterations (less than 1 second computational time) were used to acquire a result. The results presented in Table 3 verified that for any positive initial guess of any order of magnitude input into the algorithm will be iterated to a reasonably correct answer.

Enrichment Process	Initial Guess $(N^{235}/N^{238})_0$	Pre-detonation Value $(N^{235}/N^{238})_0$	Inverse Model $(N^{235}/N^{238})_0$	Percent Error
Centrifuge (with ^{236}U)	1.00×10^{10}	42.4	43.1 ± 0.431	1.61%
Diffusion (no ^{236}U)	1.00×10^{-10}	22.4	22.6 ± 0.225	0.66%

Table 3: Comparison of calculated $^{235}U/^{238}U$ values from inverse model to actual values for various initial guesses.

4.2. Sensitivity of error in calculated ^{234}U attribute

Error propagations were used to derive an equation for the error in the $^{234}U/^{238}U$ attribute in terms of the errors in the input parameters (this method was repeated for $^{235}U/^{238}U$ and the $^{236}U/^{238}U$ attributes). Using this equation, the sensitivity of the error in the $^{234}U/^{238}U$ attribute was determined by plotting the error in the calculated $^{234}U/^{238}U$ value as a function of the error in the measured ^{232}U value and the $^{234}U(n, 3n)$ microscopic cross-section. The plot depicted in Fig. 1 shows that the calculated error in the $^{234}U/^{238}U$ value varies linearly as a function of the error in the measured ^{232}U value and the error in the $^{234}U(n, 3n)$ microscopic cross-section. The linear relationship determined is important because it indicates that error in the measured ^{232}U value and the error in the $^{234}U(n, 3n)$ microscopic cross-section equally contribute to overall error in the calculated the $^{234}U/^{238}U$ value. This relationship may also be utilized to determine the point at which reducing these errors no longer reduces the overall error in the calculated the $^{234}U/^{238}U$ attribute.

