DP-MS-84-12
Rev 1

URANIUM DENSITY AND ENRICHMENT IN FUEL TUBES DETERMINED FROM ^{232}U AND ^{235}U γ-ACTIVITIES

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A paper proposed for publication in
Nuclear Instruments and Methods

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

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ABSTRACT

Gamma spectroscopy is used to determine ^{235}U density and enrichment in U-Al fuel tubes containing recycled fuel. A collimated HPGe γ-detector views the tube surface, such that U-Al disk volumes of 6.35 mm diameter and ~1.0 mm thickness are examined. The γ-activities from ^{232}U and ^{235}U, along with the tube design parameters, are used to deduce the attenuation-corrected results. Respective density and enrichment variations of <1% and <0.6ε% were measurable with 2000 sec counting time per tube location. Such measurements are useful for certifying tube quality and characterizing problems associated with blending the U-Al alloy.

1. INTRODUCTION

Gamma spectroscopy often has been applied to measure ^{235}U enrichment near the surface of thick fuels, by directly monitoring ^{235}U.[1,2,3] For such fuels, an effectively constant sample thickness is monitored, as defined by the saturated γ-attenuation depth from the fuel surface.[3] Coupled with the assumption that the surface enrichment is representative of the bulk fuel, one can also infer the overall ^{235}U density.

Thin-walled tubular fuel produced at the Savannah River Plant (SRP) does not simulate the thick fuel features described above. Gamma measurements on SRP fuel tubes are subject to varying γ-attenuation effects caused by fluctuations in both annular fuel thickness and ^{235}U density. Consequently, monitoring ^{235}U alone is insufficient. However, considerable ^{232}U activity also exists in SRP fuels, due to reprocessing and recycling. The present work examines how simultaneous monitoring of ^{232}U and ^{235}U γ-activities provides sufficient data for determining both ^{235}U density and enrichment in fuel tubes.

The goals of the measurements are to appraise ^{235}U uniformity and identify possible problems associated with tube production. During production the desired ^{235}U enrichment for the fuel is obtained by blending U_3O_8 of high enrichment with U_3O_8 of low enrichment. The resulting U_3O_8 is then blended with Al powder and configured into billets, which are later extruded into fuel tubes. If either of these blending operations is incomplete, ^{235}U

nonuniformities will appear in the extruded fuel tubes. Non-uniformities in ^{235}U enrichment indicate problems with the U_3O_8-U_3O_8 blending, while nonuniformities in ^{235}U density alone imply problems with U_3O_8-Al blending.

The development of gamma spectroscopy was motivated by drawbacks in earlier SRP methods for the fuel measurements. In the past, elemental uranium uniformity was appraised by x-ray transmission and ^{235}U enrichment was measured by mass spectroscopy, for 6.35 mm diameter disks cut from representative fuel tubes. Compared to these earlier methods, gamma spectroscopy has the advantages of (1) nondestructiveness, (2) online capability, (3) time efficiency, and (4) compact instrumentation.

2. Basis of Method

Gamma spectroscopy measures both ^{235}U enrichment and density uniformity. The ^{235}U enrichment is correlated with the $^{235}U/^{232}U$ isotopic ratio which is proportional to the 186 keV/238 keV gamma ratio. The reason for this correlation is discussed in Section 2.1. The ^{235}U density (mass per tube area) is strongly correlated with the 186 keV gamma ray of the spectrum. Both of these correlations require some gamma attenuation corrections, which are examined in Section 2.2.

2.1 General Correlations

The correlation of the ^{235}U density with the 186 keV gamma detection rate, R, is a charcteristic of the ^{235}U decay scheme and requires no further discussion.[4] However, the correlation of ^{235}U

enrichment with F = R/R' (where R' is the detection rate of the 238 keV gamma from the ^{232}U decay chain) is true only for uranium fuels that are burned up, reprocessed, and blended into reusable fuels, as they are at SRP.

Typical SRP uranium fuels have been reused in reactor irradiations over a number of years. Following each irradiation, the uranium is retrieved for fabrication of new fuel tubes, following chemical reprocessing. As the ^{235}U burns out, the ^{232}U builds in according to the activation/decay sequence shown in Figure 1. Consequently, the $^{235}U/^{232}U$ ratio decreases as ^{235}U enrichment decreases. The impact on gamma spectra for such fuels is illustrated in Figure 2. Here the gamma ratio F corresponding to $^{235}U/^{232}U$ is about 3 times greater for the 77% enrichment fuel, as compared with that of the 42% enrichment fuel. It should be noted that F depends on the buildup of ^{228}Th(1.91y) in the ^{232}U(72y) decay chain. The 238 keV gamma is from ^{212}Pb, which is in equilibrium decay with ^{228}Th.[4,5]

The ^{235}U enrichments of reprocessed U_3O_8 fuel stocks are measured accurately (±0.1e%) at ORNL before they are blended to yield a U_3O_8 fuel with the fuel design enrichment. In blending, high enrichment (ε_H) and low enrichment (ε_L) U_3O_8 components are mixed to produce some intermediate enrichment (ε) for the fuel tube. If χ_H and χ_L are the fractions of high and low enrichment U_3O_8, then

$$\varepsilon = \chi_L \varepsilon_L + \chi_H \varepsilon_H = \chi_L \varepsilon_L + (1-\chi_L)\varepsilon_H \qquad (1)$$

