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ANALYSIS OF FUEL ELEMENT CORE BLANKS FOR ARGONNE LOW POWER REACTOR BY GAMMA COUNTING

by

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I. INTRODUCTION

A rapid and nondestructive method of uranium analysis is highly desirable and useful during the fabrication phase of reactor fuels. A technique based on a determination of the differential counting rate exhibited by the 184-kev gamma radiation associated with the decay of U^{235} has been developed for the determination of the U^{235} content in Argonne Low Power Reactor fuel element core blanks. The Argonne Low Power Reactor core blanks were an aluminum-highly enriched uranium alloy containing 17.5 weight per cent uranium (approximately 40 gm U^{235}) having the following dimensions: length, 6.875 inches, width, 3.31 inches and thickness, 0.200 inch.

The gamma-ray spectrum emitted by uranium is rather complex. Using a scintillation spectrometer and scanning the spectrum, the energy is found to be concentrated primarily in two regions, at 184 and 90 kev, as shown in Fig. 1. The 184-kev gamma rays result primarily from the

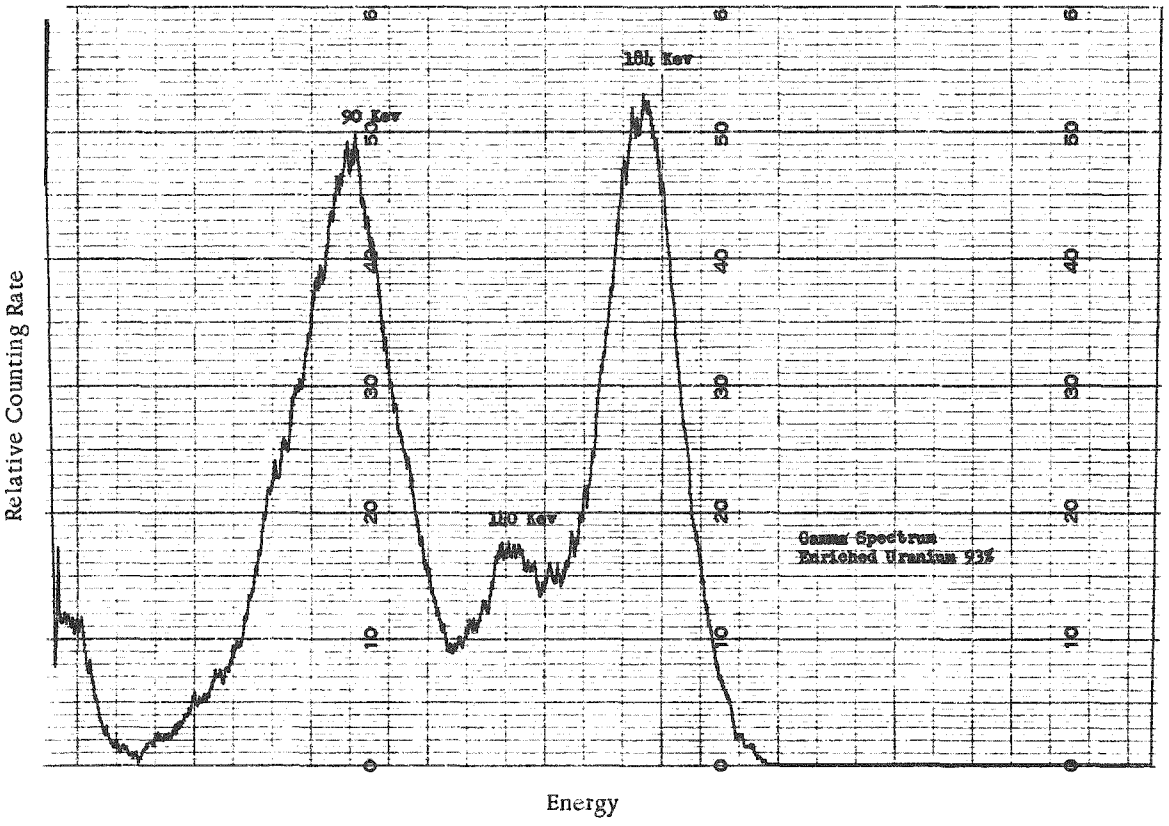

FIG. 1 Gamma-ray Spectrum of Enriched Uranium

decay of U^{235}. The gammas in the 90-kev region result from the U^{235} decay and daughter products of U^{238} and U^{235}. Using a pulse-height analyzer, it is possible to select the desired radiation emitted from the source and determine the counting rate for a given source. In this work the 184-kev gamma radiation was counted to determine the amount of U^{235} present in the individual core blanks.

