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**Nondestructive Fissile Isotopic
Measurement Technique for ^{233}U - ^{235}U
Fuels Using Prompt and Delayed
Fission-Neutron Counting**

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E. J. Allen* and S. R. McNeany*

ABSTRACT

Assay and analysis procedures were developed for nondestructive fissile isotopic measurement of mixed ^{233}U - ^{235}U fuel samples. For ^{233}U much fewer delayed neutrons are released per fission than for ^{235}U , although the number of prompt neutrons is approximately the same. By separately counting prompt and delayed neutrons released by a sample exposed to neutron irradiation, the amounts of ^{233}U and ^{235}U present in the sample can be estimated. Equations of delayed- and prompt-neutron counts versus ^{233}U and ^{235}U contents are solved simultaneously for the ^{233}U and ^{235}U contents of a sample.

Eleven samples containing mixtures of ^{233}U and ^{235}U from no ^{233}U to nearly 100% were prepared and assayed in prompt- and delayed-neutron assay devices. Constants for calibration equations, which are derived in this report, were fitted to data from nine of the samples. The maximum differences between counts calculated by the calibration equations and measured counts were 2.3% for delayed neutrons and 1.2% for prompt neutrons, indicating a good selection of the form for the calibration equations. The two remaining samples were treated as unknowns, and the uranium contents of these samples were estimated by simultaneously solving the two calibration equations. The maximum difference between estimated ^{233}U or ^{235}U content and actual content for either sample was 1.5%.

An assay procedure using neutron irradiation and delayed- and prompt-neutron counting, which can be made relatively insensitive to gamma ray background radiation by proper detector selection and shielding design, was found applicable to assay of ^{233}U - ^{235}U fuels. Nuclear facilities handling ^{233}U - ^{235}U fuels can use this analysis procedure to help satisfy material accountability and quality control requirements.

Keywords: nondestructive assay, ^{233}U measurement, ^{235}U measurement, HTGR reactors, prompt neutrons, delayed neutrons, californium-252, fissile materials, neutron sources, material accountability, quality control.

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INTRODUCTION

Thorium and ^{233}U fuels that are recycled can contain significant quantities of the ^{235}U isotope as a result of successive neutron captures in ^{233}U and mixing of ^{235}U feed with the ^{233}U . To satisfy material accountability requirements and for certain quality control applications in nuclear facilities handling these mixed ^{233}U - ^{235}U fuels, measurement of ^{233}U and ^{235}U contents in feed material, product, and waste is required. These measurements may be achieved by chemical and mass spectrometric methods or by nondestructive assay. Although nondestructive assay methods are usually less precise than chemical analysis methods, nondestructive methods typically have a shorter measurement time and avoid sampling errors. However, many nondestructive assay methods do not work well with ^{233}U fuels because of the presence of a high gamma ray background produced by decay of ^{232}U and its decay products. (The isotope ^{232}U is present in small quantities in ^{233}U fuels because it is produced along with ^{233}U when the fertile material ^{232}Th is converted in nuclear reactors.) However, several nondestructive assay methods employing neutron irradiation and prompt- or delayed-neutron detection have been developed. These methods are relatively insensitive to a gamma background.

Two such nondestructive assay techniques have been developed for application to ^{233}U - ^{235}U fuels in support of the High-Temperature Gas-Cooled Reactor (HTGR) Fuel Recycle Development Program. Both techniques use thermalized neutrons from a ^{252}Cf spontaneous-fission neutron source to cause fissions in a fuel sample. One device measures the delayed fission neutrons from the sample by rapidly transferring the sample from an irradiation chamber to a detection chamber where the delayed neutrons are counted. The second device counts the prompt and the delayed fast fission neutrons released by the sample while it is being irradiated. As the number of delayed neutrons constitute less than 1% of the total number of neutrons released per fission for ^{233}U or ^{235}U , this assay device will be referred to as the prompt-neutron assay device.

The number of delayed neutrons released per fission for ^{233}U is about half the number released per fission for ^{235}U . However, both isotopes release about the same number of prompt neutrons per fission. The large differences in the relative number of delayed and prompt neutrons released per fission for ^{233}U and ^{235}U can be used to estimate, for a particular sample, the fissile isotopic contents of that sample. If a sample containing ^{233}U and ^{235}U is irradiated and the prompt and delayed neutrons are counted, these counts can be used to estimate the ^{233}U and ^{235}U contents. Two calibration equations are solved simultaneously for ^{233}U and ^{235}U contents from the measured prompt- and delayed-neutron counts for that sample.

This paper reports the results of an experiment to investigate assay of ^{233}U - ^{235}U fuel samples by neutron irradiation and prompt- and delayed-neutron counting. Data analysis procedures useful for this assay technique are described.