Similarly, the ^{232}U enrichment ε' is related to corresponding ε'_H and ε'_L as

$$\varepsilon' = \chi_L \varepsilon'_L + (1-\chi_L)\varepsilon'_H \tag{2}$$

Upon eliminating χ_L from Equation (1) and (2), we have

$$\varepsilon' = \left[\frac{\varepsilon'_L - \varepsilon'_H}{\varepsilon_L - \varepsilon_H}\right]\varepsilon + \left[\frac{\varepsilon_L \varepsilon'_H - \varepsilon_H \varepsilon'_L}{\varepsilon_L - \varepsilon_H}\right]$$

or

$$\varepsilon' = b\varepsilon + c \tag{3}$$

For a fixed counting geometry, the detection rates for the 186 keV and 238 keV gammas are

$$R = a\varepsilon \tag{4}$$

$$R' = a'\varepsilon'$$

where a and a' are constants. Thus

$$F = R/R' = \frac{a\ \varepsilon}{a'\varepsilon'} = (a/a')\ \frac{\varepsilon}{b\varepsilon + c} \tag{5}$$

which illustrates that F is strongly correlated with ε.

Ideally, the values of a, a', b, and c are determined using samples of feed stocks of known ε and ε'; however, only the values for ε are available. Thus, an alternative approach is used in this work. We normalize Equation (3) to be

$$(a'/a)\ \varepsilon' = (a'/a)\ b\varepsilon + (a'/a)\ c$$

$$\hat{\varepsilon}' = \hat{b}\varepsilon + \hat{c} \tag{6}$$

In Figure 3, the normalized ε' or $\hat{\varepsilon}' = (a'/a)\ \varepsilon' = (R'/R)\ \varepsilon$ is plotted against ε, to demonstrate the validity of Equation (6) for fuels examined in this work. The constants $\hat{b}$ and $\hat{c}$ resulting from this plot are used in the following section, to evaluate

$$\varepsilon/\varepsilon' = (a'/a)\ \frac{\varepsilon}{\hat{b}\varepsilon + \hat{c}} \tag{7}$$

All parameters of Equation (6) are determined except the factor a'/a. It will be appreciated that absolute knowledge of a'/a is unnecessary for the relative deviation analysis used in this work.

Although a' is treated as constant, it is weakly dependent on time due to ^{228}Th buildup. However, if measurements on component samples and corresponding tubes are made at approximately the same time (or properly normalized relative to time), a' is effectively constant. This is the case for the data of Figure 3, as well as other measurements in this work.

2.2 Correlations for Fuel Tubes

In measurements with fuel tubes, effects of attenuation have to be considered. Consider the gamma detection geometry depicted in Figure 4 for a typical tube. The 186 keV gamma detection rate for an unattenuated disk of fuel with area A and thickness dx is dR_u,

$$dR_u = D\varepsilon\rho_U A dx \tag{8}$$

where

D = (186 keV γ's/sec-gm $^{235}U_3O_8$)(186 keV detection efficiency)

ρ_U = (density of U_3O_8 in fuel core, gm/cm^3)

ε = (^{235}U enrichment)

Also note that A is effectively the collimater acceptance area for the 186 keV gamma rays. Including attenuation effects of cladding and fuel core, Figure 4 predicts

$$dR = e^{-\mu_A t_c}\, e^{-\mu_f x} dR_u$$

$$R = \int_o^{t_f} dR(x) = DA\rho_U \frac{\varepsilon}{\mu_f}\left(1-e^{-\mu_f t_f}\right)e^{-\mu_A t_c} \tag{9}$$

where

R = actual attenuated 186 keV γ-detection rate, count/sec

μ_f = atten coeff of 186 keV γ in fuel core, cm^{-1}

μ_A = atten coeff of 186 keV γ in Al clad, cm^{-1}

t_f = fuel core thickness, cm

t_c = fuel clad thicknes, cm

Similarly, for the 238 keV gamma ray, the detection rate is

$$R' = D'A'\rho_U \frac{\varepsilon'}{\mu'_f}(1 - \varepsilon^{-\mu'_f t_f})e^{-\mu'_A t_c} \qquad (10)$$

where ε' is the ^{232}U enrichment and the other primed factors correlate with this gamma energy.

It is desirable to express R and R' in terms of enrichment ε and $^{235}U_3O_8$ mass/area m. Pertinent relationships are given as

$$\begin{aligned} m &= \rho_U t_f \varepsilon \\ y &= \mu_f t_t = (\rho_U g_U + \rho_A g_A)t_f = (g_U + g_A \rho_A/\rho_U)m/\varepsilon \\ y' &= \mu'_f t_f = (\rho_U g'_U + \rho_A g'_U + \rho_A g'_A)t_f = (g'_U + g'_A \rho_A/\rho_U)m/\varepsilon \end{aligned} \qquad (11)$$

where ρ_A is the Al density in the fuel core and the g_U, g_A, ...etc are the mass attenuation coefficients.[6] We also note that $\rho_A/\rho_U = (100-W_U)/W_U$, where W_U is the weight percent of U_3O_8 in the fuel core. Thus, y and y' are both functions of ε, m, and W_U, and we may write

$$\begin{aligned} R &= DA\, m e^{-\mu_A t_c}\left(\frac{1 - e^{-y}}{y}\right) = mr(m,\varepsilon,W_U,t_c) \\ R' &= D'A'm\,(\varepsilon'/\varepsilon)e^{-\mu'_A t_c}\left(\frac{1 - e^{-y'}}{y'}\right) = m(\varepsilon'/\varepsilon)r'(m,\varepsilon,W_U,t_c) \\ F &= R/R' = (\varepsilon/\varepsilon')(r/r') = (\varepsilon/\varepsilon')f(m,\varepsilon,W_U,t_c) \end{aligned} \qquad (12)$$

Here, R is primarily dependent on m, and F is primarily dependent on ε, since ε/ε' is a function of ε per Equation (7).

