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Measurement of the Relative ^{235}U Concentration and the Absolute ^{235}U Content of Individual HTGR Fuel Particles

R. W. Knoll

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UC-77 — Gas-Cooled Reactor Technology

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Reactor Division

THORIUM UTILIZATION PROGRAM (189a OH045)
Fuel Refabrication Development — Task 210

MEASUREMENT OF THE RELATIVE ^{235}U CONCENTRATION AND THE ABSOLUTE ^{235}U CONTENT OF INDIVIDUAL HTGR FUEL PARTICLES

R. W. Knoll

JULY 1976

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CONTENTS

	Page
ABSTRACT	1
INTRODUCTION	1
DESCRIPTION OF TRISO PARTICLES	2
THE DELAYED-NEUTRON METHOD	3
EXPERIMENTAL EQUIPMENT	4
SELECTION OF PARTICLES FOR THE EXPERIMENT	7
DESCRIPTION OF THE UO_2 STANDARDS	7
CONTAINMENT OF PARTICLES AND STANDARDS DURING IRRADIATION	7
IRRADIATION AND COUNTING	8
CORRECTIONS APPLIED TO THE RAW DATA	8
RELATIVE ^{235}U CONCENTRATION IN THE KERNELS	10
UNCERTAINTY IN THE RELATIVE MEASUREMENTS	12
ABSOLUTE ^{235}U CONTENT OF KERNELS	12
CONCLUSIONS	15
ACKNOWLEDGMENTS	15
REFERENCES	16

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ABSTRACT

The absolute ^{235}U content and the relative ^{235}U concentration of 77 individual TRISO-coated HTGR fuel particles were measured using delayed-neutron activation analysis. The particles were from batches A611 and A601. The measurements, which were done to determine the variation in kernel ^{235}U concentration with kernel diameter, revealed no significant size-dependent variation in the kernel uranium concentration. The absolute measurement technique and its applicability to the construction of assay machine calibration standards are discussed. With the present irradiation facility, the technique was found to be too time consuming to be used to measure the large number of particles needed for a calibration standard individually.

Key words: HTGR fuel particle, ^{235}U measurement, nondestructive assay, delayed neutrons, activation analysis.

INTRODUCTION

In a recently reported experiment,[1] the relative ^{235}U concentration of each of ten TRISO-coated HTGR fuel particles was measured using delayed-neutron activation analysis. Five particles with kernel diameters of ∿341 μm and five with kernel diameters of ∿387 μm were assayed to determine if the ^{235}U concentration in the kernel varied with kernel size. Within the limits of experimental error, the larger kernels were found to contain the same uranium concentration as the smaller ones. The present experiment expanded upon this earlier work by measuring the relative kernel ^{235}U concentration of each of 77 TRISO-coated particles with kernel diameters from 325 to 392 μm. As in the earlier work, there was no significant size-dependent variation in the ^{235}U concentration in the fuel kernels.

In addition to these relative uranium measurements, the absolute ^{235}U content of each particle was measured by comparing the delayed-neutron

counts of the particle with those of six well-characterized UO_2 standards. The absolute measurements were made to test the accuracy and convenience of the technique, with the goal of applying the technique to the construction of assay-machine calibration standards. Such calibration standards are composed of many fissile particles, and the total fissile content of the standard must be known to within a fraction of a percent. By sampling the particles that are to compose the calibration standard and then accurately measuring the ^{235}U content of the sample, the ^{235}U content of the standard could be closely estimated. The experiment showed that the technique is reasonably accurate, but it is too time consuming to be used to measure individually the large number of particles needed for an assay-machine calibration standard.

DESCRIPTION OF TRISO PARTICLES

The enriched-uranium fuel particles used in the HTGR consist of spherical kernels of a UO_2-UC_2-C mixture coated with four layers of materials designed to retain the fission products within the particle. The kernels typically range from 325 to 390 μm in diameter and contain from 40 to 70 μg of ^{235}U.

The photomicrograph of a sectioned TRISO particle in Fig. 1 shows the structure of the particle. The innermost layer consists of low-density carbon and is called the buffer coating. This buffer is coated with a layer of high-density carbon, a layer of SiC, and finally, another layer of high-density isotropic carbon. Because the particles are coated with three different materials, they are called TRISO-coated particles.

The particles are manufactured in a batch process and hence are identified by the batch number from which they came. In this experiment, 39 of the particles were from batch A611, and 38 were from batch A601. The main difference between these batches is in the ratio of UC_2 to UO_2 in the kernel. The A611 kernels contain 15% UC_2, 85% UO_2, and carbon; the A601 kernels contain 75% UC_2, 25% UO_2, and carbon.