DESCRIPTION OF ASSAY INSTRUMENTATION

The assay devices are located in the Thorium Uranium Recycle Facility (TURF) at Oak Ridge National Laboratory. To facilitate design changes and to protect personnel from the unshielded ^{252}Cf neutron source when experiments are being conducted, the devices are situated in a shielded hot cell (Hot Cell B). When not in use the ^{252}Cf source can be removed from either assay device and stored in a biologically shielded tank, allowing safe entrance into the cell.

The ^{252}Cf neutron source is contained within a 15.9-mm-OD by 375-mm-long stainless steel tube, which can be positioned into either the prompt- or delayed-neutron assay device. The source, itself, which has a point geometry, is sealed in a small metal capsule, which is screwed to the end of a polyethylene rod. The rod is sealed in the stainless steel source tube. As of July 1, 1978, the source had an activity of about 1.4×10^9 n/s. The half-life of the ^{252}Cf isotope is 2.63 years and its specific activity is about 2.3×10^{15} n/s kg.

Delayed-Neutron Assay Device

The delayed-neutron assay device comprises four main components, which are arranged as shown in Fig. 1. The first is a thermal neutron irradiator, which uses a removable ^{252}Cf neutron source. Samples activated in the irradiator are then counted in the second component, a neutron detector assembly. A pneumatic transfer system, the third component, contains 15 m of polyethylene tubing and shuttles samples between the irradiator and the detector. The transport system also has a branch for loading and unloading samples. The sequence of events necessary to automatically cycle the sample through the irradiation, decay, and counting stages is controlled by the fourth component, a programmable logic controller. The neutron irradiator and delayed neutron detector are described briefly below. A more complete description of the device is reported elsewhere.^{1,2}

Fig. 1. Delayed-Neutron Nondestructive Assay Device.

The irradiation chamber is shown in detail in Fig. 2. Basically, it is a series of concentric cylindrical annuli that surround the sample and moderate the high-energy neutrons emitted by the ^{252}Cf source. The innermost moderator is a 117.5-mm-OD by 508-mm-long annulus of machined high-density polyethylene, whose central hole is 41.3 mm in diameter. Through this hole, the pneumatic transfer tube is inserted. The annulus also contains a blind axial hole (16.3 mm diam by 354 mm deep) that lies 35.1 mm off-center and contains the ^{252}Cf source tube.

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Fig. 2. Thermal Neutron Irradiator Containing ^{252}Cf Neutron Source.

To ensure uniform irradiation of the sample (about the vertical axis of the sample), the inner polyethylene annulus and the source it contains are rotated about the pneumatic tube end at about 15 rpm. The annulus is mounted in bearings at either end and is attached at the top by a rubber belt drive to a small ac motor. Since the ^{252}Cf is essentially a point source, rotating it about the sample during irradiation gives the time-averaged effect of a ring-shaped source.

The inner polyethylene annulus is mounted on bearings that are seated in a stationary polyethylene shell having a wall thickness of 12 mm. At the bottom the shell closes to a 38.1-mm-diam hole in which the bottom of the aluminum pneumatic tube end is fixed. The polyethylene shell is encased in a 3.18-mm-thick-walled 6061-aluminum housing, which is suspended in a water-filled 0.11-m^3 (30-gal) stainless steel drum. To prevent corrosion of the aluminum housing, its outside surface is

coated with a thick layer of epoxy paint. The water in the drum helps moderate the source neutrons and reflects them back into the sample region.

The structure of the detector assembly is shown in the vertical cross sectional drawing of Fig. 3. The heart of the detector system is a circular array of eight 25-mm-diam ^3He -filled proportional counters [0.4 MPa (4 atm) gas pressure, 254 mm active length], which are embedded in a cylindrical high-density polyethylene moderator. The counters, Reuter-Stokes model U564, have closely matched operating characteristics. They are mounted in a toroidal, hermetically sealed metal can, in which they are connected in parallel to a common preamplifier output and high-voltage input.

Fig. 3. Vertical Cross-Sectional View of Delayed-Neutron Detector.

The lead annulus that surrounds the sample shields the ^3He tubes from gamma radiation emitted by ^{233}U and/or ^{235}U fission products and by ^{232}U and its decay products, which would be present² in ^{233}U fuel samples. If the ^3He tubes are not shielded, some of this

radiation would be detected along with the delayed neutrons. The 20 mm lead thickness was calculated to reduce the dose to the ^3He tubes to about 100 mrd/h, (0.28 $\mu\text{Gy/s}$) from a sample containing 1 g ^{233}U with 1200 ppm ^{232}U whose decay products had accumulated for one year. The discriminator level is set sufficiently high to reject gamma pileup from this level of gamma radiation.