As mentioned in the preceding section, the unknown factor of a'/a in Equation (7) will have no impact on the relative deviation analysis which is developed below.*

It is desired to use the measured deviations in R and F to deduce corresponding deviations in m and ε; however, deviations in W_U and t_c need to be considered also. Thus we examine

$$\begin{aligned} \Delta R &= \partial R/\partial m \cdot \Delta m + \partial R/\partial \varepsilon \cdot \Delta \varepsilon + \partial R/\partial W_U \cdot \Delta W_U + \partial R/\partial t_c \cdot \Delta t_c \\ \Delta F &= \partial F/\partial m \cdot \Delta m + \partial R/\partial \varepsilon \cdot \Delta \varepsilon + \partial F/\partial W_U \cdot \Delta W_U + \partial F/\partial t_c \cdot \Delta t_c \end{aligned} \tag{13}$$

Calculated partial derivatives for these expressions are summarized in Table 1, for the fuel tubes examined in this work. Note that the tabulated values are expressed as %-deviations in the derivatives and thus do not depend on D, A,...etc. These tabulations indicate that ΔR and ΔF are only weakly dependent on W_U and t_c, as compared with the dominant dependences of ε and m. Thus, these smaller terms are set equal to $0 \pm \sigma_S$ where σ_S is the corresponding uncertainty estimated for possible deviations. The values of σ_S are also given in Table 1, for expressions of ΔR and ΔF in the form

$$\begin{aligned} \Delta R &= \partial R/\partial m \cdot \Delta m + \partial R/\partial \varepsilon \cdot \Delta \varepsilon \pm \sigma_{SR} \\ \Delta F &= \partial F/\partial m \cdot \Delta m + \partial F/\partial \varepsilon \cdot \Delta \varepsilon \pm \sigma_{SF} \end{aligned} \tag{14}$$

The values of $\Delta\varepsilon$ and Δm are readily deduced from these as

$$\begin{pmatrix} \Delta m \\ \Delta \varepsilon \end{pmatrix} = \begin{pmatrix} \partial R/\partial m & \partial R/d\varepsilon \\ \partial F/\partial m & \partial F/\partial \varepsilon \end{pmatrix}^{-1} \begin{pmatrix} \Delta R \pm \sigma_{SR} \\ \Delta F \pm \sigma_{SF} \end{pmatrix} \tag{15}$$

---

* Absolute measurements of ε and m require evaluation of DA and D'A' (a'/a), which may be determined from standard tubes of known ε, m, W_U, and t_c.

The estimates for σ_S assume that m and ε are held constant while both inner and outer clad thicknesses (t_c' and t_c) are allowed to range between 10 and 30 mil†. It is also assumed that the tube wall thickness t is constant. With these restraints, it is seen that $t_f = t - t_c' - t_c$ will also vary, and because W_U depends on t_f for constant m and ε, it thus depends on t_c' and t_c. To express this W_U dependence, note that

$$m = \rho_U \varepsilon t_f = \frac{W_U \chi}{W_U/\rho^*_U + W_A/\rho^*_A} \varepsilon t_f \tag{16}$$

where ρ_U^* and ρ_A^* are 100% theoretical densities and χ is the fraction of theoretical density in the fuel. Solving the above expression for W_U and ΔW_U

$$W_U = \frac{100}{\left(1 + \varepsilon(t - t_c - t_c')\rho^*_A \chi/m - \rho^*_A/\rho_U^*\right)}$$

$$\Delta W_U = \frac{W_U^2}{100} \frac{\varepsilon \rho^*_A \chi}{m} \Delta(t_c' + t_c) \tag{17}$$

$$= \frac{W_U W^*_A}{100} \cdot \frac{\Delta(t_c' + t_c)}{t_f} \quad , \qquad \text{where } W^*_A = W_A + \frac{\rho^*_A}{\rho^*_U} W_U.$$

Thus, the ΔR expression (and a similar one for ΔF) for deviations in W_U and t_c is

$$\Delta R(W_U, t_c) = \frac{\partial R}{\partial W_U} \cdot \Delta W_U + \frac{\partial R}{\partial t_c} \cdot \Delta t_c \tag{18}$$

$$= \left[\partial R/dW_U \frac{W^*_A W_U}{100\ t_f}\right]\Delta t_c' + \left[\partial R/\partial W_U \frac{W^*_A W_U}{100\ t_f} + \partial R/\partial t_c\right]\Delta t_c$$

A uniform weighting of ΔR over the ranges for t_c' and t_c produces the data for calculating σ_{SR}. (Note that the magnitude of the Δt_c coefficient is smaller than $\partial R/\partial t_c$ alone). The value of σ_{SF} is obtained similarly.

† 1 mil = 0.0254 mm.

In addition to σ_S errors, each measurement of ΔF and ΔR has an error due to nuclear counting statistics. These errors are included in the reduction of the Δm and $\Delta \varepsilon$ values.

3. Instrumentation

Each fuel tube to be tested is mounted horizontally using a positioning device available with the cladding thickness monitor.[7] An HPGe detector views selected tube areas via a vertical lead collimator positioned under the tube, with a counting geometry like that shown in Figure 4. A lead plug is inserted in the tube to shield gamma rays from the far side of the tube. Gamma spectra are collected and analyzed using a multichannel analyzer/computer.

