A METHOD FOR DETERMINING MATERIAL ATTRIBUTES FROM POST-DETONATION FISSION PRODUCT MEASUREMENTS OF AN HEU DEVICE

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ABSTRACT

An algorithm was developed that uses measured isotopic ratios from fission product residue following the detonation of a nuclear weapon to compute the original attributes of the nuclear material used in the weapon. While more accurate (and more computationally intensive) methods are being explored by others, the method described here could serve as a preprocessing step to a more detailed methodology (potentially saving on computational time). This would, in turn, expedite the process of determining where the device came from, eventually leading to which terrorist group perpetrated the event. This work was restricted only to Highly Enriched Uranium (HEU) devices; however, future efforts will consider plutonium devices as well. The attributes determined include original material uranium isotopics (at present considering only ^{234}U, ^{235}U, and ^{238}U) and the type of enrichment process used to create the material (e.g., gaseous diffusion, gas centrifuge, etc.). The approach to developing this algorithm involved a simulation of the fission products and actinides present following a nuclear explosion and a detailed evaluation to determine valid ratios that could be used to work backward and achieve the original material in the device. The algorithm used was purely analytical, derived directly from burnup and radioactive decay equations. Thus, this methodology provided solutions with essentially no computational time required.

INTRODUCTION

One of the most crucial issues to national security in the United States is the ability to safeguard our country against nuclear terrorism. If national security was breached and a terrorist nuclear device was detonated in the United States, how quickly could we assess the site to determine what type of device was detonated, how powerful the device was and where it came from? Nuclear threats are not widely understood by the general population; therefore, if a terrorist device was ever detonated in our country immediate results must be produced in order to prevent mass hysteria.

The objective of the algorithm developed here was to utilize post-detonation measured isotopic ratios in order to determine the pre-detonation material attributes within reasonable accuracy. More computationally intensive (and admittedly more accurate) 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 buildup and decay equations (all first-order ordinary differential equations). It is envisioned that this methodology could serve as a preprocessor step to a more computationally intensive and more accurate system.

This work concentrated on the attribution of an HEU weapon post-detonation. Terrorist devices may differ from military nuclear weapons mainly in the sophistication of constructed (e.g., type and grade of material used and quality of tamper/reflector). Terrorist nuclear weapons that use HEU are likely to be gun-type weapons due to the simplicity of its structure. Since gun-type assembly is relatively simple, this is considered to be a likely scenario for a nuclear terrorist attack. [4]

Given a measurement of the isotopics of residue post-detonation, 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 (in fact step (7) is probably not possible, but is still an interesting problem to attempt to solve). For the purposes of this study, we have so far limited the analysis only to gaseous centrifuge and gaseous diffusion enrichment methods, since these enrichment processes are very similar in physical process. It is expected that distinguishing most other methods (such as AVLIS or EMIS) would be much simpler.

^{234}U Isotopics in Mines

Different uranium mines throughout the world are characterized by different isotopic abundances of ^{234}U which may be used as a signature to indicate 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 (53.8 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 [2]. Table I shows some of these variations in naturally occurring uranium.

TABLE I
Variations in natural uranium ^{234}U isotopic abundances from mines throughout the world. [2]

Country	Mine/Mill Facility	^{234}U / ^{238}U Atom Ratio
Gabon	Comuf Mounana	5.4344E-05
Canada	CAMECO Rabbit Lake Op.	5.4440E-05
Namibia	Roessing Uranium Mine	5.4604E-05

Enrichment Methods

Weapons-grade HEU is typically enriched to about 90% ^{235}U. The method of enrichment is a signature that may indicate where the uranium was enriched. Methods used to enrich uranium include: gas centrifuge, gaseous diffusion, laser isotope separation, chemical/ion separation and electromagnetic isotope separation. The two most common enrichment methods are gaseous centrifuge and gaseous diffusion both of which separate the uranium isotopes in a gaseous state called uranium hexafluoride.