To shield out neutron radiation originating outside the detector assembly (background radiation), the counter tube-moderator assembly is encased in a 48-mm-thick polyethylene shell. Between this shell and the inner moderator is a 0.79-mm-thick layer of cadmium. Background neutrons originating outside the detector assembly are thermalized in the shell and absorbed in the cadmium layer before they can reach the counter tubes.

Because large quantities of ^{252}Cf are stored in an adjacent hot cell at TURF, the neutron background in the vicinity of the detector is greater than can be adequately shielded by the polyethylene shell. Thus, shielding was added by placing the detector assembly in a water-filled 910 mm by 610 mm by 610 mm rectangular tank. This provides a minimum of 120 mm water around the detector assembly and reduces the typical background from about 9 counts/s to less than 2 counts/s.

Prompt-Neutron Assay Device

The prompt-neutron assay device is being developed to perform on-line fissile material assays of fuel rods produced in a reference HTGR fuel recycle facility. The measurements will be used to satisfy material accountability requirements and for quality control of fuel rod production.

A schematic diagram of the prompt-neutron assay device is given in Fig. 4. A ^{252}Cf neutron source is positioned in the center of a cylindrical drum about 0.6 m in diameter and 0.6 m in length. Surrounding the neutron source are annular regions of 18.3 mm tungsten; 23.4 mm polyethylene; and 250 mm heavy water, D_2O ; respectively. The tungsten shields source gammas and moderates through inelastic scattering the high-energy source neutrons. The polyethylene and heavy water further moderate the high-energy source neutrons.

Fig. 4. Cross-Section View of Fuel Rod Nondestructive Assay Device.

A polyethylene shell at the drum surface contains holes for the detector tubes and fuel rod samples. The arrangement of these holes is shown in detail in Fig. 5. In the experimental device only one set of sample-detector holes is used, but a production device will use a series of such channels at equal angular spacings around the drum for multiple fuel rod assays. From analysis of multiple simultaneous fuel rod assay experiments, an estimated 12 fuel rods could be assayed simultaneously in a production device with only small corrections to be applied for interference between adjacent channels. Thus, the channels will be spaced about 30° apart on the production device.

The detectors are Reuter-Stokes ^4He fast-neutron detectors with an active length of 152.4 mm, a diameter of 38.1 mm, and an anode diameter of 25 μm . The ^4He detectors are relatively insensitive to gamma radiation and the discriminator level is set sufficiently high to reject gamma pileup from source gammas or gammas originating from ^{233}U samples. The two detectors at each fuel rod channel are coupled and detector voltage is set at +2570 V.

Operation of the experimental prompt-neutron assay device is relatively simple. A fuel rod sample (~ 50 mm long and 13 mm diam) is loaded onto a grooved track that leads to the sample hole. A threaded rod driven by a stepping motor advances the sample to the desired position between the detectors. The sample can be precisely positioned (to within 25 μm) at any point along the track. After the sample is located at the correct position, counts are accumulated for a specified time. The sample is then removed and the next sample loaded onto the track. For a sample containing about 0.45 g ^{235}U or ^{233}U , counts

Fig. 5. Sample and Detector Positions in the Fuel Rod Assay Device.

collected from detection of ^{252}Cf neutrons are approximately equal to counts collected from detection of fission neutrons released by the sample.

More information on the prompt-neutron assay machine is given in ref. 2.

SAMPLE PREPARATION

Eleven capsules containing various mixtures of the fissile uranium isotopes ^{233}U and ^{235}U from 0% ^{233}U to nearly 100% ^{233}U were prepared by the procedure described in this section. These samples were then assayed in the prompt- and delayed-neutron assay devices as described in the experimental results section of this report.

Two different batches of loaded resin beads, one containing primarily ^{233}U and the other containing primarily ^{235}U were used to prepare the 11 sample mixtures. The chemical composition and the uranium isotopic composition of each batch of particles are given in Tables 1 and 2, respectively. The average diameters were about 650 μm for the particles containing ^{235}U and about 550 μm for the ^{233}U -bearing particles; the density of the particles was about 1.7 Mg/m^3 .

