Determination of the ^{235}U Enrichment of Bulk Uranium Samples Using Delayed Neutrons

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Abstract
A technique for utilizing the physics of the delayed neutron re-interrogation method [1] to determine uranium enrichment is presented in this paper. A series of active interrogation measurements was performed using pulsed 14-MeV neutrons and a polyethylene moderated ^{3}He based neutron detection system. Proof of principle measurements were performed on a set of bulk uranium oxide standards of differing enrichments. A series of measurements was performed on a set of uranium "unknowns" with and without high-Z gamma-ray shielding (lead) present. Uranium enrichment estimates were obtained for all cases including the bulk uranium samples shielded by lead. Further refinement of this technique is needed to make it a more powerful tool for non-destructive assay of bulk uranium samples.

Keywords: Uranium Enrichment, Active Interrogation, Delayed Neutrons

Introduction
Measurement of uranium enrichment is an important attribute to determine for non-destructive assay of bulk uranium samples. The "standard" methods to determine uranium enrichment rely on the observation of spontaneously emitted gamma-rays from ^{235}U and ^{238}U. The characteristic gamma-rays that identify the presence of ^{235}U are of low energy, 144, 186 and 205-keV. Due to self-shielding effects only the outer few millimeters of the uranium sample contribute to the observed radiations. If a small thickness of high-Z gamma-ray shielding is present, the gamma-ray signature for the presence of ^{235}U may be completely absent.

When attribution information from passive measurement techniques is limited because of the presence of shielding, etc., measurement of the actively induced responses can provide more information about a uranium system. The delayed neutron re-interrogation technique is an active interrogation technique that can be utilized for non-destructive assay of uranium.

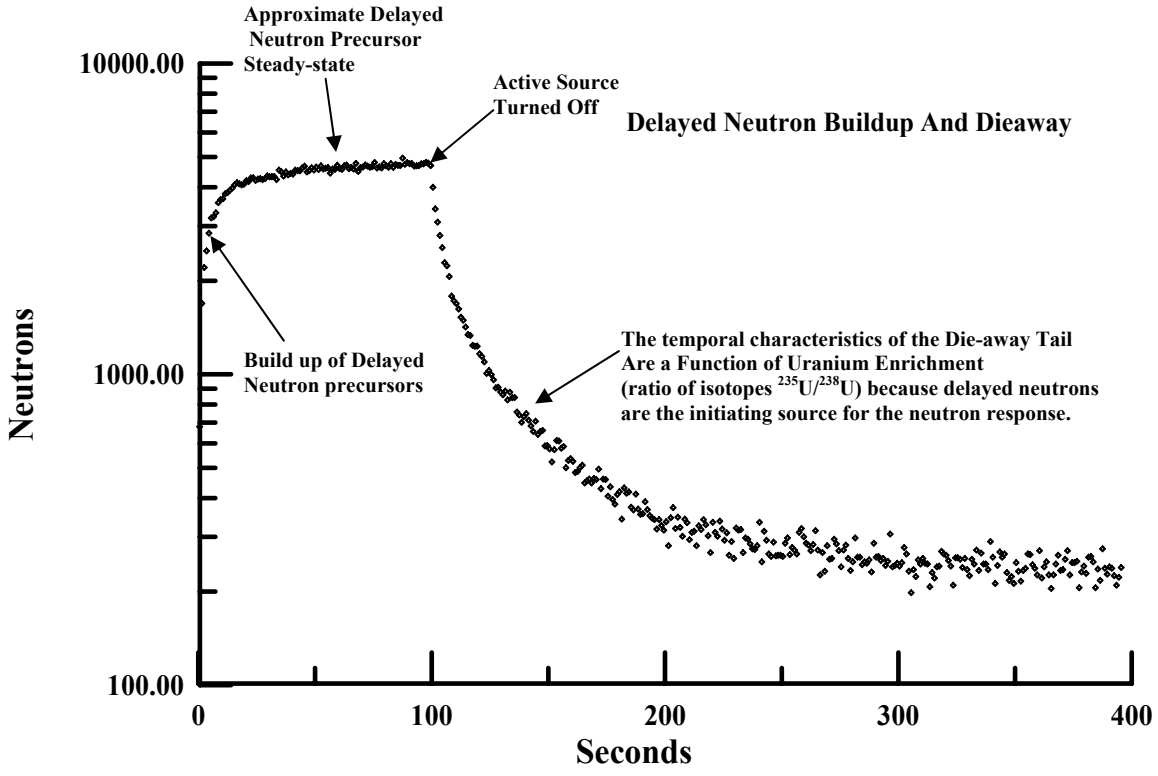
Methods for using active non-destructive assay techniques that utilize the delayed neutron driven induced neutron response have been presented by Li [2] and Campbell [3] for determining relative mixtures of actinides for small samples of a known mass. This work will demonstrate how one can use the delayed neutron re-interrogation technique for estimating ^{235}U enrichment for bulk samples of uranium of unknown mass without the need for measurement standards.