The lead collimator is 76.2 mm long and has a 6.35 mm diameter entrance (adjacent to the tube) and a 50.8 mm diameter exit (adjacent to the detector). Little collimator leakage exists with a collimator of this design.[8] A minimum of 75 mm of lead shields the detector from other areas of the tube.

The HPGe detector is an ORTEC Slimline Coaxial Model with a cooled FET preamplifier. It has an active volume of 76.7 cc, a 16.9% efficiency, and resolution of <1 keV for the gammas of interest. Detector bias of 3500 V is provided by an ORTEC Model 459 High Voltage Supply. The detector signals are processed by an ORTEC model 572 amplifier before being collected and analyzed by a 2048 channel NUCLEUS multichannel analyzer/computer. This latter unit permitted straightforward evolution of BASIC programming used in the on-line analysis.

4. Measurements

The instrument examined three different sizes of fuel tubes, with dimensions given in Table 1. Area measurements of ΔR and ΔF were made with respective 1-σ counting accuracies of <0.8% and <2.0%, using 2000 sec count times. For each tube, average values of $\bar{R}$ and $\bar{F}$ were calculated as references for ΔR and ΔF fluctuations. Corresponding $\Delta\varepsilon$ and Δm were calculated relative to tube design enrichment $\bar{\varepsilon}$ and $\bar{m}$ = 100%, using the relations in Table 1. The results are plotted as a function of tube position in Figures 5 and 6.

The resulting $\Delta\varepsilon$ variations about $\bar{\varepsilon}$ are consistent with the corresponding counting error for ε(~0.6 e%), as shown in Figure 5. By contrast, some of the Δm variations are several times larger than that predicted by counting error (~0.9%), as shown in Figure 6. Thus, these measurements imply that U_3O_8-Al blending was not completely uniform.

5. Discussion

The gamma monitor for ^{235}U uniformity is suitable for rapid non-destructive appraisal of SRP fuel tubes. The results of the above tests agree with those from earlier destructive tests, where good U_3O_8-U_3O_8 blending was established, but where poorer U_3O_8-Al blending was noted. In essence, particle segregation problems with U_3O_8-Al blending are more pronounced because of particle size differences in the U_3O_8 and Al powders blended.

Future applications with this monitor are expected to exceed those discussed in this work. At present the monitor has only been

used for measuring deviations about average or design values. However, preliminary tests indicate that absolute measurements of ε and m are possible with comparable accuracy. The monitor is already being adapted to measure ^{235}U enrichment of 1-gallon cans of uranium oxide.[3] Furthermore, the monitor is suitable for appraising high density fissile "hot spots" in fuel tubes.[8] It is anticipated that other applications will also emerge as experience is gained.

6. Acknowledgement

The author wishes to thank D. R. Johnson, W. E. Stewart, and R. L. Frontroth for acquainting him with the need for developing this monitor. He also thanks N. P. Baumann, H. B. Peacock, and D. R. Leader for technical support during the development and testing phases of the study.

The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U.S. Department of Energy.

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TABLE I

Fuel Tube Parameters*

Nominal Tube Data

Tube	W_U %	ε e%	t_c mil	t_f mil	χ %
Inner	62	65.1	20	42	90
Middle	62	60.3	20	43	90
Outer	60	44.0	20	40	90

Partial Derivatives in R and F at Nominal Tube Conditions

Tube	$\partial R/\partial m$ %/%	$\partial R/\partial \varepsilon$ %/ e%	$\partial R/\partial W_U$ %/w%	$\partial R/\partial t_c$ %/mil	$\partial F/\partial m$ %/%	$\partial F/\partial \varepsilon$ %/e%	$\partial F/\partial W_U$ %/w%	$\partial F/\partial t_c$ %/mil
Inner	0.829	0.259	0.042	-0.086	-0.068	3.105	0.003	-0.009
Middle	0.825	0.286	0.043	-0.086	-0.069	3.142	0.003	-0.009
Outer	0.844	0.349	0.040	-0.086	-0.062	3.582	0.003	-0.009

Differentials with Error Terms

Tube	Values (units) are $\Delta R(\%)$, $\Delta F(\%)$, $\Delta m(\%)$, and $\Delta\varepsilon(e\%)$	
Inner	$\Delta R = 0.829\,\Delta m + 0.259\,\Delta\varepsilon \pm 0.361$	$\Delta\varepsilon = 0.320\,\Delta F + 0.026\,\Delta R \pm 0.016$
	$\Delta F = -0.068\,\Delta m + 3.105\,\Delta\varepsilon \pm 0.040$	$\Delta m = -0.100\,\Delta F + 1.199\,\Delta R \pm 0.433$
Middle	$\Delta R = 0.825\,\Delta m + 0.286\,\Delta\varepsilon \pm 0.361$	$\Delta\varepsilon = 0.316\,\Delta F + 0.027\,\Delta R \pm 0.016$
	$\Delta F = -0.069\,\Delta m + 3.142\,\Delta\varepsilon \pm 0.040$	$\Delta m = -0.109\,\Delta F + 1.203\,\Delta R \pm 0.434$
Outer	$\Delta R = 0.844\,\Delta m + 0.349\,\Delta\varepsilon \pm 0.362$	$\Delta\varepsilon = 0.281\,\Delta F + 0.021\,\Delta R \pm 0.014$
	$\Delta F = -0.062\,\Delta m + 3.523\,\Delta\varepsilon \pm 0.040$	$\Delta m = -0.116\,\Delta F + 1.177\,\Delta R \pm 0.424$