Both gaseous diffusion and gaseous centrifuge rely on the differences in mass between ^{235}U containing molecules and ^{238}U containing molecules though they are based on different physical processes. Gas centrifuge is based on centrifugation whereas gaseous diffusion is based on

molecular effusion. Since ^{234}U is lighter than ^{235}U it enriches even more in either the gas centrifuge or gaseous diffusion process than the ^{235}U. The following equation represents the ^{234}U enrichment for gaseous centrifuge: [3]

$$\left(\frac{N^{234}}{N^{238}}\right)_0 = 0.007731\left(\frac{N^{235}}{N^{238}}\right)_0^{1.0837}\left(\frac{M^{238}}{M^{235}}\right)^{1.0837} \tag{1}*$$

where M^{238} is the atomic mass of ^{238}U and M^{235} is the atomic mass of ^{235}U. The following equation represents the ^{234}U enrichment for gaseous diffusion: [3]

$$\left(\frac{N^{234}}{N^{238}}\right)_0 = 0.008\left(\frac{N^{235}}{N^{238}}\right)_0\left(\frac{M^{238}}{M^{235}}\right) \tag{2}*$$

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. The following equation represents the ^{236}U enrichment in U.S. origin fuel:

$$\left(\frac{N^{236}}{N^{238}}\right)_0 = 0.0046\left(\frac{N^{235}}{N^{238}}\right)_0\left(\frac{M^{238}}{M^{235}}\right) \tag{3}*$$

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.

Forward Model Development

The forward model simulations used the ORIGEN2 code [1]. 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 weapon 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. The uranium was enriched to 95 atom% ^{235}U and the ^{234}U and ^{236}U concentrations were calculated for both methods of enrichment using equations (1), (2), and (3). Then, ORIGEN was used to simulate the burnup of

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* These equations were taken from TransWare Enterprises Inc., "TransFX Computer Software Manuals," July 2001.

the material in the device given a 2 kT yield. The resultant isotopics from this burnup were then decayed for 1.0 day (assumes that it will take approximately 1 day or more to acquire measured resultants from residue post-detonation).

Inverse Model Development

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 inverse model uses an iterative procedure where the pre-detonation ^{235}U/^{238}U ratio is set to an initial guess input by the user. The pre-detonation ^{234}U/^{238}U and ^{236}U/^{238}U (if applicable) ratios were calculated using Eqs. (1)-(3) and then combined with the initial guess for ^{235}U/^{238}U in order to calculate the ^{235}U enrichment of the original material using:

$$e_0^i = \frac{\left(\dfrac{N^{235}}{N^{238}}\right)_0^{i-1}}{\left(\dfrac{N^{234}}{N^{238}}\right)_0^{i-1} + \left(\dfrac{N^{235}}{N^{238}}\right)_0^{i-1} + \left(\dfrac{N^{236}}{N^{238}}\right)_0^{i-1} + 1} \tag{4}$$

where e_0^i is the pre-detonation enrichment for step i and $\left(\dfrac{N^x}{N^{238}}\right)_0^i$ is the pre-detonation atom ratio of isotope x to ^{238}U from step i-1 (or from the initial guess for the first step).

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 following equation was used to calculate the number of fissions per unit mass in the device:

$$F_T^i = \left(\frac{N^{89}}{N^{238}}\right)_T \frac{N_A E_R}{M^{238} Y^{89}} e_0^i \left(\frac{N^{238}}{N^{235}}\right)_0^{i-1} \tag{5}$$

where F_T^i is the number of fissions in the device following irradiation (i.e., at time T) per unit mass for step i, $\left(\dfrac{N^{89}}{N^{238}}\right)_T$ is the measured ^{89}Sr/^{238}U atom ratio post-detonation (i.e., at time T), N_A is Avagadro's number, E_R is the recoverable energy per fission, and Y^{89} is the cumulative fission product yield for ^{89}Sr. In using Eq. (5) we assumed that the fission product yields and recoverable energy per fission from ^{235}U was adequate (i.e., this assumes that all fissions were from ^{235}U).