II. DESCRIPTION OF EQUIPMENT

Figure 2 shows the experimental arrangement used in this work. The gamma radiation is detected by means of a sodium iodide (thallium-activated) crystal, $2\frac{1}{2}$ inches in diameter and 2 inches thick. The crystal was attached to an end window Dumont Type 6363 Photomultiplier tube. The pulse collected across a resistor at the collector anode of the photomultiplier was amplified and fed to a linear amplifier. The output of the linear amplifier was in turn fed to a single-channel pulse-height analyzer. The output of the analyzer could be accumulated for a given period of time on a decade scaler or registered on a count rate meter.

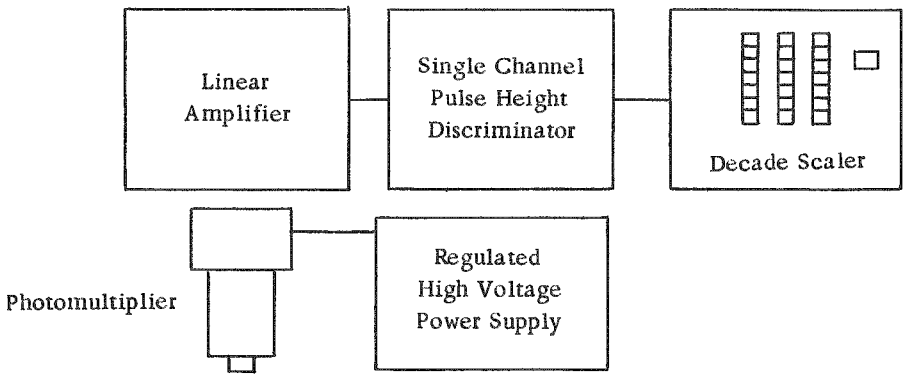

FIG. 2

Schematic Diagram of Equipment for Measuring Uranium-235 Content in ALPR Core Blanks

In this work a single-channel analyzer was used. A pulse-height analyzer is designed to measure the energy spectrum from the detector which gives an output pulse when and only when the amplitude of the input pulse is between E and $E + \Delta E$, where ΔE is known as the window width. Figure 3 shows a block diagram of the analyzer. The analyzer contains a pulse-height selecting feed back amplifier (often called a window amplifier) followed by two pulse height selectors, a memory circuit and an anticoincidence circuit. The window amplifier amplifies a segment of the pulse distribution as shown in Fig. 4. The minimum value that a pulse must have to give an output signal can be arbitrarily set. The lower pulse-height selector is biased to trigger on small impulses from the amplifier.

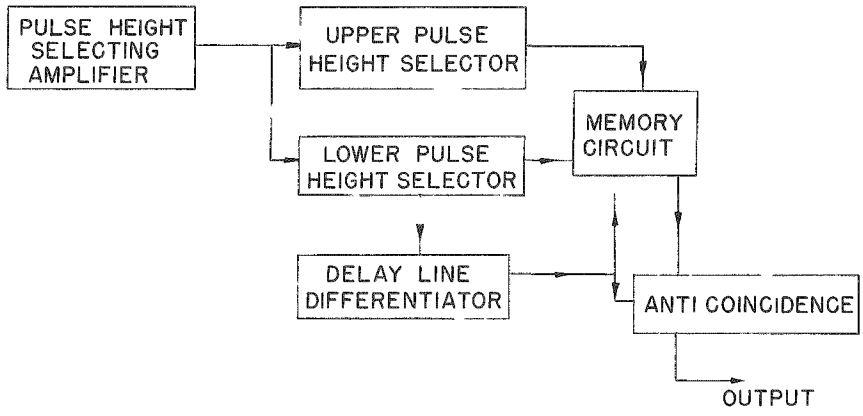

FIG. 3

Block Diagram of Single Channel Analyzer

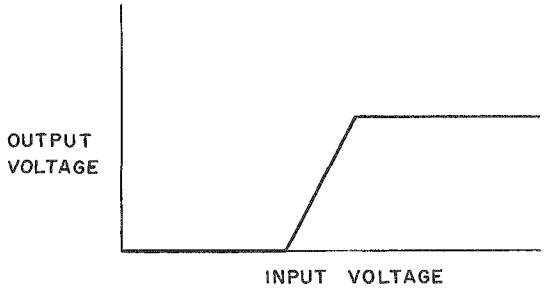

FIG. 4

Input Voltage vs. Output Voltage (Pulse-height Selecting Amplifier)

