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THE NONDESTRUCTIVE ASSAY OF EBR-II FUEL PINS

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by

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Special Materials and Services Division

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ABSTRACT

The expanding safeguards program and the increasing use of reactors as a source of power point up the desirability of having a program to assay large quantities of reactor fuel.

This report discusses the use of gamma-ray assay as an effective means of verifying ^{235}U content for accountability purposes. There were 3423 fuel pins assayed; the difference between the fuel content as determined by gamma assay and that reported by the vendor was less than 1% at the 95% confidence level.

INTRODUCTION

Fuel pins for the Experimental Breeder Reactor II (EBR-II) were nondestructively assayed by the Quality Evaluation Section of the Special Materials Division at Argonne National Laboratory. The purpose of the assay was to verify for accountability purposes that the fuel met the specifications of the transfer document within prescribed tolerances.

The EBR-II driver fuel pins assayed in this project are about 95% uranium, enriched to approximately 52% ^{235}U. They are about 18 in. long and 1/8 in. in diameter (as shown in Fig. 1), and contain 31.5-32 grams of ^{235}U. Of 36,000 pins produced, approximately 10% (3423) were assayed.

BASIC PRINCIPLES

The ^{235}U content was assayed by counting the number of 185-keV gamma rays emitted by the fuel pins. The basic equation relating to radioactive decay is

$$N = N_0 e^{-\lambda t},$$

where

N = number of atoms at time t,

N_0 = numbers of atoms at time $t = 0$,

and

λ = decay constant = $(\ln 2)/T_{1/2}$, where $T_{1/2}$ is the half-life of the isotope. The disintegration rate is then

$$\frac{dN}{dt} = -N_0\lambda e^{-\lambda t} = -N\lambda.$$

For an isotope of long half-life, such as ^{235}U whose half-life is 7×10^8 years, the disintegration rate can be considered constant.

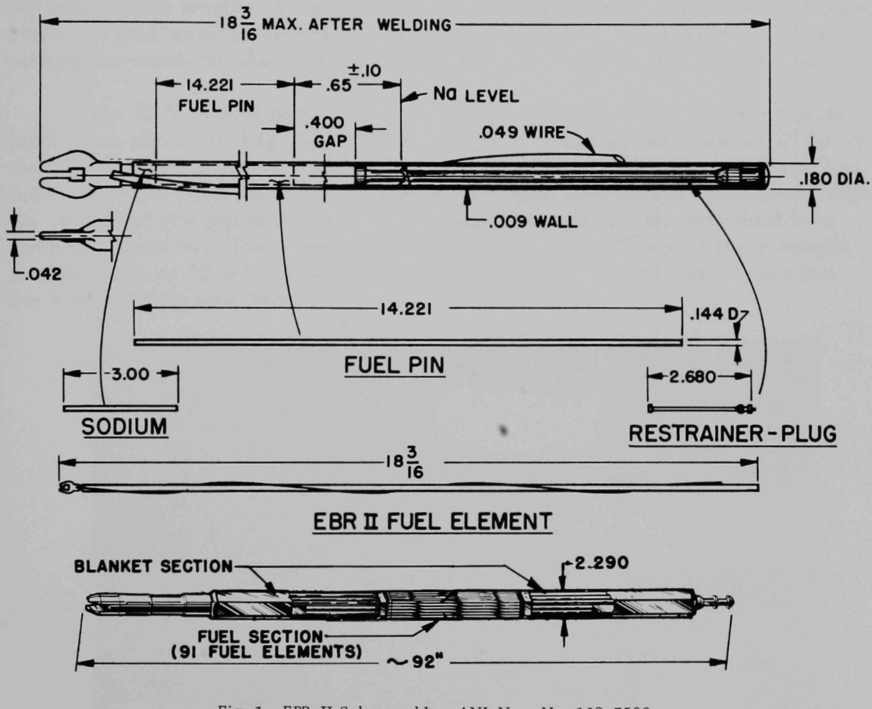

Fig. 1. EBR-II Subassembly. ANL Neg. No. 108-7599.

Uranium-235 decays by alpha emission to ^{231}Th. Associated with the alpha decay are a number of gamma-ray energies. The 185-keV gamma ray is emitted in a constant fraction of the total disintegrations. However, only a fraction of the 185-keV gamma rays emitted are actually detected and counted. If this fraction is held constant, the number of 185-keV gamma rays detected is proportional to the total number of ^{235}U atoms present. The task is to detect as large a fraction as possible of the 185-keV gamma rays

while controlling factors such as background radiation and geometry which might change this fraction. In this way, the number of 185-keV gamma rays detected from a production pin can be related to the number detected from a set of standard pins with known ^{235}U content. Through this relationship the ^{235}U content of the production pins can be calculated.