An updated ^{234}U/^{238}U value was then calculated using measurements of ^{232}U/^{238}U in the residue and the following equation:

$$\left(\frac{N^{234}}{N^{238}}\right)_0^i == \left(\frac{N^{232}}{N^{238}}\right)_T \frac{\sigma_f^{235}}{\sigma_{3n}^{234}} \frac{E_R}{F_T^i} \frac{N_A}{M^{235}} e_0^i \tag{6}$$

where σ_f^{235} is the one-group microscopic fission cross section for ^{235}U and σ_{3n}^{234} is the one-group microscopic (n,3n) cross section for ^{234}U. This equation assumes 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.

An updated ^{235}U/^{238}U value was then calculated using measurements of ^{235}U/^{238}U in the residue and the following equation:

$$\left(\frac{N^{235}}{N^{238}}\right)_0^i = \left(\frac{N^{235}}{N^{238}}\right)_T + \left(\frac{N^{235}}{N^{238}}\right)_0^{i-1} \frac{\sigma_a^{235}}{\sigma_f^{235}} \frac{F_T^i}{E_R} \frac{M^{235}}{N_A} e_0^i \tag{7}$$

where σ_a^{235} is the one-group microscopic absorption cross section for ^{235}U. This assumes that the change in ^{235}U is equal to its loss rate from absorption.

Then, an updated ^{236}U/^{238}U value was then calculated using measurements of ^{236}U/^{238}U in the residue and the following equation:

$$\left(\frac{N^{236}}{N^{238}}\right)_0^i = \left(\frac{N^{236}}{N^{238}}\right)_T - \left(\frac{N^{235}}{N^{238}}\right)_0^i \frac{M^{235}}{N_A e_0^i} \frac{F_T^i}{E_R} \left\{ \frac{\sigma_a^{235}}{\sigma_f^{235}} - 1 - \frac{\sigma_a^{235}}{2\sigma_f^{235}} \left[\left(\frac{N^{236}}{N^{235}}\right)_0^{i-1} + \left(\frac{N^{236}}{N^{235}}\right)_T \right] \right\} \tag{8}$$

This assumes the change in ^{236}U is equal to its production rate from radiative capture in ^{235}U minus the loss rate from the absorption of ^{236}U. Equation (8) 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.

The new value for the enrichment can now be calculated using Eq. (4). Equations (4)-(8) can then be repeated until the pre-detonation ^{235}U/^{238}U ratio converges to within some tolerance.

RESULTS AND DISCUSSION

The methodology described above was tested for a 2 kT detonation of a 95% 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. 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 II verified that for any positive initial guess of any order of magnitude input into the algorithm will be iterated to a reasonably correct answer.

TABLE II
Comparison of the values calculated by the inverse model with various initial guesses for the ^{235}U concentration and the actual values for the original material attributes.

Enrichment Process	Initial Guess (N235/N238)$_0$	Original Value (N235/N238)$_0$	Inverse Model (N235/N238)$_0$	Percent Error
Centrifuge (with ^{236}U)	1.00×10^{10}	42.4297	43.1132 ± 0.4309	1.6110%
Diffusion (no ^{236}U)	1.00×10^{-10}	22.4057	22.5538 ± 0.2254	0.6613%

The measured isotopic values generated from ORIGEN2 and the values computed in the inverse model for both centrifuge and diffusion enrichment processes (with and without ^{236}U) are presented in Tables III and IV. The results from the inverse model were consistently higher than the exact values for the original material attributes. The resulting error may be attributed to an assumption made when developing the algorithm that the atomic density of ^{238}U did not change with time.

In order to determine valid signatures indicating the method of enrichment, the measured values for post-detonation ^{234}U concentrations were compared. For centrifuge enriched fuel, the ^{234}U concentration was approximately 5.0 times greater than the ^{234}U concentration for diffusion enriched fuel. These significant variations in ^{234}U were used as signatures indicating the enrichment process used.

TABLE III
Comparison of the inverse model calculation and the exact value for the original material attributes for gaseous centrifuge and diffusion enrichment without ^{236}U.

