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RECEIVED BY TIG SEP 20 1972ENRICHMENT MEASUREMENT IN LOW
ENRICHED ^{235}U FUEL PELLETS

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ABSTRACT

A multi-channel analyzer and NaI-photomultiplier tube were used to measure the ^{235}U enrichment in low enriched UO_2 pellets. A selection of pellets ranging from 0.85% to 16% in enrichment and of diverse origins was found to yield an accurately linear relation between a properly defined counting rate and the percent ^{235}U enrichment. It was found necessary to perform the background subtraction carefully in order for the resulting linear plot to pass through the origin.

A simple technique for making pellet measurements was developed based on this observation which is sufficiently accurate to separate the usual neighboring values of enrichment. Field applications and experiences with this technique for low enriched fuel pellet measurements will be described.

INTRODUCTION

This application of the enrichment gauge is directed at that segment of the industry and its associated consulting and regulatory groups concerned with the isotopic assay of low enriched uranium oxide pellets. The basic technique is not new and has been previously published in the literature(1).

The enrichment of power reactor fuel pellets of various sizes can be determined by counting the 185 KeV gamma radiation and Compton continuum admitted through a mask. By using the proper mask, the detector is seeing an amount of radiation from the pellet that is dependent on isotopic enrichment and not the weight or surface area of the pellet. A proper subtraction

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of the Compton continuum yields a straight line calibration which passes through zero within the limits of error.

The method and analysis of data are presented to provide a working measurement technique for safeguards and quality control.

DESCRIPTION OF METHOD

The method has been demonstrated on pellets weighing from seven to one hundred grams net over an isotopic range of 0.85 to 16 weight percent ^{235}U (Fig 1). The pellet holder and mask over the detector on which the pellet rests is the most important piece of hardware (2). It is fabricated from lead with a minimum thickness of an eighth inch and fits in a slot near the top of the detector shield (Fig 2). Since the typical low enriched pellet is nominally three eighths inches in diameter by five eighths inches long, the hole in the mask was made a quarter of an inch wide by a half inch long arranged symmetrically about a shallow V-groove depression which places the pellet over the center of the detector. A stop helps in positioning the pellet in the V-groove in relation to the hole. The hole in the mask is small enough so that the detector sees an infinitely thick section of the pellet in relation to the 185 KeV radiation. To reach infinite thickness in this type of fuel material with this radiation energy only about two millimeters of fuel is required.

In discussing the Compton subtraction, a distinction needs to be made between what the ideal procedure would be and the compromise we were forced into because of the peculiarities of our computer-analyzer.

The full energy peak of a gamma ray of about 185 KeV would have an appearance (using a NaI p.m. tube detector) as shown in the top half of Fig. 3 if it occurred in isolation from any other. In the presence of higher energy gamma rays, the full energy peak will be superimposed on the Compton continuum caused by these. In the case of the 185 KeV peak from ^{235}U in the presence of 1001 and 765 KeV gamma radiation from ^{238}U , the appearance of the 185 KeV peak will be as shown in the lower portion of Fig. 3.

Under the circumstances illustrated in Fig 3, a reasonable definition of the net counts in the full energy peak would be to subtract the area "cdeb" from the area "dapbc", leaving shaded area "apbe". The line "be" is the extension of the Compton continuum level to the right of the peak. The continuum has been found to be fairly smooth and linear over a fairly large region. To perform this type of a Compton "background" subtraction should not be outside the capabilities of a computer based pulse height analyzer. In the present instance it was, and so a technique was evolved which, while easier to perform, is harder to explain (Fig 4).

The pulse height distribution obtained from a typical low enriched pellet is plotted in the neighborhood of the 185 KeV peak. The sides of the full energy peak are fairly linear over a large portion of their range. Extend the straight portion of each side to where they intersect the linear backward extrapolation of the Compton continuum. Through each of these points of intersection (e' and b') erect perpendiculars to the abscissa

100 gm

20 gm

11 gm

7 gm

SAMPLING OF LOW ENRICHED PELLETS

FIG. 1

FIG. 2

FIG. 3

FIG. 4

(points g and h). These are considered as defining a "lower" and "upper" channel respectively for use in the data reduction for all the sample measurements, whether "calibration" or "unknown".

The analyzer was calibrated so that channel 185 corresponded to 185 KeV. The resolution of the NaI p.m. tube was then such that g and h in Fig 4 were about 60 channels apart. The counts in 5 channels to the right of h and left of g were summed and the total multiplied by the ratio (60/10). This figure is then subtracted from the total count between the perpendiculars leaving the net shaded area "ii'pj'j".