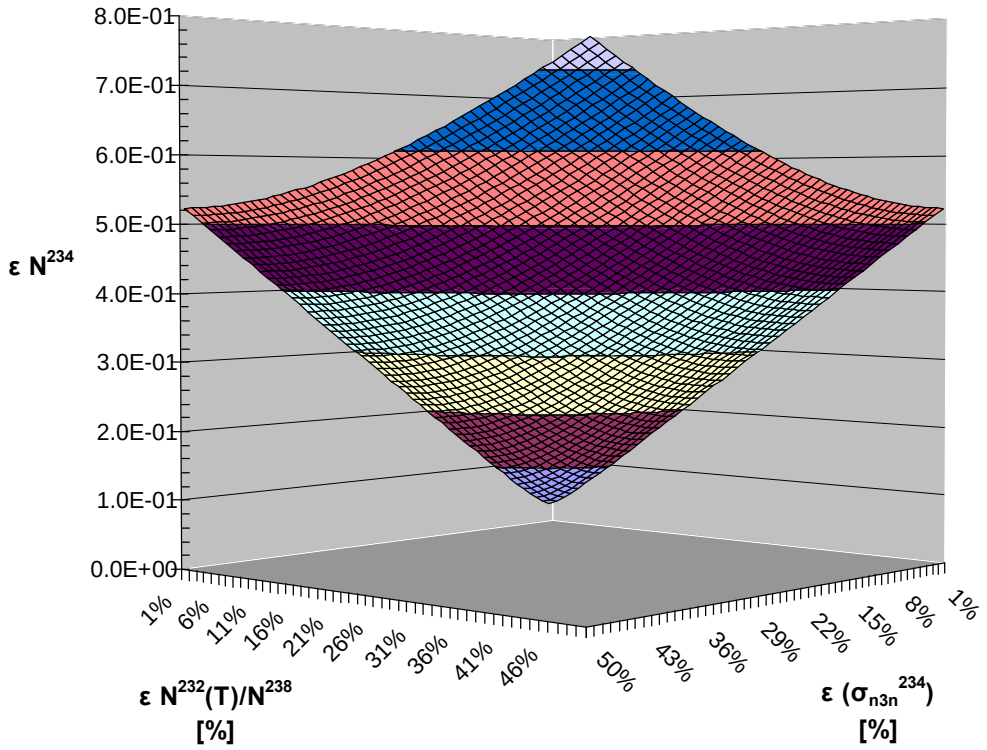

Figure 1. Error in the calculated $^{234}U/^{238}U$ value as a function of the error in the measured ^{232}U value and the $^{234}U(n, 3n)$ microscopic cross-section.

4.3. Sensitivity of error in calculated ^{235}U attribute

The error in the calculated $^{235}U/^{238}U$ attribute as a function of the error in the ^{235}U enrichment and the error in the ^{235}U microscopic fission cross-section to determine the sensitivity of the error in this attribute. The plot depicted in Fig. 2 shows that the calculated error in the $^{235}U/^{238}U$ value varies linearly as a function of the error in the ^{235}U microscopic fission cross-section and varies nonlinearly as a function of the error in the ^{235}U enrichment. The nonlinear relationship determined indicates that error in the ^{235}U enrichment contributes more towards the overall error in the calculated the $^{235}U/^{238}U$ value than the error in the ^{235}U microscopic fission cross-section does. Therefore, more effort should be spent reducing the error in the value for the ^{235}U enrichment than reducing the error in the ^{235}U microscopic fission cross-section.

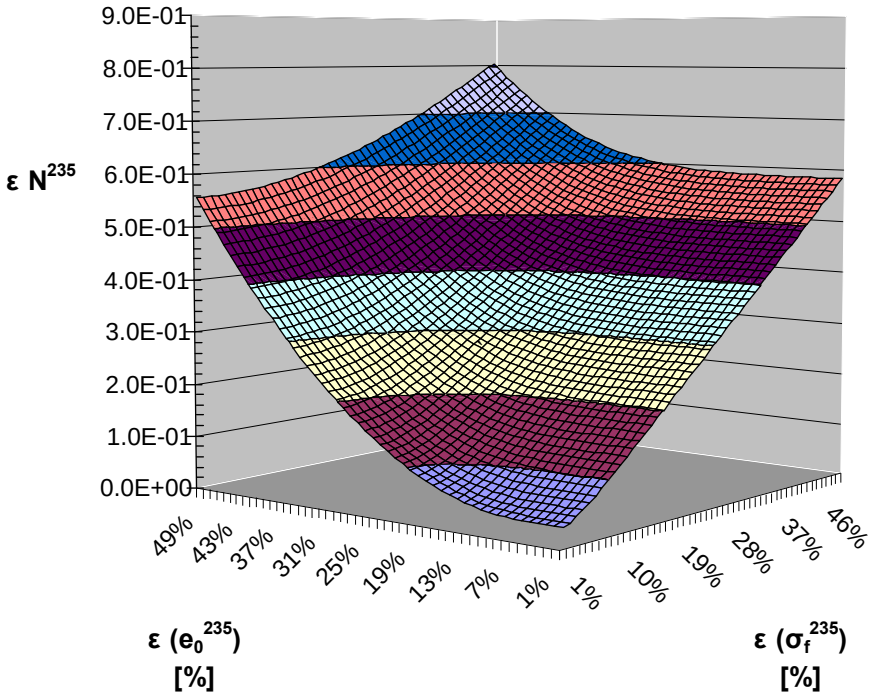

Figure 2. Error in the calculated $^{235}U/^{238}U$ value as a function of the error in the ^{235}U enrichment and the ^{235}U microscopic fission cross-section.