* Unit notation: 1 mil = 0.0254 mm

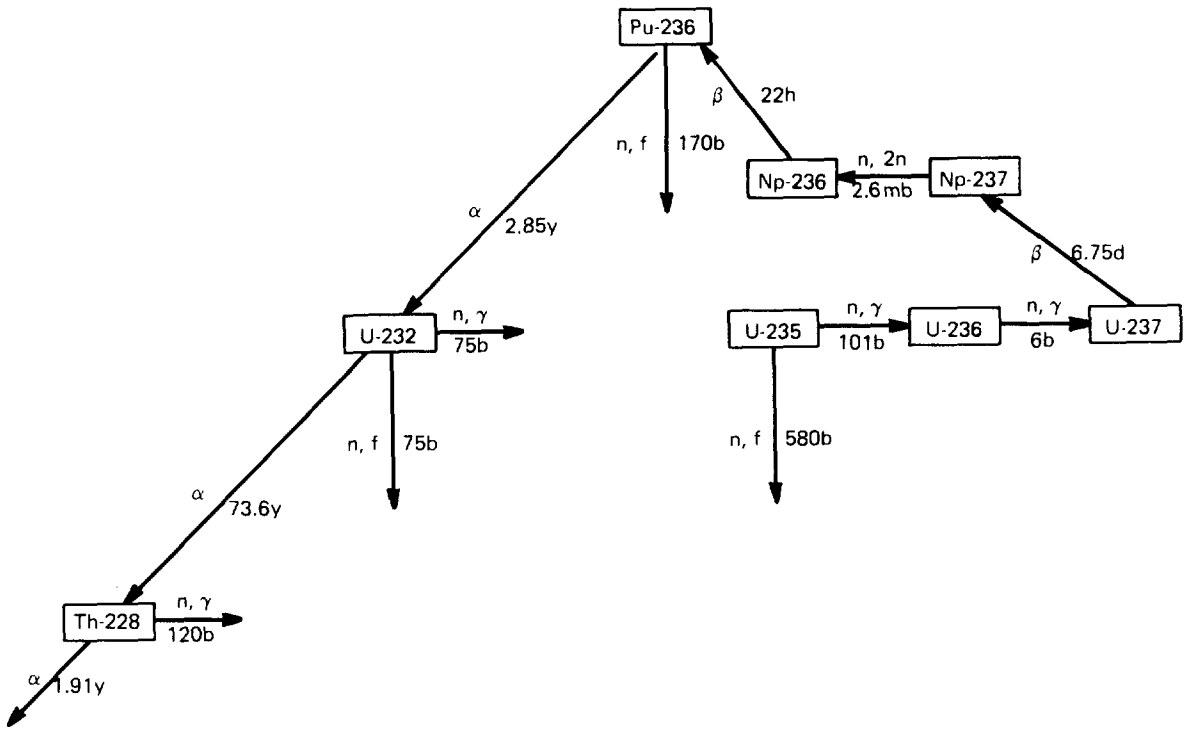

FIGURE 1. Production of ^{232}U during Burnup of ^{235}U