In effect, the average values of channels to the right and left of the peak have been used to define the bases of a trapezoid "gijh" which is being (somewhat artificially) considered as the Compton contribution to the area under the full energy peak.

This procedure yields a result which differs from that depicted in Fig 3, however, the difference is small and is exaggerated in these sketches. In fact, using this method to reduce data yields a linear relation for count rate versus enrichment passing through the origin within the calculated uncertainties.

In the field, the measurement proceeds as follows: The van carries a "library" of pellets of various low enrichments gleaned through the years from the various licensees. Knowing the range of enrichments to be measured on a given occasion, a selection of about five pellets whose contents span these enrichments is made from the library. The spectrum of the library pellet having the median enrichment is taken and plotted, and this plot is used to establish the upper and lower limits. These upper and lower limits then define the procedure (as just outlined) for getting the net counts in the full energy peak for the library pellets and the unknowns.

The net counts for the library pellets as a function of their percent enrichments are least squares fit to a straight line, including uncertainties, using the procedure as outlined (for example) in "Experimental Statistics" Handbook 91, U. S. Department of Commerce, National Bureau of Standards. The calculation is illustrated in Table I and II of the Appendix. With a little practice this procedure can be run through in perhaps 10 minutes with an electronic desk calculator.

The net count rate derived for the unknown pellets substituted into the derived calibration equation yields the percent enrichment. In our beginning work, solving for the percent enrichment was done graphically. It was found though that the potential accuracy of the method suffered because of the errors involved in plotting the calibration line and interpolating from it.

The practice to date has been to run a new calibration curve at the start of each measurement day. This has been necessary because of significant gain changes that have been observed. It is felt that a pulse height stabilizer used in connection with the analyzer might make it possible to calibrate the analyzer so that the system stays locked in indefinitely. This would cut down on the 30 minutes or more involved at the beginning of each day in setting up.

Small shifts in gain observed through the day (manifested by shift in peak position) are now corrected by small changes in the high voltage; pulse height stabilization would make this unnecessary too.

With the computer-analyzer the Region I van is using, it is very easy to automatically printout at the end of each (300 second) run, the integrated counts in the five channel region to the left, to the right, the total counts in the region between and the net counts under the peak after the Compton subtraction once these regions have been graphically determined as outlined above for one case at some intermediate enrichment. It should be remembered that one is trying to approximate in a practical, convenient, reproducible way the ideal illustrated in Fig 3, and that the ultimate test of a method should be whether a least squares fitted calibration line passes through the origin. Simply obtaining a straight line relation is not enough, because the presence of an intercept, if positive, means that the sensitivity is not as great as it could be; if negative, it means effectively that part of the range of enrichment will not be measurable. Moreover, it is felt that with a non-zero intercept there would be a certain lowering of confidence in measurements.

ANALYSIS OF DATA

The percent enrichment versus count rate for an enrichment range of 0.85 to 16% is plotted in Fig 5. The count rates were determined as described above. The pellets represented were obtained from three vendors and also include a variety of sizes as indicated in Fig 1. The least squares linear fit to the data passes accurately through the origin. The outlying curves represent the limits of error (2σ). This result indicates the possibility of utilizing the method over a wide range of enrichments.

In practice, only a small number of pellets chosen (from the "library" carried by the van) to have approximately the enrichment likely to be measured are used in the calibration. About five pellets are required for a reasonable limit of error estimation. A typical field calibration is as shown in Fig 6, along with the derived linear fit to the data. The equation is then used to determine the enrichment of individual measurements.

In Table I is shown a summary of measurements made over a period of several months. Some seven different field calibrations made at three different fuel fabrication plants are involved. The measurements are presented in order of nominal enrichment. The figure in the "gamma measured" column represents the average of six pellets. The same six pellets were then individually measured at the USAEC New Brunswick Laboratory using a mass spectrometer; an average of these is placed in the neighboring column for comparison. The data is presented in this way as a convenient method of summarizing field results; the complete listing is appended to this article. The third and twelfth grouping of six have average nominal values which appear to differ significantly from the average gamma measurement value and the average of NBL measurements on the same pellets. This is because these two groups included, respectively, one and two pellets that were inadvertently placed in the wrong nominal category. The important comparison is between the gamma derived measurements and the New Brunswick Laboratory