Table 1. Composition of Loaded Resin Beads

Primary Uranium Isotope	Content, wt %			
	Uranium	Hydrogen	Oxygen	Carbon
^{233}U	51.07	3.25	28.93	15.85
^{235}U	45.99	3.59	32.92	17.50

Table 2. Uranium Isotopic Composition of Loaded Resin Beads

Primary Uranium Isotope	Content, wt %				
	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
^{233}U	98.022	1.191	0.078	0.017	0.691
^{235}U	0.0003	0.991	93.033	0.436	5.540

Aluminum test capsules to contain the particles were fabricated according to the capsule design shown in Fig. 6. The capsules were loaded with a homogeneous mixture of ^{233}U and/or ^{235}U resin beads with sufficient glass beads to fill the entire volume of the sample region. (Keeping the volume of the mixture the same in each capsule is important to avoid neutron self-shielding problems, which are discussed later.) The uranium-bearing particles were weighed out and mixed with glass beads until 1.55 cm^3 of the total material was present. This mixture was vigorously shaken, and successive two-way equal random splits of the material were performed until eight equal batches were obtained. Each of these batches was poured, one at a time, into the test capsule, and the capsule's aluminum top was inserted and sealed. The 11 test capsule loadings are listed in Table 3 along with the background-corrected counts obtained when the samples were irradiated in the two assay devices.

Fig. 6. Encapsulated Test Sample.

Table 3. Test Capsule Loadings and Background-Corrected Counts

Sample	Ratio ^{233}U		Content, g		Neutron Counts ^a	
	^{235}U	^{233}U	^{233}U	^{235}U	Delayed (counts/90 s)	Prompt (counts/300 s)
1	0.00	0.	0.3000		86,033 ± 170	224,485 ± 534
2	1.00	0.1500	0.1500		68,218 ± 151	219,721 ± 738
3	1300	0.3000	0.0002		50,604 ± 130	217,678 ± 531
4	0.00	0.	0.4000		110,940 ± 193	287,064 ± 783
5	0.33	0.1000	0.3000		98,371 ± 181	281,788 ± 779
6	1.00	0.2000	0.2000		89,306 ± 173	286,284 ± 553
7	3.00	0.3000	0.1000		77,823 ± 161	284,259 ± 781
8	1300	0.4000	0.0003		63,285 ± 146	276,866 ± 776
9	0.00	0.	0.5000		127,491 ± 206	335,350 ± 813
10	1.00	0.2500	0.2500		105,960 ± 188	341,024 ± 816
11	1300	0.5000	0.0004		80,055 ± 164	340,286 ± 816

^a ±1 standard deviation.

X-ray radiographs of the filled capsules were taken and are shown in Fig. 7. In this figure the denser materials show as dark areas. As can be seen, the distributions of uranium particles in the capsules are generally uniform.

EXPERIMENTAL RESULTS

The 11 capsules were irradiated and counted in the delayed-neutron assay device on June 26, 1978. Each capsule was assayed three times. Each assay consisted of an irradiation period of 90 s, a decay period of 4.583 s, and a counting period of 90 s. The average of the three measurements for each capsule is given in Table 3, the counts having been corrected for background. The background was about 200 counts in a 90-s counting interval.

The capsules were irradiated and counted in the prompt-neutron assay machine on June 29, 1978. Each capsule was assayed once for 300 s except for capsules 1, 3, and 6, which were assayed twice for verification. The background-corrected counts for each capsule are listed in Table 3. Fifteen background measurements were made, the average of which was 305,075 counts. The background is due to detection of high-energy neutrons that have penetrated the moderator materials from the ^{252}Cf neutron source.

Photo 0546-79

Fig. 7. X-Radiographs of Test Samples (Samples 1 to 11 from left to right).

The background-corrected counts are plotted against total sample fissile content in Figs. 8 and 9 for the delayed- and prompt-neutron results, respectively. Notice that although large variations in delayed neutron counts are obtained as the $^{233}\text{U}/^{235}\text{U}$ ratio varies, the prompt-neutron counts are relatively insensitive to this ratio.

Fig. 8. Delayed-Neutron Counts Versus Sample Fissile Content.

Fig. 9. Prompt-Neutron Counts Versus Sample Fissile Content.

ANALYSIS OF RESULTS

Derivation of Calibration Equation

In this section, an equation is derived from basic physical considerations relating measured counts to ^{233}U and ^{235}U contents.

Let C_3 = counting rate from ^{233}U in the sample and

C_5 = counting rate from ^{235}U in the sample.

Then,

$$C_3 = \sigma_{f_3} E_3 f_3 v_3 U_3 \bar{\phi} = \alpha_3 U_3 \bar{\phi} \quad (1a)$$

$$C_5 = \sigma_{f_5} E_5 f_5 v_5 U_5 \bar{\phi} = \alpha_5 U_5 \bar{\phi} , \quad (1b)$$

where

$\bar{\phi}$ = average neutron flux in the sample,

σ_{fi} = fission cross section of uranium isotope i ,

E_i = detector efficiency for fission neutrons from isotope i ,

f_i = particle self-shielding factor for isotope i ,

v_i = number of delayed or prompt neutrons released per fission for isotope i ,

$\alpha_i = \sigma_{fi} E_i f_i v_i = \text{a constant,}$

U_i = uranium content for isotope i .