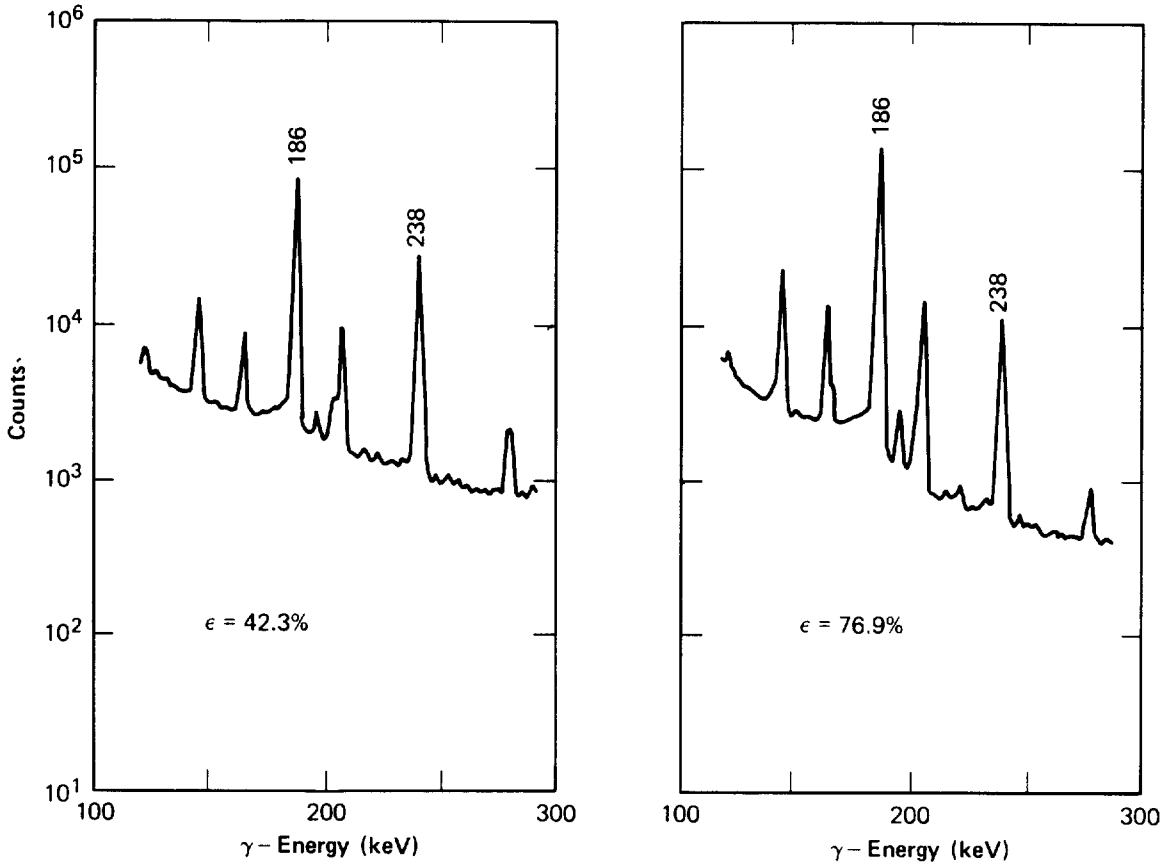

FIGURE 2. γ-Spectra of Typical SRP Fuels

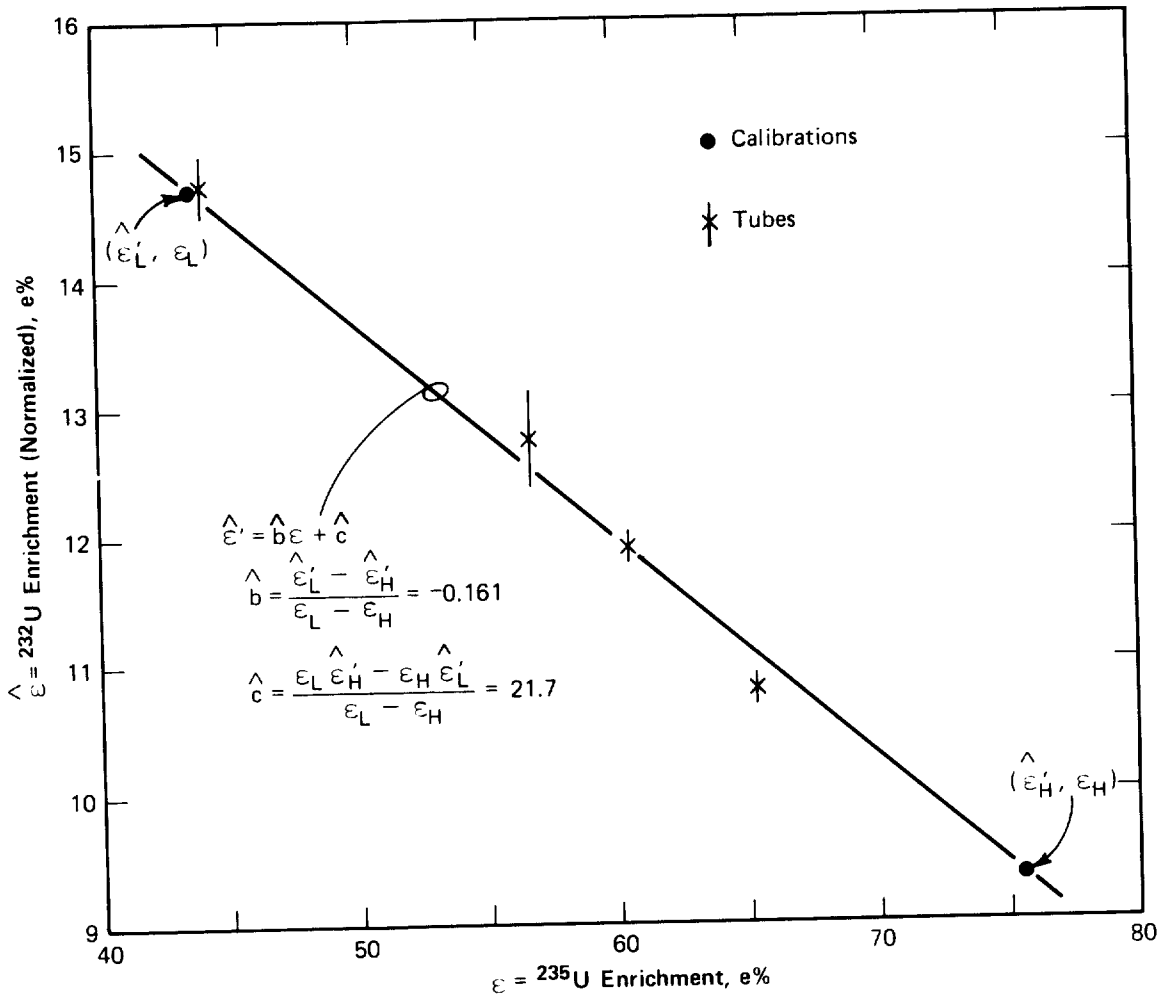

FIGURE 3. Correlation between ^{232}U and ^{235}U