Figure 5

Table I

SUMMARY OF U-235 ENRICHMENT MEASUREMENTS

<u>NOMINAL</u> <u>(FABRICATOR VALUES)</u>	<u>GAMMA MEASURED</u>	<u>NBL</u> <u>(MASS SPEC.)</u>
2.05	2.10	2.06
2.27	2.24	2.26
2.40*	2.55	2.50
2.45	2.51	2.45
2.80	2.77	2.78
2.90	3.06	2.90
2.95	3.08	2.96
2.99	3.08	2.98
3.14	3.27	3.15
3.20	3.21	3.21
3.40	3.26	3.42
3.60**	3.29	3.31
4.00	3.95	4.02

* GROUP CONTAINS ONE PELLET NOT OF NOMINAL ENRICHMENT.

** GROUP CONTAINS TWO PELLETS NOT OF NOMINAL ENRICHMENT.

Figure 6

Typical Field Calibration

$$\text{Enrichment} = -0.02 + 0.0001688 \text{ Net Counts}$$

mass spectroscopic values. The average absolute deviation is 0.07%. A statistical analysis of the total of 82 field measurements (for which Table I represents an abstract) (see Table III, Appendix) shows the deviation, mass spectroscopic minus gamma, to be -0.01 ± 0.02 absolute weight percent ^{235}U (2 σ). The bias of -0.01 weight percent cannot be demonstrated to be significant. The limit of error (2 σ) on a single pellet measurement is 0.17 weight percent of ^{235}U . Thus, there is a 95% probability that a pellet differing by as much as 0.17 weight percent in enrichment from a population can be detected to be different.

In assessing the above results it should be recognized that they were obtained under condition that are inferior to the usual laboratory environment as regards to temperature and line voltage fluctuations. It is expected that with a fixed laboratory set up dealing with a product which is more standard with regard to dimensions, compositions, etc., significantly better results could be obtained.

The success of this method for individual pellets led to interest in what results might be obtained by making spot checks of enrichment for fuel rods made up of such pellets. Fig. 7 illustrates a calibration curve obtained by making dummy sections of fuel rods out of pellets of known composition put into tubing of the type used for fuel rods. In this case only the crudest collimation was employed. Similar results were obtained with actual fuel rods, however, no amount of data comparable to that on single pellets has been accumulated as yet.

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REFERENCES

1. Russell, J. T., Method and Apparatus for Nondestructive Determination of ^{235}U in Uranium, U. S. Patent 3,389,254, June 18, 1968 (To United States Atomic Energy Commission).
2. Cohen, I. and Gundersen, G., Field Experience with the AEC Measurement Van, Proceedings of the Twelfth Annual Meeting of the Institute of Nuclear Materials Management, West Palm Beach Shores, Florida, CONF-710617-3 (1971).

Figure 7

Fuel Rod Calibration

$$\text{Enrichment} = -0.45 + 0.0001357 \text{ Net Counts}$$

Appendix

Table I (Appendix)

Least Squares Procedure for Fitting
Enrichment vs. Count Rate

X denotes counts per time interval Y denotes percent enrichment

$\Sigma X =$ _____ $\Sigma Y =$ _____
 $\bar{X} =$ _____ $\bar{Y} =$ _____

Number of points: $n =$ _____

Step (1) $\Sigma XY =$ _____

(2) $(\Sigma X)(\Sigma Y)/n =$ _____

(3) $S_{xy} =$ Step (1) - Step (2)

(4) $\Sigma X^2 =$ _____

(7) $\Sigma Y^2 =$ _____

(5) $(\Sigma X)^2/n =$ _____

(8) $(\Sigma Y)^2/n =$ _____

(6) $S_{xx} =$ Step (4) - Step (5)

(9) $S_{yy} =$ Step (7) - Step (8)

(10) $b_1 = \frac{S_{xy}}{S_{xx}} =$ Step (3) $\div$ Step (6)

(14) $\frac{(S_{xy})^2}{S_{xx}} =$ _____

(11) $\bar{Y} =$ _____

(15) $(n - 2) s_y^2 =$ Step (9) - Step (14)

(12) $b_1 \bar{X} =$ _____

(16) $s_y^2 =$ Step (15) $\div$ (n - 2)

(13) $b_0 = \bar{Y} - b_1 \bar{X} =$ Step (11) - Step (12)