4.4. Sensitivity of error in calculated ^{236}U attribute

The sensitivity of the error in the $^{236}U/^{238}U$ value was determined by plotting the error in the calculated $^{236}U/^{238}U$ value as a function of the errors in the ^{236}U and the ^{235}U microscopic absorption cross-sections. The plot depicted in Fig. 3 shows that the calculated error in the $^{236}U/^{238}U$ value varies linearly as a function of the errors in the ^{236}U and the ^{235}U microscopic absorption cross-sections. The linear relationship determined indicates that the error in the ^{235}U microscopic absorption cross-section affects the overall error in the calculated the $^{236}U/^{238}U$ value more than the error in the ^{236}U microscopic absorption cross-section. This is because increasing the error in the ^{235}U microscopic absorption cross-section increases the overall error in the calculated the $^{236}U/^{238}U$ value significantly more than increasing the error in the ^{236}U microscopic absorption cross-section does. Therefore, more effort should be spent reducing the error in the ^{235}U microscopic absorption cross-section than reducing the error in the ^{236}U microscopic absorption cross-section.

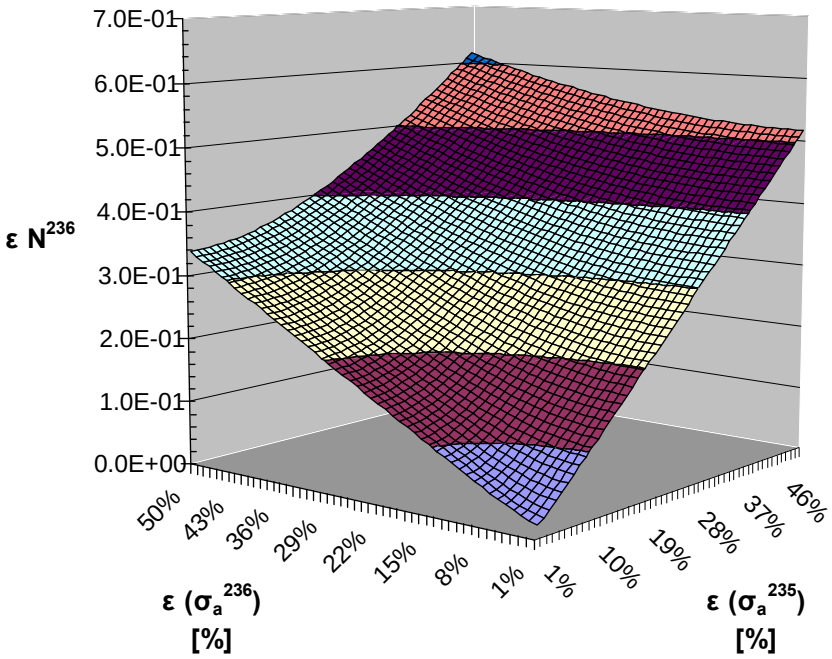

Figure 3. Error in the calculated $^{236}U/^{238}U$ value as a function of the errors in the ^{236}U and the ^{235}U microscopic absorption cross-sections.

5. ^{234}U Isotopics in Mines

After determining the enrichment process and the pre-detonation uranium isotopic ratios, this information may then be used to calculate the pre-enrichment $^{234}U/^{238}U$ isotopic ratio. Since different uranium mines throughout the world are characterized by different isotopic abundances of ^{234}U, the source (mine or otherwise) from which feed uranium was taken can be determined by comparing the calculated pre-enrichment $^{234}U/^{238}U$ isotopic ratio to a set of measured $^{234}U/^{238}U$ ratios taken from various mining or milling facilities throughout the world. Thus, the natural variation of ^{234}U throughout the world provides a unique signature indicating the geographic origin of the material.

^{234}U has a relatively short half-life and exists in secular equilibrium with ^{238}U. Thus, the ratio of ^{234}U to ^{238}U should equal to the ratio of the half-lives (55.0 ppm). Variations in the ratio of $^{234}U/^{238}U$ may result from processes that disrupt the decay chain of ^{238}U to ^{234}U [5]. All of the measured $^{234}U/^{238}U$ values shown in Table 4 were determined using thermal ionization mass spectrometry where the $^{235}U+$ ion beam intensity was adjusted to correct for mass discrimination using the measured $^{235}U/^{238}U$ ratio obtained by gas source mass spectrometry.