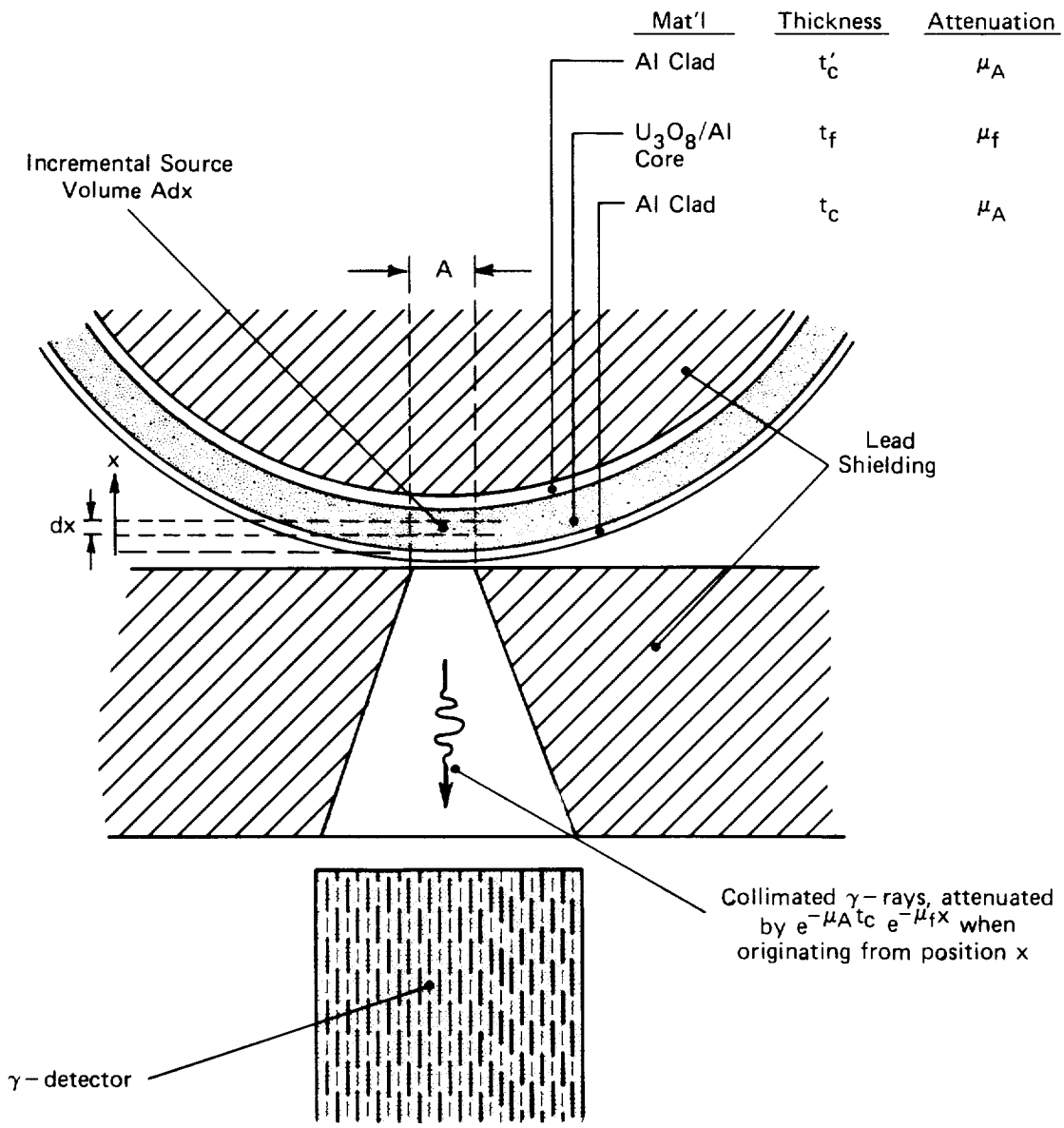

FIGURE 4. Gamma Detection Geometry for Fuel Tubes

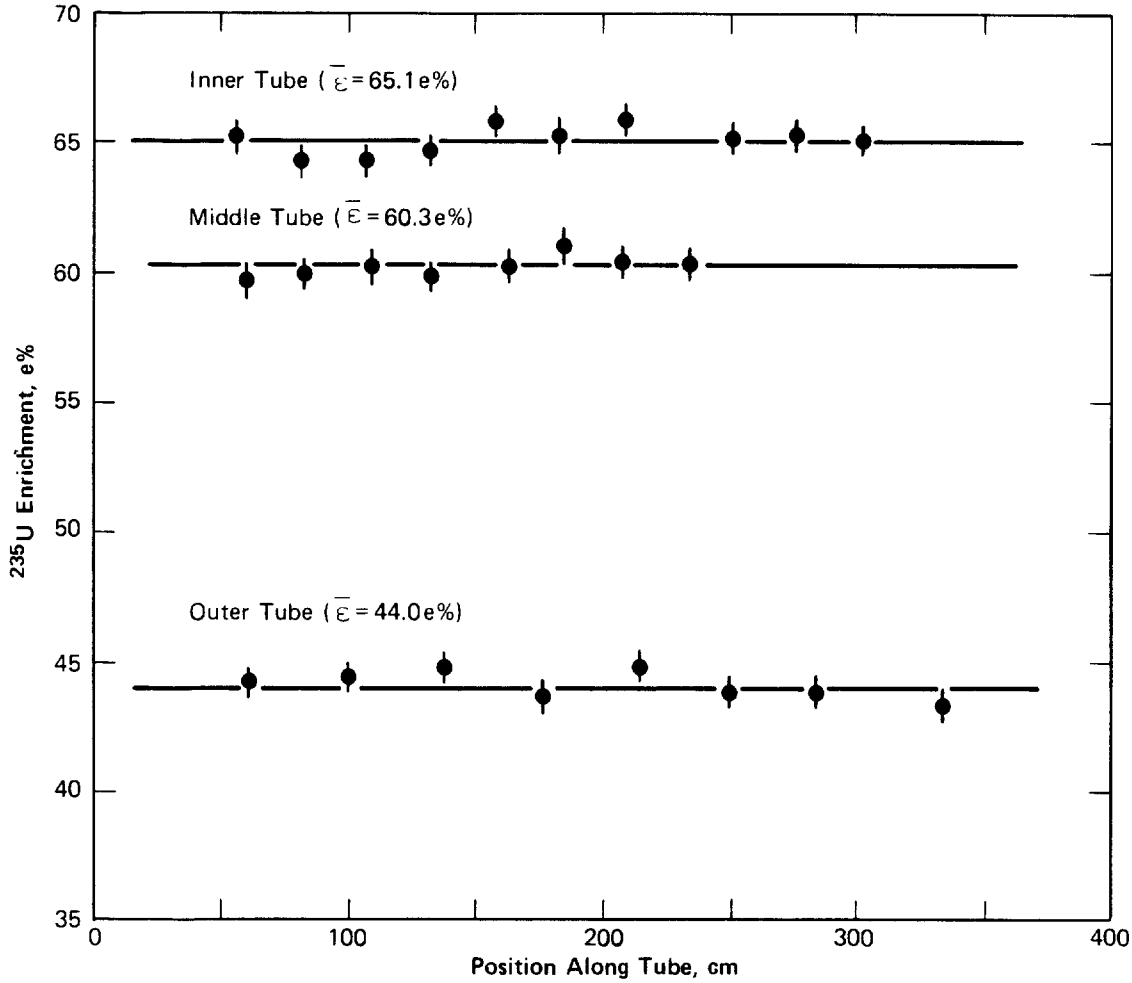