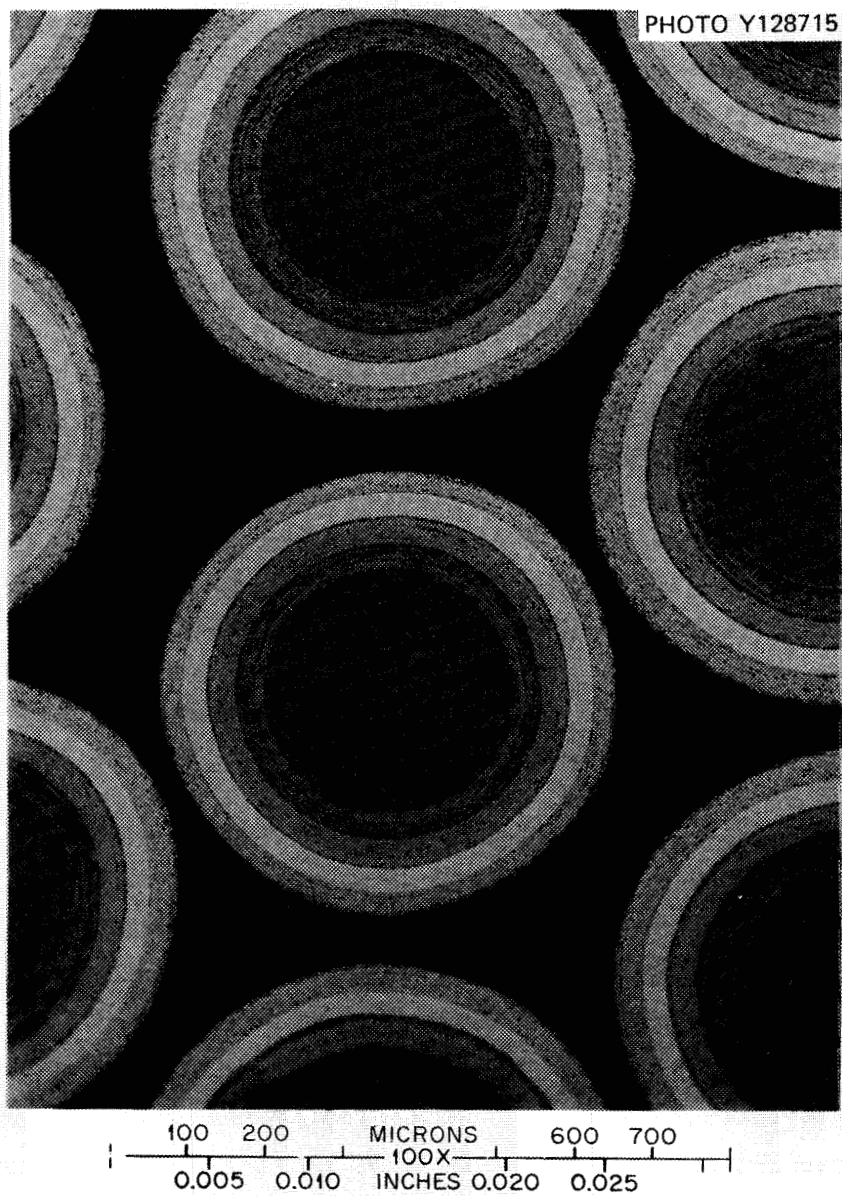

Fig. 1. Photomicrograph of sectioned TRISO-coated fuel particles.

THE DELAYED-NEUTRON METHOD

The delayed-neutron method of determining the ^{235}U content of a fuel particle takes advantage of neutrons that are emitted by some ^{235}U fission products (delayed-neutron precursors) shortly after fission occurs. These neutrons are emitted almost simultaneously with β^- decay of the precursors

and have half-lives[2] of from 0.2 to about 55 sec. By irradiating a fuel particle in a thermal-neutron flux and then quickly transferring the particle to a counting chamber, one obtains a number of counts N_1 that directly corresponds to the number of delayed neutrons emitted during the counting period. The absolute ^{235}U content of the particle is found by comparing N_1 with the delayed-neutron counts from a standard of known ^{235}U content, which has been irradiated and counted under the same conditions as the particle. Alternatively, the ^{235}U loading of a particular particle, relative to the loadings of a number of other similar particles, is measured by identically irradiating and counting all the particles and then comparing the counts of the particle to those of the other particles.