1. Delayed Neutron Re-interrogation Technique

Both pulsed neutrons and pulsed photons can be used as the interrogating source when employing the delayed neutron re-interrogation technique. For the enrichment measurement described below, a continuous direct current active source would work as well.

The creation of an intrinsic steady-state source of delayed neutrons is accomplished by repetitious interrogation using an active source to create fission products throughout the uranium. The emission of a delayed neutron occurs randomly and later in time with respect to the parent fission that produced the delayed neutron precursor. The delayed neutrons either leak from the system or initiate further fission chains. After a finite time, the delayed neutron precursor populations reach apparent steady-state levels and the continued active interrogation results in creating a near steady-state source of neutrons that is distributed throughout the uranium that re-interrogates the material. The intrinsic neutron source strength is dependent upon the intensity and ability of the active source to induce fissions in the uranium during a pulse. Once the active source is "turned off", the neutron response is being driven by a time-dependent decreasing population of delayed neutron precursors. The abundances and kinetic behavior of the induced fission products that emit delayed neutrons drive the induced neutron response. Fig. 1 illustrates this physics.

Figure 1: The time dependence of the neutron response that illustrates the delayed neutron buildup, approximate equilibrium, and die-away for an active measurement performed on a 91% enriched ^{235}U oxide sample interrogated by pulsed 14-MeV neutrons.



2. Use of Delayed Neutrons For Enrichment Determination

The kinetic properties of delayed neutrons are very useful for interpreting dynamic neutron measurements that involve active interrogation of uranium. The uniqueness of the delayed neutron emission properties (abundances and temporal properties) for ^{235}U and ^{238}U provides signatures for the delayed neutron population buildup and the delayed neutron population die-away. The time behavior of the neutron response is a function of the delayed neutron precursor population created by fissions of the actinide isotope mixture. The isotope mixture (or enrichment for the case of uranium) can be estimated by examining the shape and time behavior of the neutron response. Since high-Z materials are not as effective shielding neutrons as gamma rays, use of an active technique may make possible an enrichment estimate for a bulk uranium sample (of unknown mass) shielded by a high-Z material.

3. Theory

A theoretical expression for enrichment related to the actively induced delayed neutron driven neutron response can be derived starting from an equation presented by Li [see ref. 2]. A six-group delayed neutron precursor model is used. The measured delayed neutron count rate $C_T(t)$ for a small fissile sample irradiated by some flux rate Φ of fission inducing particles is given by equation (3.1).

$$C_T(t) = \varepsilon \cdot \left(\frac{\nu_d m N_A \sigma_f \Phi}{M} \right) \sum_{i=1}^{6} \beta_i \frac{1}{\lambda_i} \left(1 - e^{-\lambda_i t_b}\right) \left(e^{-\lambda_i t_c}\right) \left(1 - e^{-\lambda_i t}\right) = \varepsilon \cdot f(t_b, t_c, t) \cdot m \tag{3.1}$$

Where:

$C_T(t)$ = measured delayed neutron count rate

ε = efficiency of the neutron detection system

ν_d = average number of delayed neutrons emitted per fission

m = mass of fissile actinide (g)

N_A = Avogadro's number

σ_f = fission cross section of interrogating particles (bn)

Φ = particle flux to which sample is exposed ($cm^{-2}s^{-1}$)

M = atomic mass number of fissionable nuclide (g/mol)

β_i = fraction of delayed neutrons emitted in group i

λ_i = decay constant of delayed neutron in group i

t_b = irradiation time (s)

t_c = cooling time (s)

t = measurement time (s)

f = delayed neutrons born per unit mass (a function of time)

If one assumes that the fission product inventory is in equilibrium due to an infinitely long irradiation time and the cooling time is assumed to be zero (immediate measurement of the sample after irradiation), equation (3.1) becomes

$$C_T(t) = \varepsilon \cdot \left(\frac{\nu_d m N_A \sigma_f \Phi}{M} \right) \sum_{i=1}^{6} \beta_i e^{-\lambda_i t} = \varepsilon \cdot f(t) \cdot m . \qquad (3.2)$$

The quantity $\left(\frac{m N_A \sigma_f \Phi}{M} \right)$ is proportional to the delayed neutron precursor population reached in the mixture during irradiation.