Two output signals are taken from the pulse-height selector; one is used to trigger the memory circuit, and the other is differentiated by a short delay line to obtain two pulses. The pulse marking the end of the input pulse to the lower pulse selector is used to hold on the memory circuit and is also used to feed to one grid of the anticoincidence circuit. The upper pulse-height selector is triggered only if the output pulse from the window amplifier is above a certain preset value, $E + \Delta E$. The output pulse from this selector is fed to the memory circuit. The difference between the size of the pulse which triggers the lower pulse-height selector and the size of the pulse which triggers the upper pulse-height selector is determined by the window width. The memory circuit provides an input pulse to the anticoincident circuit only when the upper pulse-height selector is triggered. When only the lower selector is triggered, there is no coincidence and therefore an output pulse.

Since the scintillator crystal does not surround the specimen, only a fraction of the total emitted radiation by the specimen impinges on the scintillation crystal. For an absolute determination, the counting rate must be corrected for a "geometry factor," for self-absorption in the specimen, for scattering in the specimen, for the number of incident gamma rays which pass through the scintillator crystal without undergoing any interaction and counting losses in the electronic system.

Self-absorption in the specimen itself is an important factor to be considered in this technique. Although the U^{235} is the primary source of the 184-kev radiation, both the U^{235} and the U^{238} and any alloying material present absorb the radiation. The self-absorption increases as the thickness of the specimen increases. The expression for the self-absorption is

$$N = N_0 \left[\frac{1 - e^{-\mu t}}{\mu t}\right] . \qquad (1)$$

Here N_0 is the number of disintegrations per unit time in the specimen and N is the number of gamma rays which emerge from the specimen; also μ is the linear absorption coefficient of the core material and t is the thickness of the specimen. In the case of alloys, μ is determined by both the absorption coefficient of the uranium and the absorption coefficient of the alloy material.

A number of the gamma rays incident on the scintillator crystal pass through without undergoing any interaction. The number of gammas which do not produce scintillations is determined by the thickness of the crystal and the absorption coefficient of sodium iodide for the incident quantum of radiation. The fraction of the incident gamma rays which are absorbed and produce scintillations is given by the following expression:

$$\text{Fraction Absorbed} = (1 - e^{-\mu' t'}), \qquad (2)$$

where μ' is the linear absorption coefficient of sodium iodide for gamma rays of a given energy and t' is the thickness of the crystal. With the thick crystal used in this work, nearly all of the 184-kev gamma rays are absorbed.

The gamma radiation is scattered both in the specimen and by any materials surrounding or in the neighborhood of the source. By proper shielding of the detector the effect of scatter by the surroundings can be kept to a minimum.

The electronic counting system is not able to count all of the radiation that produces scintillations because the electronic system has a dead time. This means that after an event is registered there is a certain period of time required for the electronic system to prepare itself to register the next event. If two or more events are too close together, they will not be registered as separate events. Consequently, corrections must be made to the observed counting rate. In the single-channel analyzer not only the pulses which have an amplitude $E + \Delta E$, but also the pulses which have an amplitude greater than $E + \Delta E$, can be counted, the latter being sometimes referred to as the "over count." In determining dead time the actual count in the window plus the over count must be determined, because the pulse of amplitude greater than $E + \Delta E$ also causes the amplifier to be inactive for

a certain time after their arrival. The usual expression for determining the loss of count is given by

$$N_0 = \frac{N}{1 - NT}, \tag{3}$$

when N_0 is the true number of events, N is the number of measured events and T is the dead time of the instrument.