EXPERIMENTAL EQUIPMENT

The particles were activated in the pneumatic tube facility[3] in the Oak Ridge Research Reactor (ORR) (Fig. 2). The tube leads from a laboratory beneath the reactor up into beam hole HN-3 of the reactor and ends at the core face. Small polyethylene rabbits are used to carry samples throughout the tube system. In the laboratory, the pneumatic tube splits into two sections, one of which leads to a loading receptacle and the other to a neutron moderator and detector assembly that consists of a large paraffin cylinder into which the pneumatic tube runs. Six BF_3 neutron detector tubes, each 5.08 cm OD, are embedded in the paraffin symmetrically about the central tube. A 7.62-cm-thick lead annulus between the central tube and the BF_3 tubes absorb gamma rays emitted by the sample. To count the delayed neutrons emitted by the sample, the detector tubes are connected to a preamplifier, amplifier, high-speed scaler, and high-voltage power supply. This setup is shown in Fig. 3.

The system works in the following manner. A rabbit containing the sample to be assayed is loaded into the tube at station 1 (see Fig. 2) and shot up to the face of the reactor core. The sample is irradiated here for a preset time t_i, after which it is shot back down to the moderator and detector assembly. The sample is allowed to decay for a short time t_d, and then its delayed neutrons are counted over a time interval t_c. All functions, including the timing, counting, and movement of the rabbit

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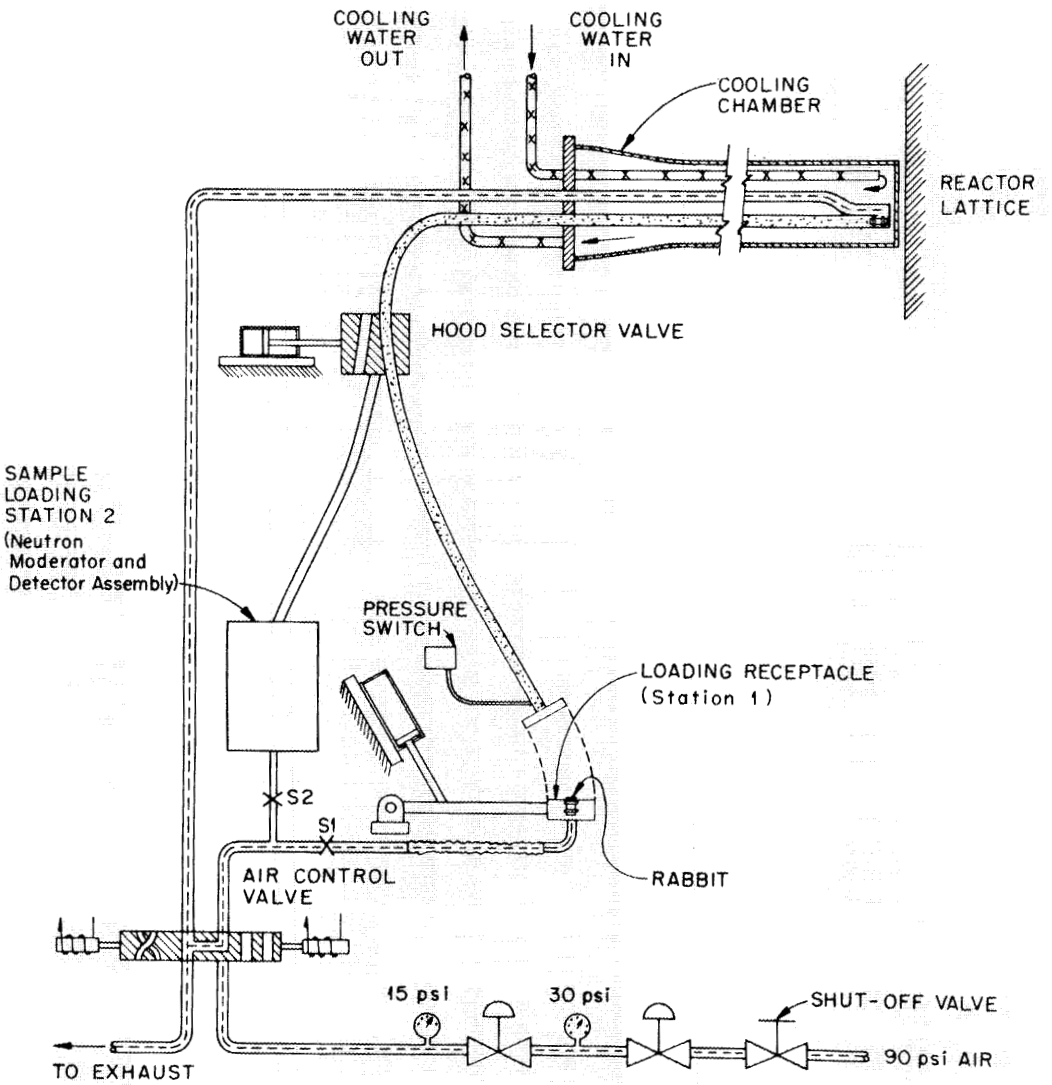

Fig. 2. Pneumatic tube system — ORR.

through the tubes, are controlled automatically. Since certain errors inherent in the irradiation and counting process are related to t_i, t_d, and t_c, these time intervals are chosen to minimize the errors. Reference 3 describes how this is done.