For a sub-critical multiplying mixture with **n** fissile actinide components, the expression for the measured neutron count rate can be approximated by equation (3.3).

$$C_T(t) = \varepsilon \cdot K_l \cdot \sum_{j=1}^{n} f_j(t) \cdot m_j \qquad (3.3)$$

Where

K_l = sub-critical neutron multiplication factor associated with the fissile mixture.

and

$f_j(t)$ = time-dependent functional form of the "starter" delayed neutron population born per unit mass due to fission of component j actinide during the irradiation.

Hence, for a sample composed of **n** fissile isotopes

$$C_T(t) = D \cdot \sum_{j=1}^{n} A_j \cdot f_j(t) \qquad (3.4)$$

Where

$$D = \varepsilon \cdot K_l \cdot N_A \cdot \Phi , \qquad (3.5)$$

$$A_j = \frac{\nu_{dj} \cdot m_j \cdot \sigma_{fj}}{M_j} \tag{3.6}$$

and

$$f_j(t) = \sum_{i=1}^{6} \beta_{ij} e^{-\lambda_{ij} t} \quad . \tag{3.7}$$

For a two component system, equation (3.4) simplifies to

$$C_T(t) = B_1 \cdot F_1(t) + B_2 \cdot F_2(t) \tag{3.8}$$

where

$$B_1 = D \cdot A_1 \qquad \text{and} \qquad B_2 = D \cdot A_2 \, .$$

Enrichment with respect to fissile component 1 is defined as

$$E_1 = \frac{m_1}{m_1 + m_2} \tag{3.9}$$

Using Equations (3.6) and (3.8) and after performing some simple algebra, equation (3.9) becomes

$$E_1 = \frac{\nu_{d2} \cdot \sigma_{f2} \cdot B_1 \cdot M_1}{\nu_{d2} \cdot \sigma_{f2} \cdot B_1 \cdot M_1 + \nu_{d1} \cdot \sigma_{f1} \cdot B_2 \cdot M_2} \tag{3.10}$$

The coefficients B_1 and B_2 are determined from the best fit of the data using the functional form of equation (3.8).

The error associated with the enrichment estimate of equation (3.10) can be quantified using the formula [4, 5]

$$\sigma_{E_1}{}^2 = \left(\frac{\partial E_1}{\partial B_1} \right)^2 \cdot \sigma_{B_1}{}^2 + \left(\frac{\partial E_1}{\partial B_2} \right)^2 \cdot \sigma_{B_2}{}^2 \tag{3.11}$$

This error formulation assumes that the uncertainty in the enrichment estimate is associated with only the uncertainties of fitting the coefficients B_1 and B_2 . The nuclear property constants for ^{235}U and ^{238}U are assumed to have no uncertainties associated with them.

4. Experimental Protocol

For the proof of principle measurements, a set of six uranium oxide standards [6] were used as bulk uranium samples. All the samples were contained in standard tinned steel food cans (>99 wt. % Fe). Each sample had about the equivalent uranium mass but differing enrichments of ^{235}U. Table 1 lists the characteristics of the samples.

Table 1: ^{235}U and ^{238}U isotopic Content of the Uranium Oxide Samples Used for the Uranium Enrichment Determination Proof of Principle Measurements

Standard ID	^{235}U weight %	^{238}U weight %	Oxide Mass (grams)	U Mass (grams)	^{235}U Mass (grams)	^{238}U Mass (grams)
UISO-91	91.419	7.33	1178	990	904	86
UISO-66	66.317	32.8	1171	990	658.8	331.2
UISO-52	52.426	46.2	1171	989	515.4	473.6
UISO-38	37.848	61.6	1172	991	372.1	618.9
UISO-27	27.000	72.3	1174	991	265.0	726.0
UISO-13	13.098	86.7	1171	991	128.4	862.6

A set of measurements were performed on some “unknown” uranium metal samples with and without shielding materials present. Table 2 lists the characteristics of the “unknown” samples used in the measurements. For the shielded cases, the uranium samples were placed in a close fitting lead cylinder with a wall thickness of 1.59 cm (top and bottom included).

Table 2: Characteristics of the Uranium Samples Used as Part of the “Unknown” Measurements Data Set.