Enrichment Process	Atomic Ratio	Measured Value (T = 1.1 days)	Original Value (T = 0 days)	Inverse Model (T = 0 days)	Percent Error
Centrifuge (no ^{236}U)	N235 / N238	33.921569	35.500800	36.0751 ± 0.3605	1.6179%
	N234 / N238	0.857089	0.868500	0.8831 ± 0.0127	1.6889%
	N236 / N238	0.397059	0.000000	0.0047 ± 0.0075	-
Diffusion (no ^{236}U)	N235 / N238	21.401713	22.405700	22.5538 ± 0.2254	0.6613%
	N234 / N238	0.177368	0.179200	0.1816 ± 0.0047	1.3090%
	N236 / N238	0.251309	0.000000	0.0273 ± 0.0026	-

TABLE IV
Comparison of the inverse model calculation and the exact value for the original material attributes for gaseous centrifuge and diffusion enrichment with ^{236}U.

Enrichment Process	Atomic Ratio	Measured Value (T = 1.1 days)	Original Value (T = 0 days)	Inverse Model (T = 0 days)	Percent Error
Centrifuge (with ^{236}U)	N235 / N238	40.540784	42.429656	43.1132 ± 0.4106	1.6110%
	N234 / N238	1.024335	1.037963	1.0552 ± 0.0151	1.6641%
	N236 / N238	0.668319	0.195176	0.2041 ± 0.0110	4.5841%
Diffusion (with ^{236}U)	N235 / N238	23.860440	24.980279	25.3754 ± 0.2536	1.5817%
	N234 / N238	0.197745	0.199842	0.2042 ± 0.0029	2.2035%
	N236 / N238	0.394534	0.114909	0.1213 ± 0.0065	5.5651%

TABLE V
Comparison of the inverse model calculation and the exact value for the original material attributes for gaseous diffusion enrichment with and without ^{236}U.

Enrichment Process	Atomic Ratio	Original Values (T = 0 days)	Inverse Model (T = 0 days)	Percent Error
Diffusion (with ^{236}U)	N235 / N238	24.980279	25.3754 ± 0.2536	1.5817%
	N234 / N238	0.199842	0.2042 ± 0.0029	2.2035%
	N236 / N238	0.114909	0.1213 ± 0.0065	5.5651%
Diffusion (no ^{236}U)	N235 / N238	22.405700	22.5538 ± 0.2254	0.6613%
	N234 / N238	0.179200	0.1816 ± 0.0047	1.3090%
	N236 / N238	0.000000	0.0273 ± 0.0026	-

After the enrichment process has been determined, whether or not ^{236}U existed in original weapons material must be established. The values computed in the inverse model for diffusion enriched fuel with and without ^{236}U are presented in Table V. For diffusion enriched fuel (with ^{236}U), the ^{236}U value from the inverse model was approximately 4.5 times greater than the ^{236}U value for diffusion enriched fuel (without ^{236}U).

In the derivation of Eq. (8) it was assumed that the ratio of ^{236}U/^{235}U was a linear function with respect to time and could therefore be easily integrated. The assumption was verified in the following figure which depicts the ratio as a function of irradiation time. A linear trend line was used to fit to the data points.
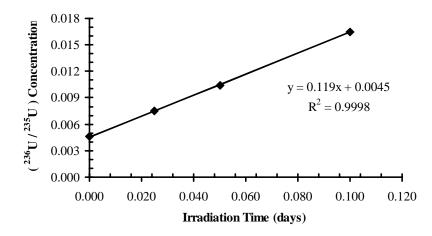

Figure 2. ORIGEN2 calculation of (^{236}U/^{235}U) isotopic ratio as a function of irradiation time for irradiation of 95% enriched uranium enriched.

CONCLUSIONS

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. The results computed in the inverse model for the original attributes of the HEU fuel were compared for gaseous centrifuge and gaseous diffusion enriched fuel, both with and without ^{236}U present in the original material. The values computed in the algorithm were consistently higher than the exact values; however, the resulting percent differences, ranging from 0.6631% to 5.5651%, were always within the acceptable uncertainties. The determined signature that indicated the use of a centrifuge enrichment process to create the weapons material was based on the ^{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. Also, there was no testing of "spoofing" mechanisms performed.

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.

ACKNOWLEDGMENTS

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