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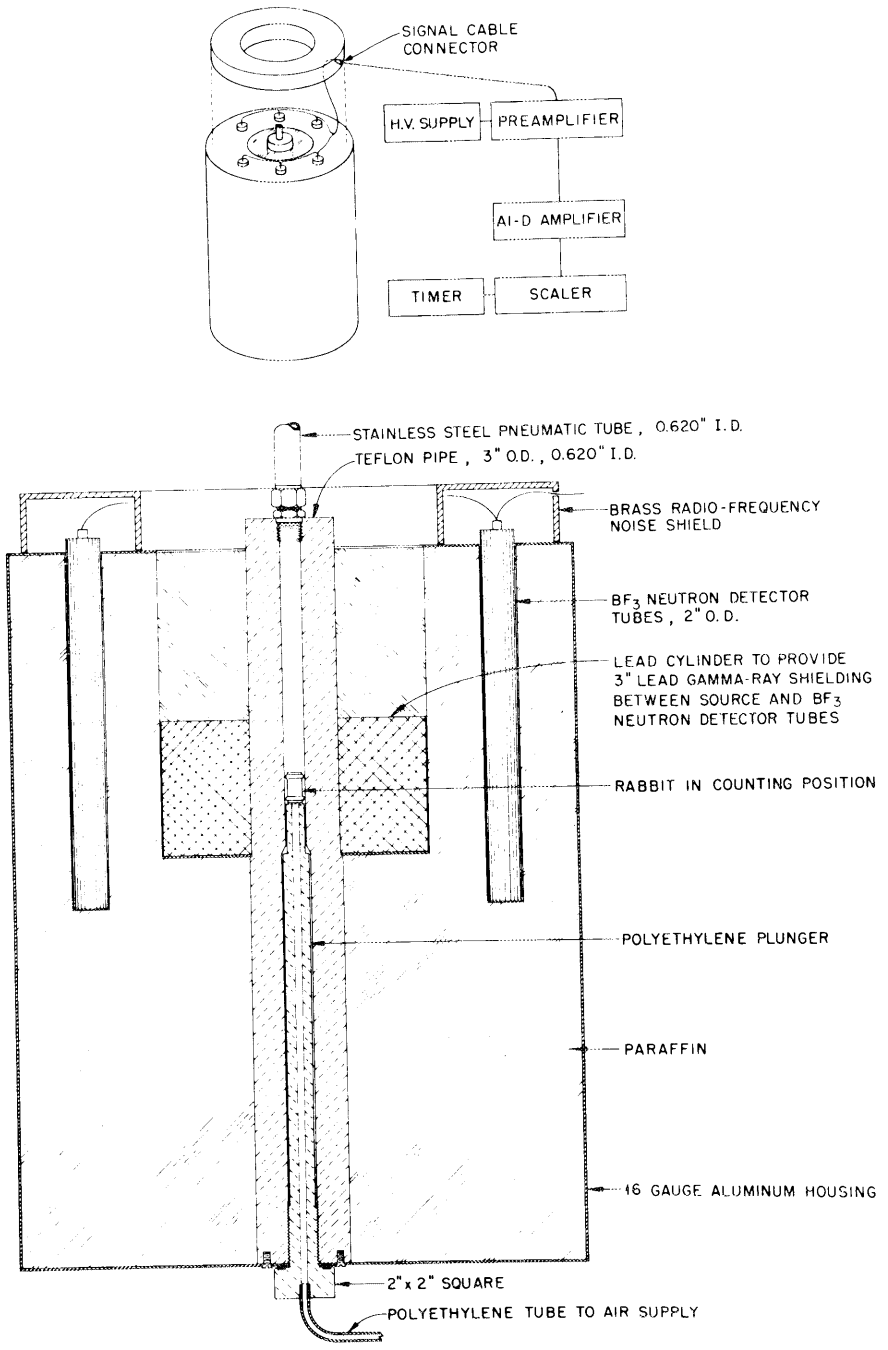

Fig. 3. Delayed-neutron moderator and detector assembly and electronic components.