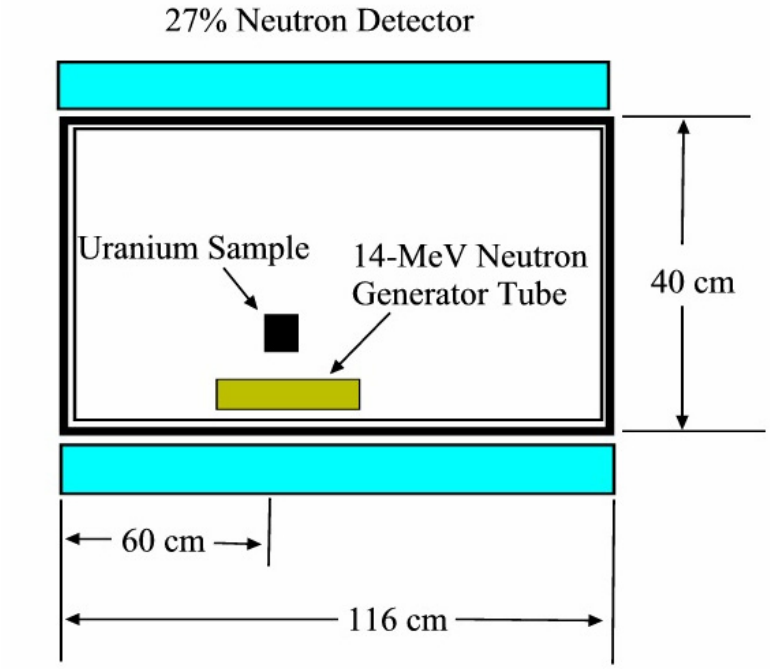
ID	Form and Geometry	Total Mass (g)	Weight % ^{235}U
Highly Enriched Uranium (HEU) “unknown”	Metal Right cylinder	987	93.15
Depleted Uranium (DU) “unknown”	Metal Right cylinder	1003	0.22

The neutron detection system used for the measurements was built using forty-eight ^{3}He tubes (2.54 cm diameter by 100 cm length filled to a gas pressure of 0.2 MPa) arranged into sixteen modules. Each module contained three ^{3}He tubes contained in a cavity made of polyethylene. The modules were arranged to form an internal cavity with dimensions 40 cm x 40 cm x 116 cm open at both ends to allow access for placement of samples. The internal surface of the cavity was covered by a thin layer of cadmium. The detection efficiency for fission energy neutrons was 27 %.

The active interrogation source used for the measurements was a commercially available pulsed 14-MeV neutron generator system (MF Physics model CC A-210) [7]. The system was operated at pulse repetition rate of 50 Hz with a 14 MeV neutron output of

approximately 10^6 neutrons per pulse into 4π steradian. Fig. 2 shows a schematic of the geometry for the experimental measurements.

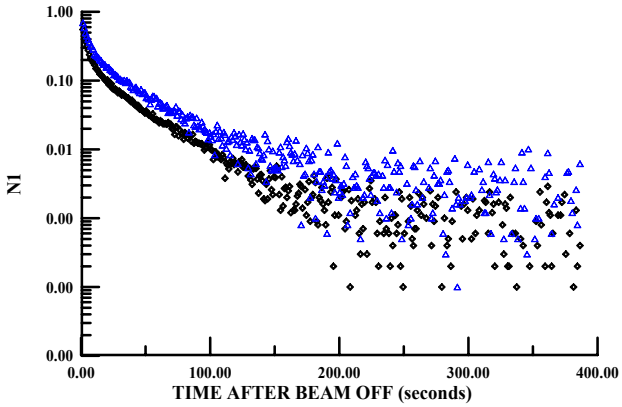
Figure 2: Geometry of the Active Uranium Enrichment Determination Measurement



For performing the measurements and analysis, it is important to utilize a list mode data acquisition system as part of the experimental setup. A list-mode data set allows one to archive the original data set and manipulate the data in multiple ways to determine the best analysis technique. The data acquisition employed for the measurements was the custom Los Alamos-designed pulse arrival-time recording module PATRM [8].

Once an intrinsic steady-state source of delayed neutrons is achieved in the bulk sample of uranium, the active source is "turned off" and the neutron response is recorded. When the active source is turned off, the neutron response appears to be a sum of decaying exponentials (see Fig. 3) that are functions of the delayed neutron precursor population driving the system. The die-away data was measured for a total of 400 seconds after the active source was turned off.