The dead time of the instrument can be determined by using the two-source method, in which two nearly identical sources are used. In the method the sum of the measured activities of two single sources is compared with the activity of the sum of the sources. Obviously, the latter is smaller because the relative loss is larger. Let n_1, n_2, and n_{12} be the true number of events per unit time for source 1, source 2, and both sources, respectively, including background; let n_b be the background, and let m_1, m_2, m_{12} and m_b equal the corresponding recorded counting rates. Then,

$$n_1 + n_2 = n_{12} + n_b$$

or

$$\frac{m_1}{1 - m_1T} + \frac{m_2}{1 - m_2T} = \frac{m_{12}}{1 - m_{12}T} + \frac{m_b}{1 + m_bT}, \tag{4}$$

leading to a quadratic equation for T. The development of the solution in powers of $x = m_1 + m_2 - m_{12} - m_b$ yields

$$T = T_1 \left[1 + \frac{T_1}{2}(m_{12} - 3m_b)\right] \tag{5}$$

where

$$T_1 = \frac{x}{2(m_1 - m_b)(m_2 - m_b)} .$$

Because of the small value of x, a difference of nearly equal quantities, considerable accuracy is required for m_1, m_2, and m_{12}. From this experiment the dead time of the system can be calculated.

III. EXPERIMENTAL DATA

Preliminary experiments were made using a group of uranium foils, all of equal area and enrichment but differing in thickness. Data obtained using these foils is given in Table I and plotted as the solid curve of Fig. 5. In Table I, both the thickness of the foils and their masses per cm^2 are given. In Fig. 5 the abscissa is given only in mass per cm^2, which is proportional to the total weight of the uranium-235 present. The ordinate is counts per second. It could be expected that the curve in Fig. 5 would follow a straight line because, as the mass of U^{235} increases, there is a proportional increase in the number of disintegrations occurring per unit time.

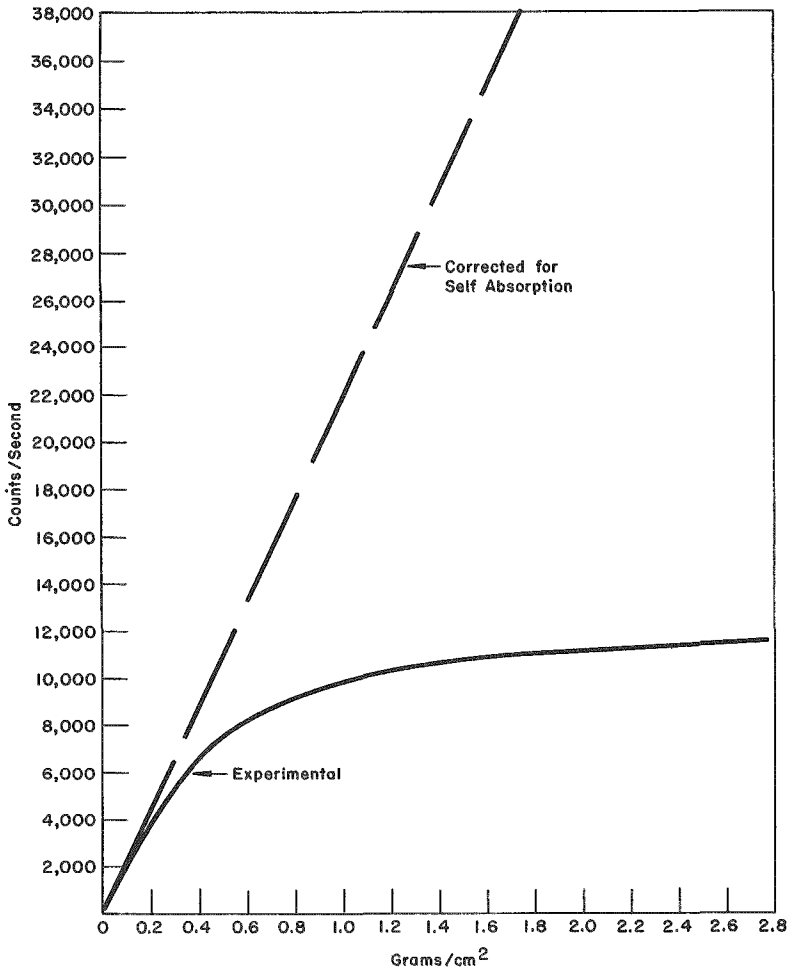

FIG. 5

Counting Rate as a Function of U^{235} Content

The fact that the shape of the experimental curve is different indicates that corrections previously discussed must be made to the counting rate. The dashed curve in Fig. 5 shows the counting rate data corrected only for self-absorption using Eq. 1. The value of the mass absorption coefficient used in these calculations was 1.50 cm^2/gm.* Table II was obtained from Table I by dividing the measured counting rate (solid curve, in Fig. 5), by the mass of the foil. This data is plotted in Fig. 6. This curve shows that as the thickness of the foil increases the greater is the correction for self-absorption and counting loss.