SELECTION OF PARTICLES FOR THE EXPERIMENT

The fuel particles were randomly chosen from their respective batches by a riffling process, after which they were radiographed and placed in separate numbered vials. The diameter of each fuel kernel was measured from the radiographs using a microscope equipped with a cross-hairs measuring device. To guard against error, two separate sets of diameter measurements were made on each particle.

DESCRIPTION OF THE UO_2 STANDARDS

Each standard consisted of a small fragment of UO_2 that was centered axially in a 0.130-cm-diam by 0.381-cm-long aluminum capsule sealed with a welded aluminum plug. Each UO_2 fragment had been broken off from a UO_2 wire and carefully weighed to an accuracy of about ±1 μg. The weight of the particles ranged from 77 to 105 μg, and the uranium was enriched in ^{235}U to 99.85%. Thus, the ^{235}U content of the standards ranged from ∿68 to 92 μg.

CONTAINMENT OF PARTICLES AND STANDARDS DURING IRRADIATION

High-density polyethylene rabbits with inside dimensions of 0.9 cm diam × 1.95 cm long were used to transport the particles and standards through the pneumatic tube system. Each rabbit contained a snug-fitting graphite cylinder with an indentation machined into the center of its top surface. A fuel particle or a UO_2 standard was placed within the indentation, centered 1.24 cm from the inside bottom of the rabbit. Because the thermal-neutron flux decreased rapidly with axial distance from the rabbit bottom, the particle or standard was prevented from moving about by a polyethylene disk that trapped the particle or standard within the indentation (see Fig. 4).

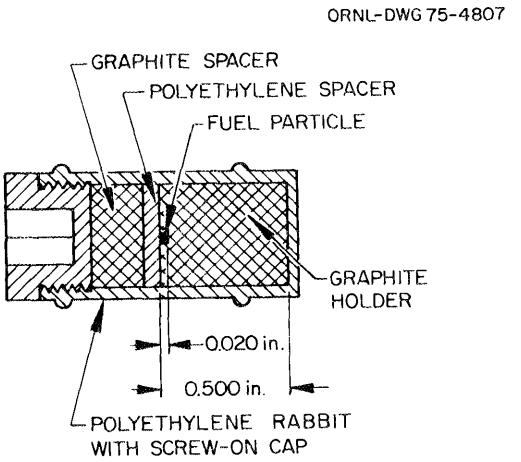

Fig. 4. Rabbit containing graphite holder and particle (1 in. = 2.54 cm).

IRRADIATION AND COUNTING

Because of the relatively large number of particles, the irradiation and counting spanned a three-day period during which the ORR operated at full power. The particles were run in groups of from 10 to 30. Each particle in a group was irradiated for 20 sec, allowed to decay for 60 sec, and then counted for 80 sec. After all particles in the group had been run once, the UO_2 standards were irradiated and counted in the same manner. Each particle in the group was then irradiated and counted a second time in the original order. Then the standards were run again, and the experiment proceeded to the next group of particles, which was run in the same manner. Background checks were made at the beginning and end of each day by irradiating and counting a rabbit containing only a graphite insert. The background counts were always negligible compared to the counts from the fissile material.

CORRECTIONS APPLIED TO THE RAW DATA

Corrections were applied to each delayed-neutron count to account for the temporal variation in the thermal-neutron flux and to compensate for loss of counts due to detector deadtime and thermal-neutron self-absorption

in the particles and standards. Each delayed-neutron count was corrected for the change in the flux level relative to the flux level at the beginning of the experiment. The temporal change in the flux was determined from the run-to-run change in the delayed-neutron counts of the UO_2 standards. The UO_2 standard counts tended to decrease irregularly over the course of the experiment. Relative to their initial values, the monitor counts had decreased about 4.5% by the end of the experiment. Thus, the delayed-neutron counts of the particles that had been assayed near the end of the experiment were corrected by increasing the counts by 4.5%.

Count loss due to detector deadtime was accounted for by using the standard deadtime correction, modified to take into account the exponential decrease in count rate over the counting period.* The correction ranged from 3.6% for the smallest particles to about 7% for the largest monitors.

In order to compare particle-to-particle and particle-to-monitor delayed-neutron counts, it was also necessary to correct for thermal-neutron self-absorption. The larger a particle and the more concentrated its ^{235}U, the more its inner volume is shielded from the thermal-neutron flux. Since a large kernel will absorb fewer neutrons per unit volume than a small kernel, it will appear to contain less ^{235}U than it actually does. To correct for this effect in the kernels, self-shielding factors derived from the Wälti technique were used.[4,5] These corrections depended on kernel diameter and density and were in the range of 4%. An analytical formula was used to correct for self-shielding in the UO_2 monitors.[6] The UO_2 fragments were assumed to be spherical, although in reality they were irregularly shaped. Because of the high ^{235}U concentration in the UO_2, these self-shielding factors were in the range of 12 to 14%.