Figure 3: The Normalized Delayed Neutron Driven Neutron Response for the ~91% Enriched Uranium Oxide Sample (Upper curve) and a Depleted Uranium ~0.2% Enriched Sample (Lower curve).



Using Keepin's six group fast fission delayed neutron precursor parameters [9] for ^{235}U and ^{238}U in equation (3.8), a best fit determination of the parameters B_1 and B_2 is performed on the measured response data using the JANDEL SigmaPlot software [10]. The "best fit" values of B_1 and B_2 , Keepin's fast fission delayed neutron yield data, and 14-MeV neutron fission cross section data [11] (see Table 3) are used in equation (3.10) to estimate the enrichment. For comparison purposes, fits were performed using data for varying lengths of time after the active source was turned off (i.e. first 60 seconds after source was turned off, first 100 seconds after source was turned off, .etc).

Table 3: Delayed Neutron Yields and 14-MeV Neutron Fission Cross sections for ^{235}U and ^{238}U.

Isotope	ν_d neutrons per fission	$\sigma_f(14\ \text{MeV})$ (bn)
^{235}U	0.0165	2.08
^{238}U	0.0412	1.128

No corrections due to background were made to the data and we are currently ignoring any contribution to the measured response due to the presence of other uranium isotopes in the samples (^{234}U and ^{236}U).

5. Results and Discussion

Table 4 gives the results of the analysis for the proof of principle measurements performed on the set of UISO standards.

Table 4: Enrichment Estimates Inferred From The Delayed Neutron Driven Neutron Response For the UISO Standards.

Case	Die-Away Time For Fit (seconds)	^{235}U Enrichment Estimate (weight %)	Error For Enrichment Estimate (weight%)	Case	Die-Away Time For Fit (seconds)	^{235}U Enrichment Estimate (weight %)	Error For Enrichment Estimate (weight %)
UISO-91	60	91	± 3	UISO-66	60	76	± 5
	100	88	± 2		100	71	± 4
	200	85	± 2		200	69	± 3
	300	84	± 1		300	69	± 2
UISO-52	60	41	± 6	UISO-38	60	49	± 5
	100	42	± 4		100	41	± 4
	200	41	± 3		200	36	± 3
	300	42	± 2		300	36	± 2
UISO-27	60	45	± 5	UISO-13	60	31	± 6
	100	38	± 4		100	28	± 4
	200	37	± 3		200	24	± 3
	300	37	± 2		300	23	± 3

Table 5 presents the set of measurements performed on the "unknown" uranium samples with and without lead shielding.

Table 5: Enrichment Estimates Inferred From the Delayed Neutron Driven Neutron Response For the Set of "Unknown" Measurements.

Case	Die-Away Time For Fit (seconds)	Enrichment Estimate (weight %)	Error For Enrichment Estimate (weight %)	Case	Die-Away Time For Fit (seconds)	Enrichment Estimate (weight %)	Error For Enrichment Estimate (weight %)
DU "unknown"	60	9	± 3	HEU "unknown"	60	92	± 2
	100	6	± 2		100	90	± 2
	200	3	± 2		200	88	± 1
	300	2	± 1		300	88	± 1
DU "unknown" Shielded by Pb	60	10	± 4	HEU "unknown" Shielded by Pb	60	88	± 4
	100	6	± 3		100	88	± 3
	200	4	± 2		200	86	± 1
	300	3	± 2		300	86	± 1

6. Summary and Conclusions

The above results show some promising trends that one can estimate uranium enrichment for a bulk uranium sample of unknown mass using the delayed neutron re-interrogation technique. Estimates for enrichment were obtained in the presence of gamma-ray shielding which would not have been possible using traditional gamma-ray measurement techniques. Further refinement will make this technique a very powerful tool for non-destructive assay of bulk uranium samples.

One possible way to get more accurate enrichment estimates is to develop a more sophisticated fitting routine to determine the "best fit" of the data. One should re-evaluate

some of the initial assumptions that went into modeling the delayed neutron driven neutron response (i.e. the delayed neutron precursor groups are in equilibrium due to an infinitely long irradiation time). A more sophisticated model for the delayed neutron driven neutron response that accounts for multi-energy induced fissions (fission energy induced fissions verses 14-MeV induced fissions) could be developed which may help reduce the error in the estimates. Also, other multi-group representations of the delayed neutron kinetic parameters could be incorporated into the analytical expression for the delayed neutron driven neutron response which may provide a model which allows a better fit to the data. Investigation of these possible refinements will be topics for future research.

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