EQUIPMENT

The detection equipment includes a lithium-drifted germanium (GeLi) detector having a volume of about 20 cc and a resolution of about 8 keV at 1.33 MeV. The data collected were fed to a 400-channel analyzer used in the multiscale mode. A single-channel analyzer built into the 400-channel analyzer was used to select the 185-keV peak from the spectrum. The counts for each pin were stored in a separate channel of the analyzer.

An X-Y positioning table was used to move the pins under the detector (as shown in Fig. 2). The table had a scanning distance of 27 in. in the X direction and 8 in. in the Y direction with a positioning accuracy of 0.001 in. The pins were supported on the table with nylon spacers so that the length of the pin was along the X direction and the tip was held in a vertical position. The pins could then be rotated 180° along their length. A maximum of 10 pins could be equally spaced, 0.811 in. apart, across the table at any one time.

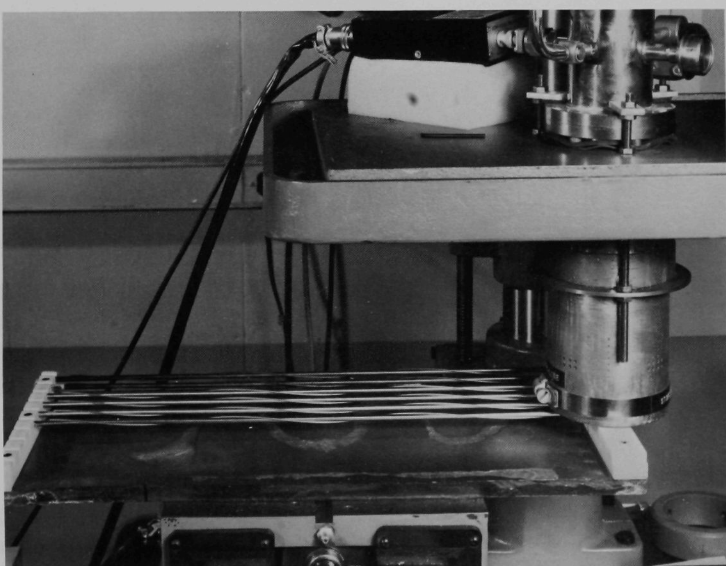

Fig. 2. Scanning Table with EBR-II Fuel Pins. ANL Neg. No. 150-513.

The table was controlled by a preset indexer, which was supplied with the table, and a preset-indexer control of our own design. The preset indexer allowed us to select the distance of travel in each direction. The distance traveled was 16.000 in. in the X direction and 0.811 in. in the Y direction. The 16 in. in the X direction was enough so that the detector started beyond one end of the fuel (approximately 13 in. long) and finished past the other end. The 0.811 in. in the Y direction was equal to the spacing between individual pins, so that moving the table once in the Y direction would move a new pin into position for counting.

The preset-indexer control allowed selection of either ± direction for both X and Y motion and the number of pins to be scanned. The control permitted the indexer to move the table so that each pin was scanned under the detector. When all the pins had been scanned, the control stopped the indexer and waited to be reset. The pins were then turned over and scanned in the opposite direction. (The equipment described above is shown in Fig. 3.)

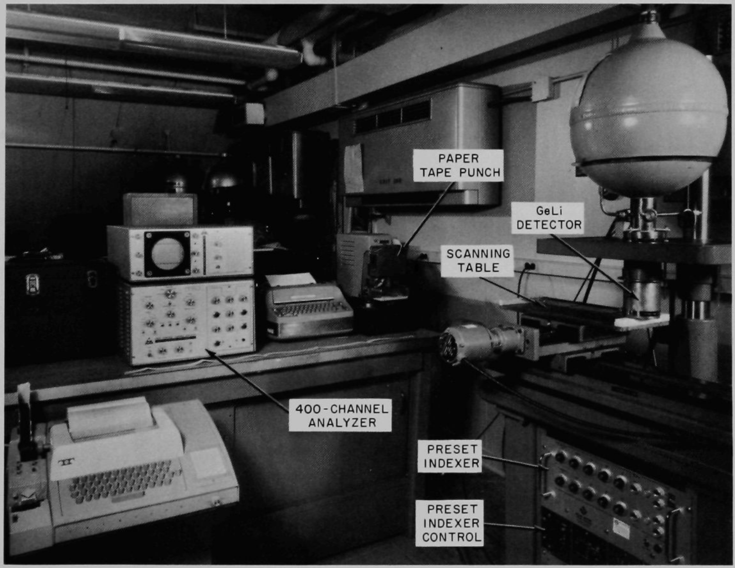

Fig. 3. Gamma-ray Assay Equipment. ANL Neg. No. 150-515.