Sample No.	Country of Origin	Milling Facility	$^{234}U/^{238}U$ Atom Ratio	Statistical Uncertainty
1	Finland	Askola	5.444E-05	8.0E-08
2	Finland	Paukkajanvaara	5.126E-05	7.6E-07
3	Australia	Ranger Mine	5.455E-05	4.4E-07
4	Australia	Dam Operations	5.341E-05	6.2E-07
5	Canada	Cogema Resources	5.385E-05	6.0E-07
6	Canada	CAMECO Key Lake Op.	5.397E-05	3.4E-07
7	Gabon	Comuf Mounana	5.434E-05	4.2E-07
8	Czech Republic	DIAMO, Straz pod Ralskem	8.355E-05	4.9E-07
9	Canada	CAMECO Rabbit Lake Op.	5.444E-05	4.8E-07
10	Namibia	Roessing Uranium Mine	5.460E-05	4.1E-07
11	France	Cogema Lodeve	5.154E-05	2.8E-07
12	France	CETAMA Amethyste	5.340E-05	3.3E-07

Table 4: Variations in measured $^{234}U/^{235}U$ atom ratios from mines throughout the world [5], [6].

A plot of the measured $^{234}U/^{235}U$ atom ratios with associated uncertainties for all twelve samples is depicted in Fig. 4. Sample 8 from the Czech Republic has a significantly greater $^{234}U/^{235}U$ atom ratio than any other sample which cannot be explained by geological processes. One possibility may be a result of anthropogenic contamination with plutonium, especially ^{238}Pu [5]. This contamination may have occurred as a result of the Chernobyl accident. A more in depth comparison of the variation in the measured $^{234}U/^{235}U$ atom ratios with associated uncertainties with sample 8 omitted is depicted in Fig. 5.

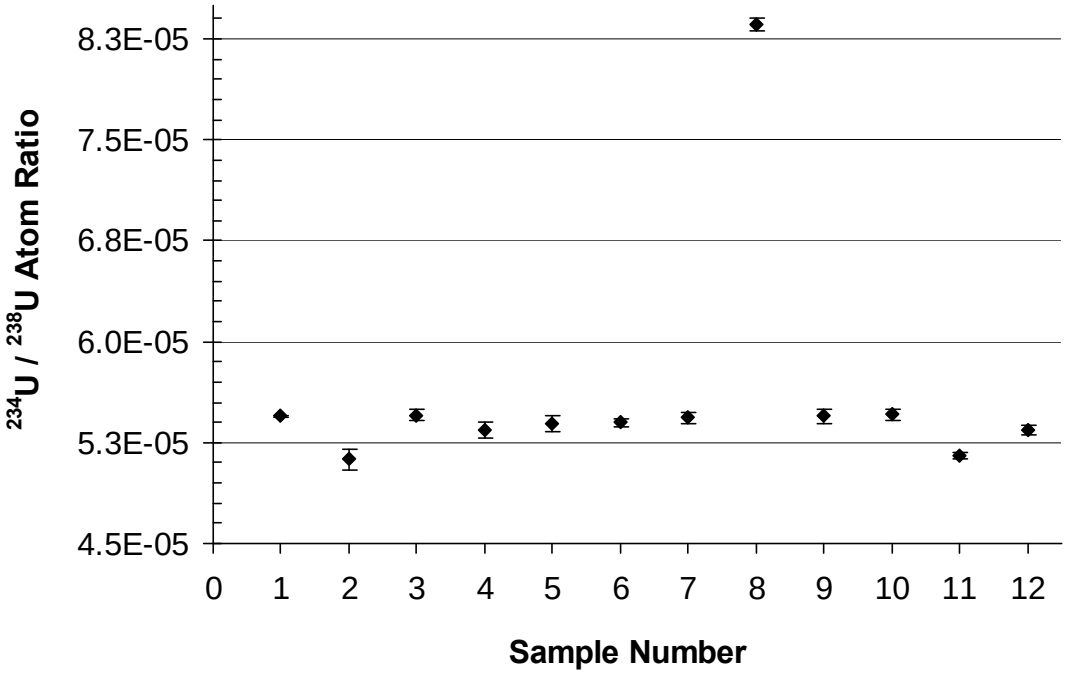

Figure 4. The $^{234}U/^{238}U$ atom ratio measured in all twelve samples.