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*The standard deadtime correction is $N = M/(1 - \dot{M}T)$, where M = observed counts, $\dot{M}$ = observed counts/sec, T = detector deadtime, and N = actual counts. This formula is not valid when the count rate is changing rapidly over the counting period, as in this experiment. Because of the 60-sec decay period between the irradiation and counting of the fuel particle, it was assumed that all delayed-neutron activity came from the 56- and 23-sec delayed-neutron groups. Taking into account the exponential decay of these two groups over the counting period, the deadtime correction was found to be $N = M/(1 - 1.4\ \dot{M}T)$.

RELATIVE ^{235}U CONCENTRATION IN THE KERNELS

After the above corrections were applied, the delayed-neutron count of each particle became directly proportional to its ^{235}U content. The relative ^{235}U concentration of each kernel was found by dividing its corrected delayed-neutron count by its volume and normalizing with respect to the mean concentration. The kernels were assumed to be spheres; so their volumes were calculated as $V_i = (\Pi/6)\ D_i^3$, where D_i is the kernel diameter measured from radiographs of the kernels as explained earlier.

The normalized relative ^{235}U concentration in the A611 and A601 kernels is plotted vs kernel diameter in Figs. 5 and 6, respectively, and the results are summarized in Table 1. The lines drawn through the points in Figs. 5 and 6 are least-squares fits, and as can be seen, the slope of the A611 line is slightly negative. Based on the values given by the line, the difference in ^{235}U concentration between a 390- and a 330-μm-diam A611 kernel is about 1%. However, this 1% is within the limits of error of the

Table 1. Particle parameters and summary of results

	Sample A611	Sample A601
Sample size	39	38
$\bar{D}$, mean kernel diameter of sample, μm	362.0	352.4
Mean relative ^{235}U concentration		
All particles	1.000 ±0.0182	1.000 ±0.0109
Particles with kernel diameter $>\bar{D}$	0.9987±0.0143	0.9988±0.0098
Particles with kernel diameter $<\bar{D}$	1.000 ±0.0208	1.000 ±0.0122
Density of kernel,[a] g/cm	3.102	3.034
Uranium in kernel,[a] wt %	71.57±0.19	79.02±24
^{235}U enrichment,[a] %	92.97	93.00
Average ^{235}U per particle measured with absolute delayed-neutron activation analysis, μg	51.6±1	51.0±1

[a]Properties obtained from Procedure No. MET-FCT-DP-10.

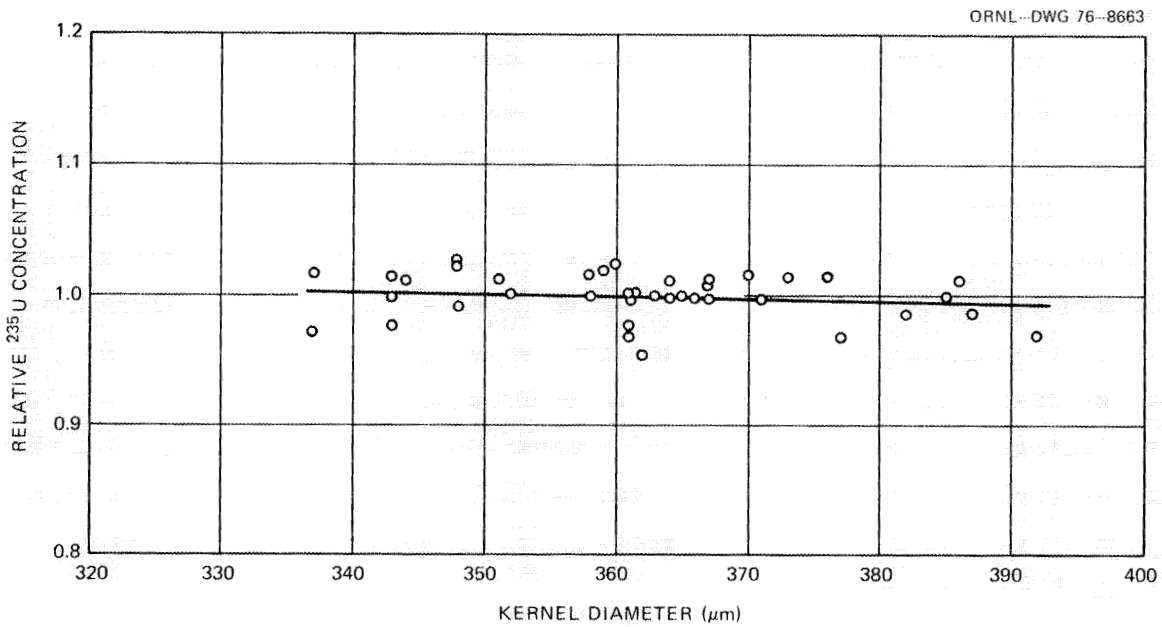

Fig. 5. Relative ^{235}U concentration of fuel kernels from batch A611.