Pulse pileup in the electronics can cause erroneous count data. To account for this, a correction system was devised. A pulse of variable amplitude and frequencies of 62.5, 125, 250, 500, or 1000 Hz was fed into the preamplifier through the test input. The amplitude of the pulser pulse was adjusted to fall into the window of the single-channel analyzer that had

previously been set for the 185-keV peak. The output of the single-channel analyzer was fed back to the correction system, where the pulses in coincidence with a signal from the pulser (i.e., the pulser pulses) were separated from the true counts, which were fed back to the multichannel analyzer to be counted. The pulser pulses were amplified and fed to the preset indexer, which fed them to the stepping motors. Since the pulser pulses were treated like any other pulses from the time they entered the preamplifier until they were just ready to be counted, a high count rate would cause some of them to be lost as well as some of the 185-keV pulses. If pulser pulses were lost, the table received fewer pulses and ran slower. Therefore, when counting losses were high, the pin moved slower under the detector.

The pulser rate was selected so that the pulser pulses would not greatly increase the counting losses but would still sample the counting system frequently enough to make a good correction. The speed of the table could then be adjusted by means of a divider, which would divide the frequency of the pulses fed to the indexer by 2, 4, 8, 16, or 32. The possible range was 500-1.95 pulses/sec, or 500-1.95 mils/sec. The practical maximum was 250 mils/sec, the maximum speed at which the table operated reliably.

PROCEDURE

Background count was determined by counting an empty location on the table loaded with standards. Since the background was different for each position on the table and varied with the number of pins on the table, the one background count was related to the count at any position by a series of actual background counts taken before the start of work on the production pins. The time needed for background counts made it impossible to count background for each position during production. The data as stored in the 400-channel analyzer were typed out on the attached typewriter and punched on paper tape. The typewriter readout was used to verify the paper tapes.

The standards included six pins ranging from a depleted pin to a 70%-enriched pin. A 52%-enriched pin, similar to the production pins, was included.

The standards and background were counted twice each day, once in the morning with one side up, and once in the afternoon with the other side up. The production pins were also counted on both sides. The pin numbers and vendor weight for each day were punched on paper tape using a teletype. This tape along with the data tapes was read into a small computer. The program took the standard count data along with the known ^{235}U content of each standard and calculated a straight line using a least-squares technique. The data for the production pins were fitted to the standard line, and the ^{235}U content of each pin was calculated along with a confidence and difference

between the assay value and the vendor value of ^{235}U. The computer then typed a table containing the information in a form that was suitable for a report. Some typical results are shown in Table I.

TABLE I. Typical Results of Fuel Assay of EBR-II Fuel Pins

	Weight of ^{235}U, g, as Determined by		
Pin Number	Gamma Assay	Vendor Chemistry	Difference
	Batch Number 682		
41754	31.92	32.12	-0.20
43357	32.17	32.06	0.11
45847	32.15	32.13	0.02
46313	31.89	31.85	0.04
47425	31.92	32.15	-0.23
47935	32.02	31.88	0.14
48162	31.94	31.92	0.02
48296	31.84	31.86	-0.02
48401	31.99	31.89	0.10
48408	31.83	32.00	-0.17
Total	319.66	319.86	-0.20
	Batch Number 683		
38411	32.12	32.04	0.08
39318	32.02	31.78	0.24
39748	31.90	31.87	0.03
40379	31.86	31.94	-0.08
40397	31.91	31.81	0.10
40800	31.73	31.86	-0.13
41002	32.11	31.88	0.23
41467	31.97	32.03	-0.06
41692	31.96	31.85	0.11
42343	32.11	31.90	0.21
Total	319.70	318.96	0.74
	Batch Number 684		
32152	31.88	31.94	-0.06
32338	31.94	31.95	-0.01
32509	31.88	31.98	-0.10
32904	31.87	31.83	0.04
35327	31.80	31.90	-0.10
35379	31.79	31.74	0.05
35801	31.88	31.77	0.11
36727	31.73	31.95	-0.22
36734	31.86	31.94	-0.08
37028	31.87	31.87	0.00
37101	32.04	31.96	0.08
Total	350.53	350.83	-0.30

RESULTS

A total of 3423 pins from 362 batches were counted. The total weight of these pins, as determined by gamma assay, was 108,815.45 g; the vendor's reported weight was 108,853.23 g. The difference between gamma assay and vendor's weight was -37.78 ± 52.88 g at the 95% confidence level. The average weight for each pin was 31.789 and 31.800 g, respectively, giving a mean difference of 0.011 g or 0.035%. Chemical analysis was done by Argonne on 64 of the batches. Comparison of the results for these batches follows:

Method of Determination	Mean Weight of ^{235}U, g
Gamma Assay	31.80 ± 0.3
Vendor chemistry	31.80 ± 0.2
ANL chemistry	31.73 ± 0.4