(Note that isotope 3 is ^{233}U and isotope 5 is ^{235}U .)

$$C = \text{total counting rate} = C_3 + C_5 = \bar{\phi}(\alpha_3 U_3 + \alpha_5 U_5) \text{ either prompt or delayed neutrons.} \quad (2)$$

The form of Eq. (2) is valid for either prompt- or delayed-neutron counting. The average flux and the constants α_3 and α_5 will be different for the two.

It is necessary to estimate the average neutron flux in the sample. If we assume that the sample is purely absorbing and all neutrons passing through the sample traverse the average chord length of the sample, Nisle's self-shielding approximation is obtained.^{3,4}

$$\bar{\phi} \approx \frac{\phi_0}{\text{Rav} \Sigma_a} [1 - \exp(-\Sigma_a \text{Rav})] \quad (3)$$

and

$$\Sigma_a \approx \Sigma_{a3} + \Sigma_{a5} = \sigma_{a3} U_3 / V + \sigma_{a5} U_5 / V, \quad (4)$$

where

ϕ_0 = neutron flux in a nonabsorbing sample,

Rav = average chord length of the sample,

σ_{ai} = absorption cross section of isotope i ,

V = volume of the sample.

Substituting Eqs. (3) and (4) into Eq. (2), Eq. (5) is obtained:

$$C = \frac{\phi_0 V}{\text{Rav}} \frac{(\alpha_3 U_3 + \alpha_5 U_5)}{(\sigma_{a3} U_3 + \sigma_{a5} U_5)} \{1 - \exp[-(\text{Rav}/V)(\sigma_{a3} U_3 + \sigma_{a5} U_5)]\}. \quad (5)$$

Note that the total counting rate given by Eq. (5) depends on the geometry of the sample through Rav and V . Equation (5) can be simplified to Eq. (6) if the sample geometry is the same for all samples:

$$C = \left(\frac{A_3 U_3}{U_3 + RU_5} + \frac{A_5 U_5}{U_3 + RU_5} \right) \{1 - \exp[-D(U_3 + RU_5)]\}. \quad (6)$$

Equation (6) has four constants — A_3 , A_5 , R , and D — which are different for delayed-neutron counting than for prompt-neutron counting. Equation (6) is the equation to be solved simultaneously for ^{233}U and ^{235}U contents given the prompt- and delayed-neutron counts.

If λ_3 is set equal to zero in Eq. (6), it reduces to Eq. (7). The form of Eq. (7) is also given by Forster et al.⁵ as a calibration equation to be used for assay of ^{235}U -containing fuel rods by a neutron irradiation and neutron detection assay method.

$$C = (A_5/R)[1 - \exp(-TRU_5)] \quad (7)$$

Determination of Calibration Constants

The four constants in Eq. (6) were determined by a Gauss-Newton nonlinear least squares fit⁶ to the data given in Table 3, excluding the measured counts for capsules 5 and 7. These samples were assumed to have unknown uranium contents. The calculated constants are given in Table 4 for both the prompt- and delayed-neutron results.

Table 4. Constants for Eq. (6) Obtained by Nonlinear Least Squares Fit to Data in Table 3 Excluding Capsules 5 and 7

Constant	Prompt Neutrons	Delayed Neutrons
A_3 , counts	1,388,356	380,922
A_5 , counts	1,595,862	748,838
R	2.1315	2.7672
D , g ⁻¹	0.56247	0.46518

The counts calculated with Eq. (6) and the constants given in Table 4 are listed in Table 5. Counts calculated with Eq. (6) and the measured counts agree well, even for samples 5 and 7, which were not included in estimation of the calibration constants. The maximum percentage difference between measured and calculated counts is 2.3% for delayed-neutron counts and 1.2% for prompt-neutron counts. The good agreement indicates that Eq. (6) is a reasonable selection for the calibration equation.

Table 5. Comparison Between Measured Counts and Counts Calculated with Eq. (6) and Constants Given in Table 4

Sample	Delayed-Neutron Counts			Prompt-Neutron Counts		
	Measured	Calculated	$1 - \frac{\text{Calculated}}{\text{Measured}}$	Measured	Calculated	$1 - \frac{\text{Calculated}}{\text{Measured}}$
1	86,033	86,690	-0.0076	224,485	226,180	-0.0076
2	68,218	69,322	-0.0162	219,721	221,265	-0.0070
3	50,604	49,676	+0.0183	217,678	215,713	+0.0090
4	110,940	108,905	+0.0183	287,064	285,216	+0.0064
5	98,371	99,215	-0.0086	281,788	284,193	-0.0085
6	89,306	88,665	+0.0072	286,284	282,949	+0.0116
7	77,823	77,179	+0.0083	284,259	281,464	+0.0098
8	63,285	64,759	-0.0233	276,866	279,911	-0.0110
9	127,491	128,437	-0.0074	335,350	337,583	-0.0067
10	105,960	106,386	-0.0040	341,024	339,433	+0.0047
11	80,055	79,155	+0.0112	340,286	340,589	-0.0009