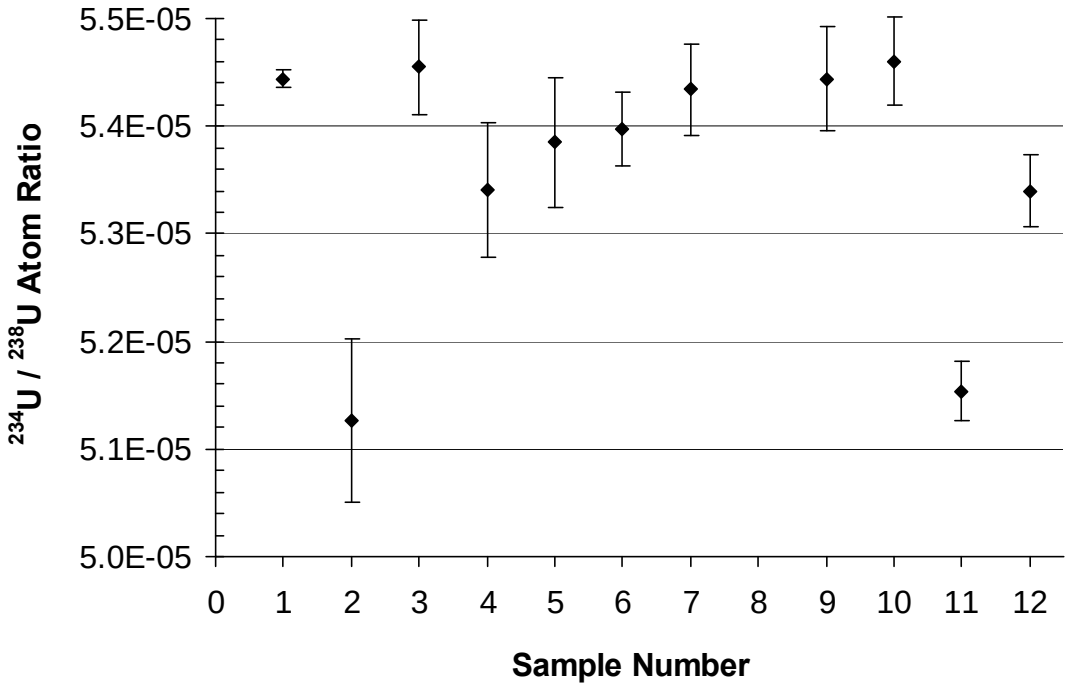

Figure 5. Expanded plot of the measured $^{234}U/^{238}U$ atom ratios excluding sample 8 [5], [6].

6. Discussion and Conclusion

6.1. Discussion

Various methods that could potentially be used to disguise the origin of the nuclear material used in HEU weapon prior to it being detonated were assessed in order to determine their effects on the validity of the algorithm. The first spoofing technique assessed was contamination of the original material used in the HEU weapon with fission products such as ^{137}Cs or ^{60}Co. This will result in higher measured post-detonation concentrations of the fission products used to contaminate the original weapons material. The total number of fissions in the device per unit mass will be affected if the fission products used to contaminate the original material are the same as the fission products used in this calculation. Using two fission products the significantly increases the probability of determining that original material was contaminated because there is a smaller probability that the two fission products used in the algorithm were also used to contaminate the original material. If only one of the fission products that was used in the algorithm was also used to contaminate the original material, then the total number of fissions in the device calculated using one fission product will differ significantly from the value calculated using the other fission product. Thus, indicating that one of the fission products was either present in the original material or else measured incorrectly.