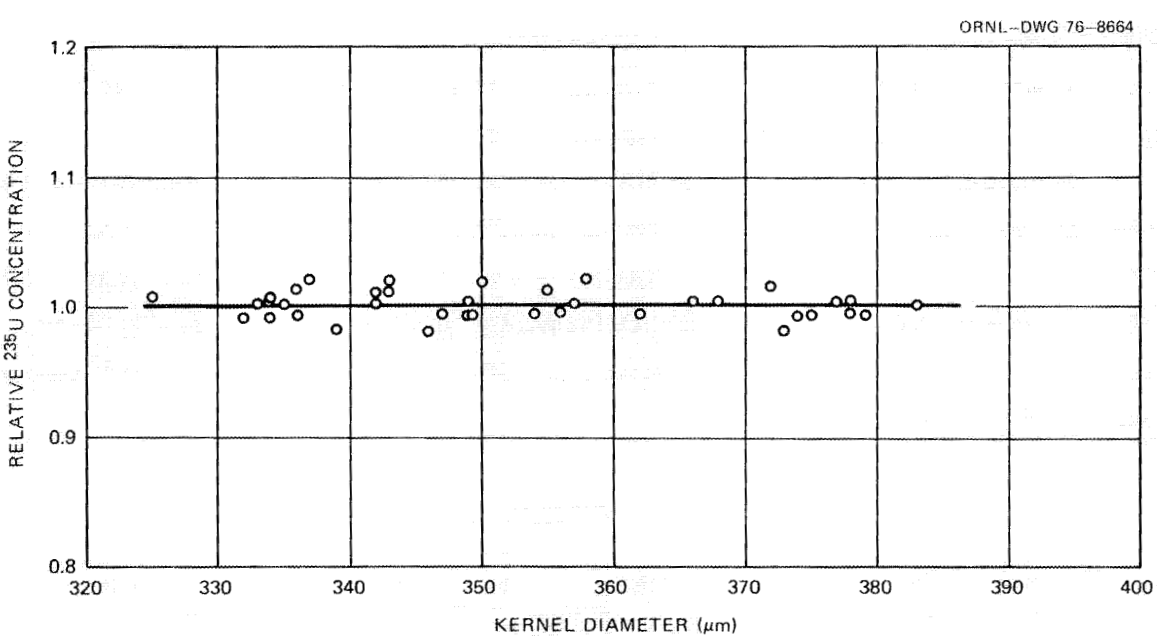

Fig. 6. Relative ^{235}U concentration of fuel kernels from batch A601.

experiment. The slope of the A601 line is essentially zero, indicating no variation in uranium concentration with kernel diameter. The relative standard deviation in the average ^{235}U concentration of all 39 kernels from batch A611 is ±1.8%, compared to ±1.1% for the 38 kernels in batch A601.

Another way of determining size-dependent uranium concentration variation is by comparing the average ^{235}U concentration of kernels with diameters smaller than the sample mean diameter $\overline{D}$ with the average concentration of kernels with diameters greater than $\overline{D}$. In sample A611, the normalized average relative ^{235}U concentration for the particles with kernel diameters greater than $\overline{D}$ (362 μm) was 0.9987 ± 0.0143 vs 1.000 ± 0.0208 for kernels smaller than $\overline{D}$. In sample A601, the average ^{235}U concentration of the larger kernels (diameter >352 μm) was 0.9988 ± 0.0098 vs 1.000 ±0.0122 for the smaller kernels.

UNCERTAINTY IN THE RELATIVE MEASUREMENTS

Some of the uncertainty in the relative uranium concentration is due to the experimental equipment and to the correction factors applied to the counts. To estimate the error due to the irradiation and counting facility, three nearly identical A611 fuel particles were irradiated and counted four times each over a time span of several hours. The standard deviation in the delayed-neutron counts of each of these particles was about 0.3%, which can be attributed to the equipment itself.

The error in the correction factors is difficult to determine because the minute-to-minute neutron flux variations and the uncertainty in the detector deadtime were not accurately known. Because the total magnitude of the corrections was <15% for the largest particles, relatively large errors in the correction factors would affect the relative uranium measurements by only a few percent.