Estimation of ^{233}U and ^{235}U Contents in Samples by Simultaneously Solving the Calibration Equations

Calibration equations, one for prompt-neutron assay and the other for delayed-neutron assay, were derived and the calibration constants determined in the preceding paragraphs. Each calibration equation is of the form of Eq. (6). Equation (6) for prompt- or delayed-neutron counts is a nonlinear function of ^{233}U and ^{235}U contents of the sample. If the prompt- and delayed-neutron counts for a sample are taken, the two calibration equations can be solved simultaneously for ^{233}U and ^{235}U contents. An iterative procedure is required. One that converges very rapidly and appears to be stable is an extension of Newton's method⁷ for solving nonlinear equations.

Let

$$f(x, y) = D_m - D(x, y) \quad , \quad (8b)$$

and

$$f(x, y) = P_m - P(x, y) \quad (8a)$$

where

- P_m = measured prompt-neutron counts,
 D_m = measured delayed-neutron counts,
 $F(x, y)$ = calibration equation of the form of Eq. (6)
 for prompt-neutron counts,
 $D(x, y)$ = calibration equation of the form of Eq. (6)
 for delayed-neutron counts,
 x = ^{233}U content,
 y = ^{235}U content.

When both $f(x, y)$ and $g(x, y)$ are equal to zero, then a solution is found for x and y that will yield the measured delayed- and prompt-neutron counts.

If $f(x, y)$ and $g(x, y)$ are expanded in a Taylor Series and set equal to zero, Eqs. (9a) and (9b) are obtained.

$$f(x_2, y_2) \approx f(x_1, y_1) + f_x(x_1, y_1)(x_2 - x_1) + f_y(x_1, y_1)(y_2 - y_1) = 0 \quad (9a)$$

$$g(x_2, y_2) \approx g(x_1, y_1) + g_x(x_1, y_1)(x_2 - x_1) + g_y(x_1, y_1)(y_2 - y_1) = 0 \quad (9b)$$

where

- x_1 = first estimate of ^{233}U content,
 y_1 = first estimate of ^{235}U content,
 x_2 = second estimate of ^{233}U content,
 y_2 = second estimate of ^{235}U content,
 f_x = partial derivative of $f(x, y)$ with respect to x ,
 f_y = partial derivative of $f(x, y)$ with respect to y ,
 g_x = partial derivative of $g(x, y)$ with respect to x ,
 g_y = partial derivative of $g(x, y)$ with respect to y .

Equations (9a) and (9b) are linear and can be solved directly for x_2 and y_2 . Iteration continues until the desired convergence on the estimated uranium contents is obtained.

Two samples were considered to be unknowns, and the data from these samples were not included in the determination of the calibration constants of Eq. (6). Using the measured prompt- and delayed-neutron counts of these samples, Eqs. (9a) and (9b) are iterated and the ^{233}U and ^{235}U contents of these samples estimated. Table 6 lists the results of these iterations. As can be seen, the iterative procedure converges very rapidly.

Table 6. Results of Iterative Procedure as Applied to Samples 5 and 7

Iteration	Content in Sample 5, g		Content in Sample 7, g	
	^{233}U	^{235}U	^{233}U	^{235}U
1	0.2000	0.2000	0.2000	0.2000
2	0.1027	0.2931	0.3087	0.0960
3	0.0991	0.2967	0.3046	0.1000
4	0.0991	0.2967	0.3046	0.1000

Estimated ^{233}U and ^{235}U contents and those contained within the capsules are compared in Table 7. The maximum percentage difference between actual and estimated contents of ^{233}U or ^{235}U in the two samples is 1.5%.

Table 7. Comparison Between Estimated and Actual ^{233}U and ^{235}U Contents in Samples 5 and 7

	^{233}U Contents			^{235}U Contents		
	Actual (g)	Estimated (g)	$1 - \frac{\text{Estimated}}{\text{Actual}}$	Actual (g)	Estimated (g)	$1 - \frac{\text{Estimated}}{\text{Actual}}$
5	0.1000	0.0991	0.0090	0.3000	0.2967	0.0110
7	0.3000	0.3046	-0.0151	0.1000	0.1000	0.0