Another spoofing technique assessed was boosting the weapon prior to detonation. In a boosted nuclear weapon, a mixture of deuterium (D) and tritium (T) gas is injected into the central core of ^{235}U metal sphere, called the “pit”. The implosion of the pit causes the ^{235}U to fission which in turn causes the atoms in the D-T mixture to undergo fusion. The fusion reaction produces large quantities of high energy neutrons (approximately 14 MeV) which travel through the compressed pit causing additional fission reactions [7]. The boosting of a nuclear weapon greatly increases the yield by causing more of the material to fission during detonation. Therefore, if calculated yield of an HEU weapon was on the order of 100 kT or greater it was probably boosted. In the case where a weapon was boosted prior to being

detonated but was a fizzle, then the atoms in the D-T mixture did not undergo fusion and post-detonation measurements of both deuterium and tritium could be obtained.

The last spoofing technique assessed was using a combination of plutonium and uranium metal or Mixed Oxide fuel (MOX) fuel as the original material in the weapon. This presents the most difficult problem because not only will of the fission product concentrations be higher but any signatures indicating the method of enrichment will disappear. In this case, it might be useful to combine techniques used to determine the original material in both an HEU and plutonium device.

6.2. Conclusion

In this work, an algorithm was developed that uses measured isotopic ratios from fission products and actinides present following the detonation of a nuclear weapon to compute the original material attributes of the weapon. The algorithm was comprised of analytical inversions of first-order differential equations derived directly from burnup and radioactive decay equations. The following post-detonation isotopic ratios were used: $^{89}Sr/^{238}U$, $^{95}Zr/^{238}U$, $^{232}U/^{238}U$, $^{234}U/^{238}U$, $^{235}U/^{238}U$, and $^{236}U/^{238}U$. The primary advantage gained from this methodology was it provided accurate solutions with essentially no computational time required. Error propagations were used to determine the sensitivity of the error in the calculated original ^{234}U, ^{235}U, and ^{236}U attributes for the HEU fuel. The errors in the calculated $^{234}U/^{238}U$ and $^{236}U/^{238}U$ attributes were linearly related the errors in measured parameters. The error in the calculated $^{235}U/^{238}U$ attribute varied nonlinearly as a function of the ^{235}U enrichment placing a significant importance on ensuring the accuracy of this value. The determined signature that indicated the enrichment process used to create the weapons material was based on the measured $^{234}U/^{238}U$ ratio. A source of error that was not assessed exists in the cross-section data used throughout the algorithm from the ORIGEN2 library for an FFTFC reactor. In this work, we were only testing the feasibility of the algorithm and did not consider its relationship to an actual weapon detonation. Thus, testing of this methodology using cross-section data obtained for an actual device detonation would improve the viability of the algorithm.

This work is important to homeland security and a significant prototype to data protocol in the event of a terrorist attack in our country. The algorithm developed was restricted only to HEU devices; however, future efforts will consider plutonium devices as well. It is also necessary to analyze how elements disperse in the environment and what current technology is available to measure isotopic fission fragments in the environment. All of the above aspects will affect the validity of the algorithm and if it could in fact be used if a terrorist device was detonated in the U.S.

7. Acknowledgements

I would like to thank Lee Cadwallader from Idaho National Laboratory for all of his help, as well as, Diane Fischer and Mark Laughter from Oak Ridge National Laboratory for the help they provided.

This research was performed while on appointment as a U.S. Department of Homeland Security (DHS) Scholar under the DHS Scholarship and Fellowship Program, a program administered by the Oak Ridge Institute for Science and Education (ORISE) for DHS through an interagency agreement with the U.S Department of Energy (DOE). ORISE is managed by Oak Ridge Associated Universities under DOE contract number DE-AC05-00OR22750. All opinions expressed in this paper are the author's and do not necessarily reflect the policies and views of DHS, DOE, or ORISE.

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