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*G. W. Grodstein, X-ray Attenuation Coefficients from 10 kev to 100 Mev, NBS Circular 583.

Table I

COUNTING DATA FROM URANIUM FOILS

t, gm/cm^2	Thickness, mils	counts/sec	Correction for Self-absorption, counts/sec
0.0518	1.08	1,276	1,340
0.105	2.19	2,432	2,682
0.158	3.29	3,445	3,989
0.210	4.37	4,334	5,257
0.250	5.21	4,898	6,157
0.464	9.66	7,220	10,873
0.714	14.87	8,767	16,044
0.949	19.77	9,575	20,701
1.199	24.98	10,320	26,254
1.663	34.64	10,889	36,010

Table II

COUNTING RATE PER GRAM OF U^{235}

Thickness, mils	t, gm/cm^2	Grams U^{235}	counts/sec/gm
1.08	0.0518	2.22	575
2.19	0.105	4.53	537
3.29	0.158	6.83	504
4.37	0.210	9.16	473
5.21	0.250	10.90	449
9.66	0.464	20.15	358
14.87	0.714	31.05	282
19.77	0.949	41.22	232
24.98	1.199	52.12	198
34.64	1.663	72.27	151
54.43	2.613	113.49	100

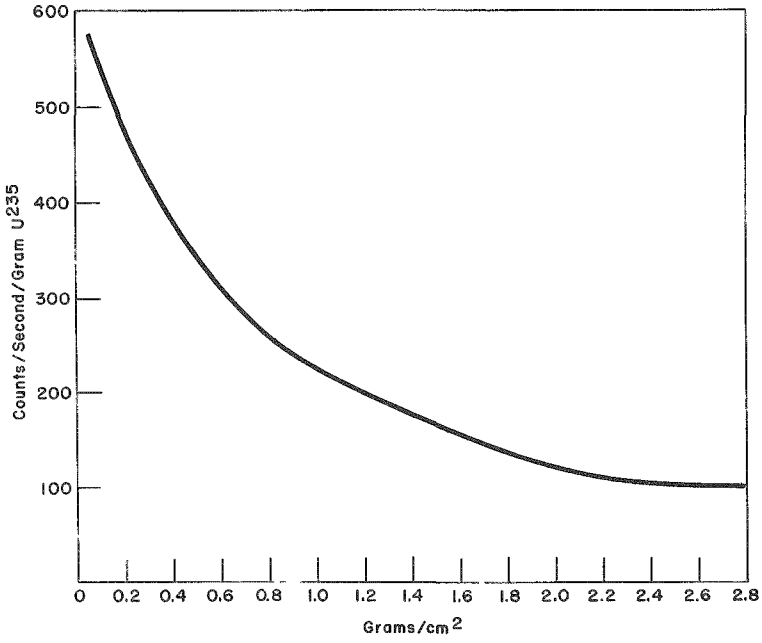

FIG. 6

Counting Rate per Gram Versus Thickness of the Source

Preliminary work to develop a suitable technique for Argonne Low Power Reactor core blanks consisted of counting a large number of core blanks. The scintillator was positioned far enough above the core so that the entire core was counted at one time. Two cores of the same mass giving the same number of counts were found. One of these two core blanks was arbitrarily taken as the standard. The other was analyzed chemically to determine the amount of U^{235} present. Two other cores, one with a counting rate less than the standard and one with a counting rate greater than the standard, were also analyzed chemically. A plot of the counting rates of the three specimens versus the mass of U^{235} was made. Table III shows the counting rates and U^{235} contents for the three cores. These data are plotted in Fig. 7; the calibration curve was linear for this limited range.