APPLICATIONS

For material accountability requirements and for certain quality control applications in nuclear facilities that handle mixed ^{233}U - ^{235}U fuels, measurement of ^{233}U and ^{235}U in feed material, product, and waste is required. Certain nondestructive measurement techniques using neutron interrogation and neutron detection, as those described in this report, work well for ^{233}U fuels since measurements obtained with such techniques are not significantly affected by gamma radiation originating from ^{233}U and ^{232}U or their decay products. Many other nondestructive techniques, such as passive calorimetry or gamma signature methods, would be highly affected by decay of ^{232}U and its decay products. Chemical analysis methods could be used, but measurement time would be much longer than for nondestructive methods and each container would have to be opened for sampling. Also, representative sampling would be difficult with chemical analysis methods. The techniques described in this report can be used to determine individual ^{233}U and ^{235}U contents of mixed ^{233}U - ^{235}U fuels and would therefore be very useful in nuclear facilities that handle such mixed fuels. The precision of this isotopic assay method, which is discussed in detail in the appendix, depends on the $^{233}\text{U}/^{235}\text{U}$ mass ratio and the net prompt- and delayed-neutron counts.

As an application example, consider a fuel recycle facility that produces HTGR fuel rods containing ^{233}U and ^{235}U . Assume that an on-line prompt-neutron assay machine measures the total fissile content in each fuel rod for quality control and material accountability. As the fissile content measurement provided by the prompt-neutron assay machine depends somewhat on the $^{233}\text{U}/^{235}\text{U}$ ratio, samples will be regularly sent to a sample inspection station. At the sample inspection station the samples will be assayed in a delayed-neutron assay machine. Delayed- and prompt-neutron counts will be compared, and the $^{233}\text{U}/^{235}\text{U}$ ratio will be estimated by use of a calibration equation, such as Eq. (6). This estimate will be used with the prompt-neutron measurements for accurate determination of total fissile content in each fuel rod.

Assume that the $^{233}\text{U}/^{235}\text{U}$ ratio is regularly estimated to within about 5%. The total fissile content, as provided by measurements with the prompt-neutron assay machine, will be biased somewhat by the error in the $^{233}\text{U}/^{235}\text{U}$ ratio. The magnitude of the bias can be readily estimated by rewriting Eq. (6), the calibration equation:

$$U = -\frac{1+S}{D(S+R)} \ln \left[1 - \frac{C(S+R)}{SA_3 + A_5} \right], \quad (10)$$

where

S = $^{233}\text{U}/^{235}\text{U}$ ratio,

U = total fissile content = $^{233}\text{U} + ^{235}\text{U}$, g,

A_3, A_5, D , and R = calibration constants for prompt-neutron assay.

Assume that $C = 286,284$, $S_1 = 0.95$, $S_2 = 1.00$, $S_3 = 1.05$ and A_3 , A_5 , D , and R have the values given in Table 4. The estimated total fissile contents are 0.40556, 0.40567, and 0.40577 g for the various values of the $^{233}\text{U}/^{235}\text{U}$ ratio, respectively. The percentage variation in the estimated fissile content as provided by the prompt-neutron assay machine is about 0.05% for a 10% variation in the $^{233}\text{U}/^{235}\text{U}$ ratio. This is a negligible bias and indicates that the above procedure for measurement of total fissile content of fuel rods in a fuel recycle facility handling ^{233}U - ^{235}U fuels is acceptable.

CONCLUSIONS

Fuel samples containing mixtures of ^{233}U and ^{235}U can be accurately assayed by nondestructive methods employing neutron interrogation and counting delayed and prompt fission neutrons. The number of delayed neutrons per fission is significantly less for ^{233}U

than for ^{235}U . Measuring both the delayed neutrons and prompt neutrons provides estimates of the ^{233}U and ^{235}U contents.

An experiment was performed for 11 mixtures of ^{233}U and ^{235}U over the entire range of $^{233}\text{U}/^{235}\text{U}$ ratios. Constants in a calibration equation, given by Eq. (6), were fitted to data from nine of the samples. The maximum percentage difference between measured counts and those calculated with the calibration equation and the fitted constants was 2.3% for delayed neutrons and 1.2% for prompt neutrons. Contents of the two remaining samples were assumed to be unknown and were not used in the fitting procedure for the calibration constants. The counts obtained for these samples were inserted into the calibration equations for prompt- and delayed-neutron counting, and the resulting equations were solved simultaneously for ^{233}U and ^{235}U sample contents. The maximum percentage difference in estimated ^{233}U or ^{235}U content from the actual capsule contents for either sample was 1.5%.

The results of the ^{233}U - ^{235}U mixture experiment indicate that ^{233}U and ^{235}U can be rapidly and accurately estimated by neutron irradiation and delayed- and prompt-neutron detection. The analysis procedure described in this report can be used in ^{233}U - ^{235}U fuel production facilities to partially satisfy material accountability and quality control requirements.