FIGURE 5. Measured ^{235}U Enrichment Fluctuations for Fuel Tubes

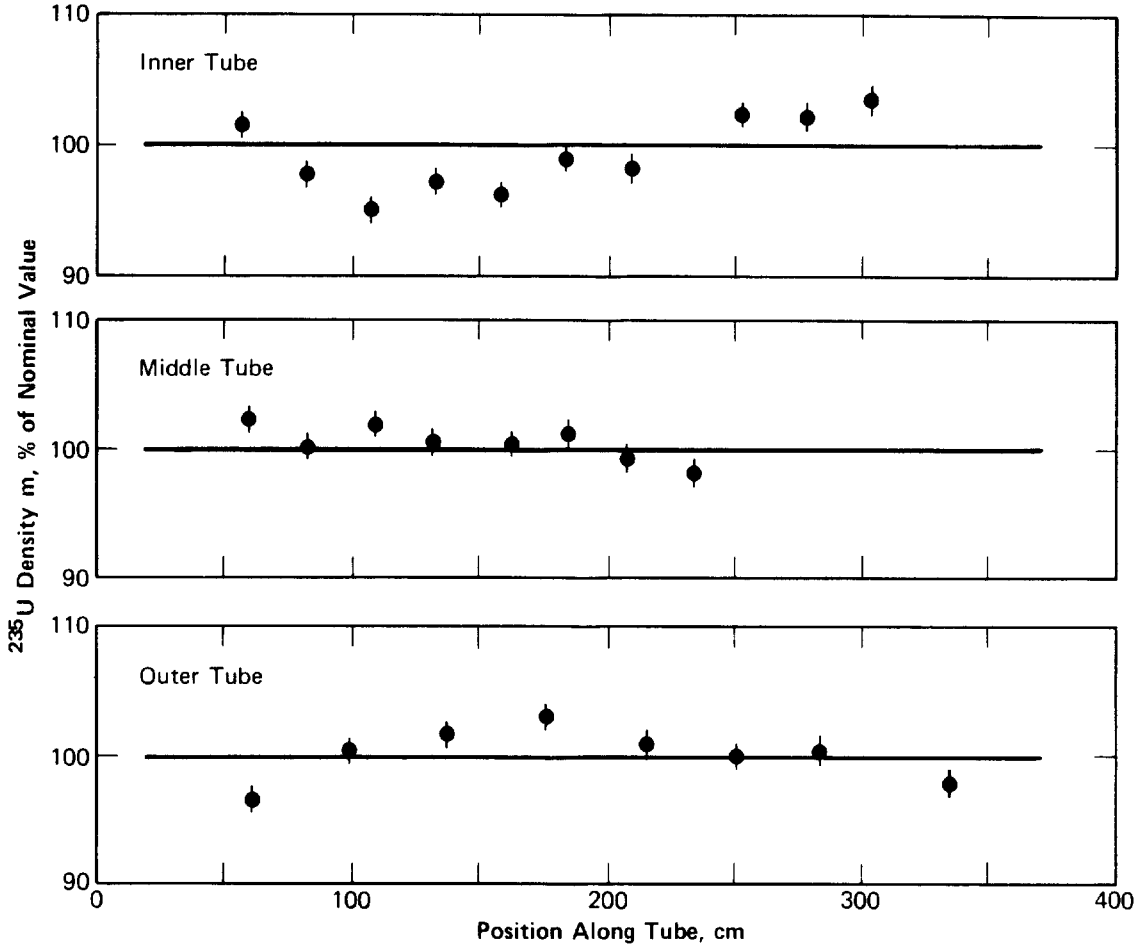

FIGURE 6. Measured ^{235}U Density Fluctuations for Fuel Tubes