Table III

COUNTING RATE VERSUS U^{235} CONTENT

U^{235} Content, gm	Counting Rate, counts/sec
37.452	10,016
38.812	10,200
42.257	10,626

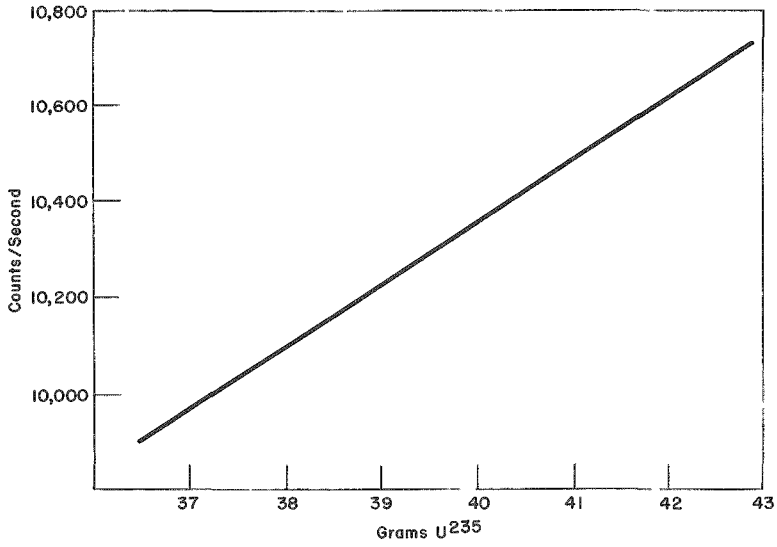

FIG. 7

Calibration Curve
for ALPR Core Blanks

Therefore, the change in the counting rate was directly proportional to the change in U^{235} content, the slope of the graph being the constant of proportionality. The constant of proportionality contains the necessary correction terms. However, this particular constant of proportionality is applicable only to Fig. 7. Using this information, the counting rate for each core was compared to the counting rate of the standard to get the U^{235} content.

Because a comparison technique is being used, explicit correction factors can be neglected. In addition, the constant terms in Eq. 8 given in Appendix A contain correction terms. The core blanks had approximately 0.3 gm U^{235} per cm^2. From Fig. 5 it can be seen that the self-absorption for this region is relatively small. Likewise the counting rate is low enough so that this correction is small also.

To determine the accuracy of the method experimentally, six core blanks were counted before and after pickling. Table IV shows the data obtained. As can be seen, the cores lost from 100 to 300 mg U^{235} in the pickling operation. These results agree with the calculation of the mass of U^{235} lost by counting.

The following techniques were also used to determine the sensitivity of the equipment to changes in U^{235} content. A one-mil strip of enriched uranium foil, with a mass of approximately 0.8 gm, was mounted on the standard core blank and counted. The foil was placed at various positions on the top and bottom of the core blank. It was found that variations in the position of the foil caused a variation in the counting rate of

Table IV

CHANGE IN COUNTING RATE DUE TO REMOVAL OF U^{235}

Core Number	Relative Counting Rates		U^{235} Content (gm)		Loss of U^{235} (gm)	
	Before Pickling	After Pickling	Before Pickling	After Pickling	By Counting	By Computation*
24-2	0.9944	0.9914	38.4	38.2	0.2	0.3
24-3	1.0053	1.0018	39.2	39.0	0.2	0.3
24-4	1.0056	1.0019	39.3	39.0	0.3	0.3
24-5	1.0080	1.0034	39.4	39.1	0.3	0.3
24-6	1.0057	1.0025	39.3	39.0	0.3	0.3
24-8	1.0036	1.0023	39.1	39.0	0.1	0.3

* These data were computed on the assumption that the core alloy was homogeneous. It appears from the other data that the uranium concentration may vary throughout the thickness of the core.

only about 0.3%. There was a 1.4% change in the overall counting rate due to the 1.8% change in source strength. By taking the average of a number of counts with the foil in different positions on the top and bottom of the core, the change in counting rate due to a known change in U^{235} content was obtained.

The following procedure was adopted for the determination of the U^{235} content in Argonne Low Power Reactor cores. The 184-kev gamma from U^{235} was counted differentially with a 2% window width, giving a counting rate of approximately 5,000 counts per second. Each core was counted for five minutes. The standard core was counted after each five cores. The five cores counted between counts of the standard were compared to the average of the two counts for the standard. The detailed procedure for calculating the U^{235} of the Argonne Low Power Reactor cores is given in Appendix A.