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APPENDIX

Precision of Isotopic Assay Method

Precision of Isotopic Assay Method

This appendix estimates the precision of the isotopic assay technique described in the body of the report. First, we assume that the net response of both the delayed-neutron assay device and the prompt-neutron assay device can be written as a linear function of the isotopic mass contents of samples. This relation is expressed by

$$C_D = a_3 U_3 + a_5 U_5 , \quad (A1)$$

$$C_P = b_3 U_3 + b_5 U_5 , \quad (A2)$$

where

$$\begin{aligned} C_D &= \text{net delayed-neutron counts,} \\ C_P &= \text{net prompt-neutron counts,} \\ U_3 &= {}^{233}\text{U content of sample,} \\ U_5 &= {}^{235}\text{U content of sample,} \\ a_3, a_5, b_3, b_5 &= \text{calibration constants.} \end{aligned}$$

Equations (A1) and (A2) can be solved for ${}^{233}\text{U}$ and ${}^{235}\text{U}$ contents as follows.

$$U_3 = \frac{C_D b_5 - C_P a_5}{a_3 b_5 - b_3 a_5} , \quad (A3)$$

$$U_5 = \frac{C_D b_3 - C_P a_3}{a_5 b_3 - b_5 a_3} , \quad (A4)$$

By use of propagation of error formalism,⁸ an equation is written that relates the variance of ^{233}U and ^{235}U determination to the variance of the responses of the two assay devices:

$$\sigma^2(U_i) = \left(\frac{\delta U_i}{\delta C_D} \right)^2 \sigma^2(C_D) + \left(\frac{\delta U_i}{\delta C_P} \right)^2 \sigma^2(C_P) , \quad (\text{A5})$$

where

$\sigma^2(U_i)$ = variance of the i th nuclide determination,

$\sigma^2(C_D)$ = variance of the response of the delayed-neutron assay device,

$\sigma^2(C_P)$ = variance of the response of the prompt-neutron assay device.

Here, it is noted that responses of the two devices are independent; thus no covariance terms appear in Eq. (A5). By using Eqs. (A3) and (A4) to calculate the derivatives and by performing some algebraic manipulations, one can obtain the following relationships:

$$\frac{\sigma(U_3)}{U_3} = K_1 \left\{ \left[\frac{\sigma(C_D)}{C_D} \right]^2 \left[\frac{a_3}{a_5} + \frac{U_5}{U_3} \right]^2 + \left[\frac{\sigma(C_P)}{C_P} \right]^2 \left[\frac{b_3}{b_5} + \frac{U_5}{U_3} \right]^2 \right\}^{1/2} \quad (\text{A6})$$

$$\frac{\sigma(U_5)}{U_5} = K_2 \left\{ \left[\frac{\sigma(C_D)}{C_D} \right]^2 \left[\frac{a_5}{a_3} + \frac{U_3}{U_5} \right]^2 + \left[\frac{\sigma(C_P)}{C_P} \right]^2 \left[\frac{b_5}{b_3} + \frac{U_3}{U_5} \right]^2 \right\}^{1/2} \quad (\text{A7})$$

where

$$K_1 = (b_3/b_5 - a_3/a_5)^{-1},$$

$$K_2 = (b_5/b_3 - a_5/a_3)^{-1},$$

$\sigma(U_3)/U_3$ = coefficient of variation of ^{233}U assay,

$\sigma(U_5)/U_5$ = coefficient of variation of ^{235}U assay.

After the constants α_3 , α_5 , b_3 , and b_5 are determined for the two assay devices, Eqs. (A6) and (A7) can be used to study the precision of the isotopic assays. Since ^{233}U and ^{235}U emit approximately the same number of prompt fission neutrons when exposed to a thermal neutron flux, b_3/b_5 is expected to be approximately 1. Also, since ^{235}U emits nearly twice as many delayed neutrons as ^{233}U emits, α_3/α_5 will be about 0.5. Using the assay device responses measured in this work, the following ratios were found:

$$\alpha_3/\alpha_5 = 0.594 \ ,$$

$$b_3/b_4 = 0.981 \ .$$

A calculational study was performed to determine how the coefficient of variation of the isotopic determinations responded to various isotopic ratios. Using response precisions of 0.25% for both assay devices (i.e., $\sigma(C_D)/C_D = \sigma(C_P)/C_P = 0.0025$), the results of Fig. A1 were obtained. It is seen that isotopic assays become more precise as isotopic abundance increases, but that they are always less precise than the 0.25% device response precision. In addition, the precision of the assays varied in direct proportion with the assay response precisions. For example, if the assay response precision of Fig. A1 changed from 0.25% to 0.50%, both curves would be shifted upward by a factor of 2.

Fig. A1. Coefficients of Variation of ^{233}U and ^{235}U Measurements for Prompt- and Delayed-Neutron Assay Response Precisions of 0.25%.

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