$s_r =$ _____

Equation of the line:

$$Y = b_0 + b_1 X$$

$s_{b_1} =$ _____

$s_{b_0} =$ _____

Estimated variance of the slope:

$$s_{b_1}^2 = \frac{s_y^2}{S_{xx}} = \text{Step (16)} \div \text{Step (6)}$$

Estimated variance of intercept:

$$s_{b_0}^2 = s_y^2 \left\{ \frac{1}{n} + \frac{\bar{X}^2}{S_{xx}} \right\} =$$

Note: The following are algebraically identical:

$$S_{xx} = \Sigma (X - \bar{X})^2; S_{yy} = \Sigma (Y - \bar{Y})^2; S_{xy} = \Sigma (X - \bar{X})(Y - \bar{Y}).$$

Table II (Appendix)

Give a $(1 - \alpha)$ Confidence Interval Estimate For a Single (Future) Value of Y Corresponding To a Chosen Value of $X = X'$.

Procedure	Example
(1) Choose the desired confidence level, $1 - \alpha$	(1) Let: $1 - \alpha = .95$ $\alpha = .05$
(2) Obtain s_r from Worksheet.	(2) From Worksheet 5-5.2 $s_r = 26.418$
(3) Look up $t_{1-\alpha/2}$ for $n - 2$ degrees of freedom in Table A-4.	(3) $t_{.975}$ for 9 d.f. = 2.262
(4) Choose X' , the value of X at which we want to make an interval estimate of a single value of Y .	(4) Let $X' = 300$, for example.
(5) Compute:	(5)
$W_s = t_{1-\alpha/2} s_r \left[1 + \frac{1}{n} + \frac{(X' - \bar{X})^2}{S_{xx}} \right]^{.5}$	$W_s = (2.262) (26.418) (1.0463)$ $= 62.5$
and	
$Y_c = \bar{P} + b_1 (X' - \bar{X})$	$Y_c = 247.5$
(6) A $(1 - \alpha)$ confidence interval estimate for Y' (the single value of Y corresponding to X') is given by	(6) A 95% confidence interval estimate for Y at $X = 300$ is the interval 247.5 ± 62.5 , i.e., the interval from 185.0 to 310.0
$\bar{P} + b_1 (X' - \bar{X}) \pm W_s$ $= Y_c \pm W_s$	

TABLE III (APPENDIX)

LOW ENRICHED FUEL PELLET DATA

NOMINAL (FABRICATOR VALUE)	GAMMA MEASURED	NBL (MASS SPEC.)	NOMINAL (FABRICATOR VALUE)	GAMMA MEASURED	NBL (MASS SPEC.)
2.05	2.09	2.06	2.99	2.98	2.98
	2.10	2.06		3.15	2.98
	2.06	2.06		3.12	2.98
	2.13	2.06		3.04	2.98
	2.07	2.06		3.11	2.98
	2.14	2.06		3.01	2.98
2.27	2.34	2.26	3.14	3.07	3.15
	2.30	2.26		3.07	3.15
	2.24	2.26		3.20	3.15
	2.26	2.26		3.22	3.15
	2.19	2.27		3.15	3.15
	2.33	2.27		3.01	3.15
2.40	3.05	2.92*	3.20	3.27	3.24
	2.43	2.42		3.18	3.19
	2.45	2.41		3.19	3.19
	2.47	2.42		3.25	3.21
	2.48	2.41		3.15	3.19
	2.52	2.43		3.24	3.22
2.45	2.42	2.45	3.40	3.25	3.42
	2.49	2.46		3.28	3.43
	2.59	2.45		3.40	3.42
	2.50	2.45		3.37	3.42
	2.50	2.45		3.37	3.42
	2.56	2.45		3.38	3.42
2.80	2.89	2.78	3.60	3.47	3.61
	2.80	2.77		3.45	3.61
	2.83	2.77		3.00	3.13*
	2.81	2.78		3.55	3.61
	2.79	2.77		3.64	3.61
	2.71	2.78		2.15	2.26*
2.90	2.90	2.90	4.00	3.91	4.01
	2.84	2.90		3.80	4.02
	2.86	2.90		3.96	4.01
	2.87	2.91		4.01	4.01
	2.91	2.90		3.97	4.02
	2.92	2.89		4.06	4.02
2.95	3.02	2.97		3.94	4.02
	3.11	2.95		4.06	4.02
	3.04	2.95		4.01	4.02
	2.99	2.95		3.92	4.02
	3.17	2.95			
	3.13	2.95			

* PELLET NOT OF NOMINAL ENRICHMENT