Experience has indicated that a slight improvement in stability of the photomultipliers can be obtained by putting the phototube and scintillator in a temperature-regulated box. The counting rates given in Table IV were too high and caused fatigue and instability in the phototube current. The counting rate was lowered to approximately 5,000 counts per second for counting Argonne Low Power Reactor cores. The counting rate was lowered without altering the experimental setup by inserting a lead foil absorber between the specimen and scintillator crystal.

IV. SCANNING OF FUEL ELEMENTS

This equipment and technique can also be used to measure the total U^{235} content in a fuel element and to indicate inhomogeneity in the fuel. For measuring the U^{235} content of completed fuel elements, a scanning device is used to move the fuel element in front of the scintillator crystal. Heavy lead shielding is provided so that the detector can view only the section of the fuel element directly under the detector. The speed of traverse must be carefully regulated since the accuracy of the technique depends upon the constancy of the speed of the motor which drives the scanner. The counts are accumulated over the period of a scan on a decade scaler. For indicating inhomogeneity, a count-rate meter rather than a scaler is used. The count-rate meter is generally used as a differential count-rate meter rather than as an absolute instrument in this application. This is done by balancing out part of the constant output so that variations in counting rate are more easily observed. The output of the differential count rate meter is recorded on a strip chart recorder. Determination of the total U^{235} content and the inhomogeneity in fuel can be done at the same time.

Four fuel elements containing cores which had previously been counted were scanned. The four fuel elements were approximately 30 inches long and required approximately 8 minutes for scanning. The cores had been pickled after the initial gamma counting and before rolling. Table V shows the results obtained. Only fair correlation was obtained; however, since the cores had been pickled after gamma counting, this may account for some of the discrepancies. As mentioned previously, it appears that the uranium concentration may vary throughout the thickness of the core.

Table V

SCANNING OF ARGONNE LOW POWER REACTOR FUEL ELEMENTS

Core Number	counts Per second	Accumulated Counts for Five Minutes	Relative Counting*	Plate Number	Accumulated Counts for Scan of Plate	Relative Count **
19-2	9,066	2,719,707	1.00	E-66	1,916,063	1.00
20-18	9,168	2,750,418	1.011	E-69	1,938,682	1.012
19-17	9,088	2,726,319	1.002	E-72	1,927,578	1.006
19-16	9,121	2,736,376	1.006	E-73	1,950,935	1.018

* Count relative to Core No. 19-2.

** Count relative to Plate No. E-66.

V. CONCLUSION

The technique described above has been used to determine the U^{235} content of Argonne Low Power Reactor core blanks. The U^{235} content could be determined to ± 0.1 gram in core blanks which had a U^{235} content of approximately forty (40) grams. Some of the previously mentioned fuel element cores were rolled into fuel plates and scanned. Fair correlation was obtained between these results and the results obtained by counting the core blank.

APPENDIX A

PROCEDURE FOR COUNTING ALPR CORE BLANKS

Each core was counted for five minutes and the standard core was counted after every fifth core. The standard core blank contained 38.81 grams U^{235}. A change in counting rate of 29,800 per five-minute interval (99.3 counts/sec) is equal to 1 gram U^{235}.

To determine the U^{235} content the following measurements and calculations are made.

(a) Count each ALPR core blank for five minutes.
(b) Count the standard core blank after every fifth unknown core blank for five minutes.
(c) Insert the five-minute accumulated count for the unknown core blank (C_u) in equation 8 below.

Equation 8 below gives the number of grams (ΔW) to be added or subtracted from the standard.

$$\Delta W = W_u - W_s \tag{6}$$

where

W_u = mass of U^{235} in unknown

W_s = mass of U^{235} in standard

$$\Delta W = \frac{1}{\text{counts/gm}} \left[C_u - C_s \right] = \frac{C_s}{\text{counts/gm}} \left[\frac{C_u}{C_s} - 1 \right] \tag{7}$$

where

C_u = accumulated counts for unknown

C_s = accumulated counts for standard

$$\Delta W = \frac{1{,}624{,}000}{29{,}800} \left[\frac{C_u}{1{,}624{,}000} - 1 \right] \tag{8}$$