ABSOLUTE ^{235}U CONTENT OF KERNELS

The absolute amount of ^{235}U in each particle, W_i, was determined as $W_i = C_i/a$, where W_i is micrograms of ^{235}U in particle i, C_i is corrected delayed-neutron counts in particle i, and a is delayed-neutron counts per

microgram of ^{235}U in UO_2 standards. The constant "a" was found by dividing the corrected delayed-neutron counts of each of the six UO_2 monitors by the weight of ^{235}U in the monitor and averaging the resulting six numbers. The graphed points in Figs. 7 and 8 give the absolute ^{235}U content of each particle as determined by the delayed-neutron method.

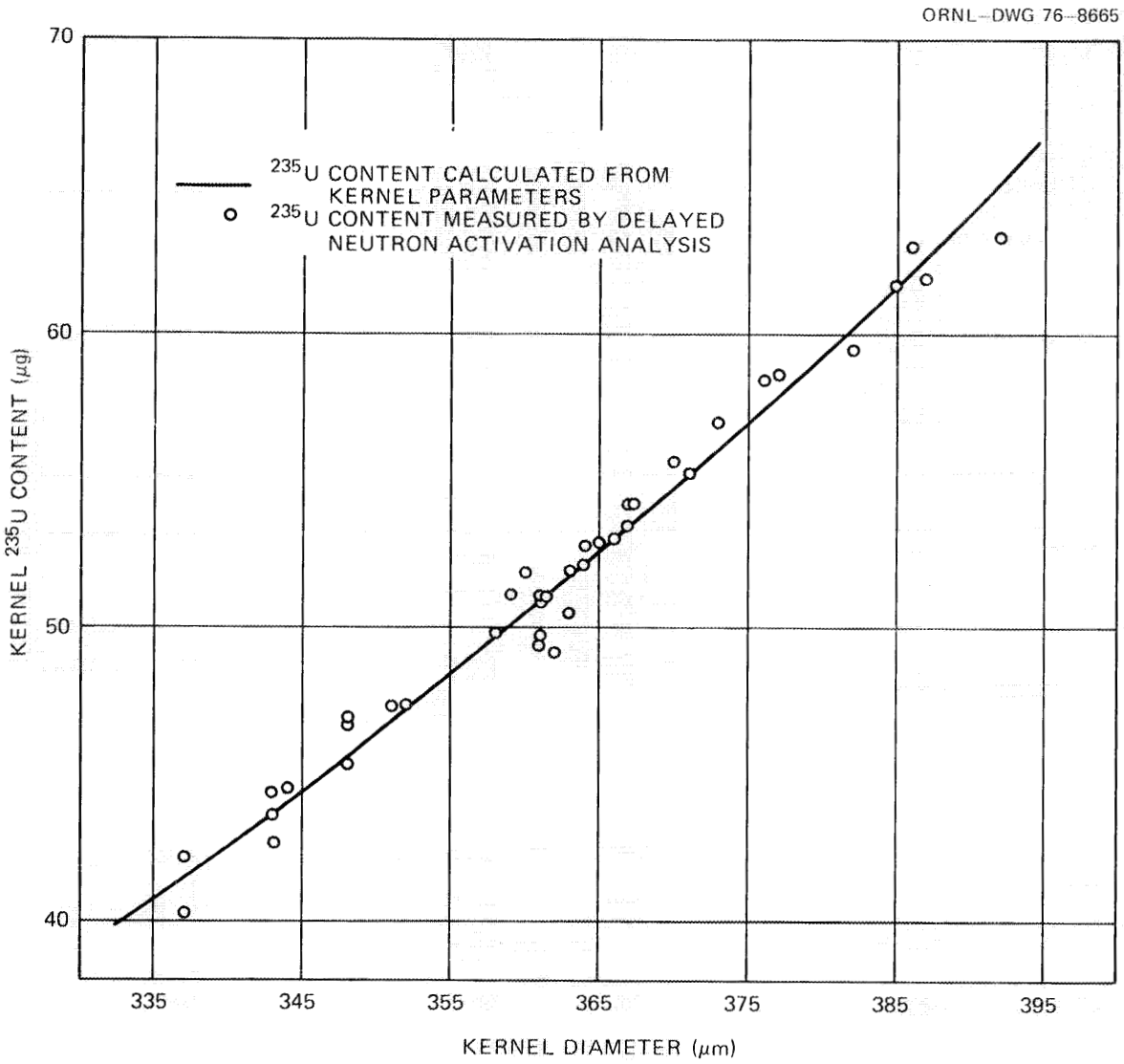

Fig. 7. Comparison of measured kernel ^{235}U content with calculated values for sample A611.

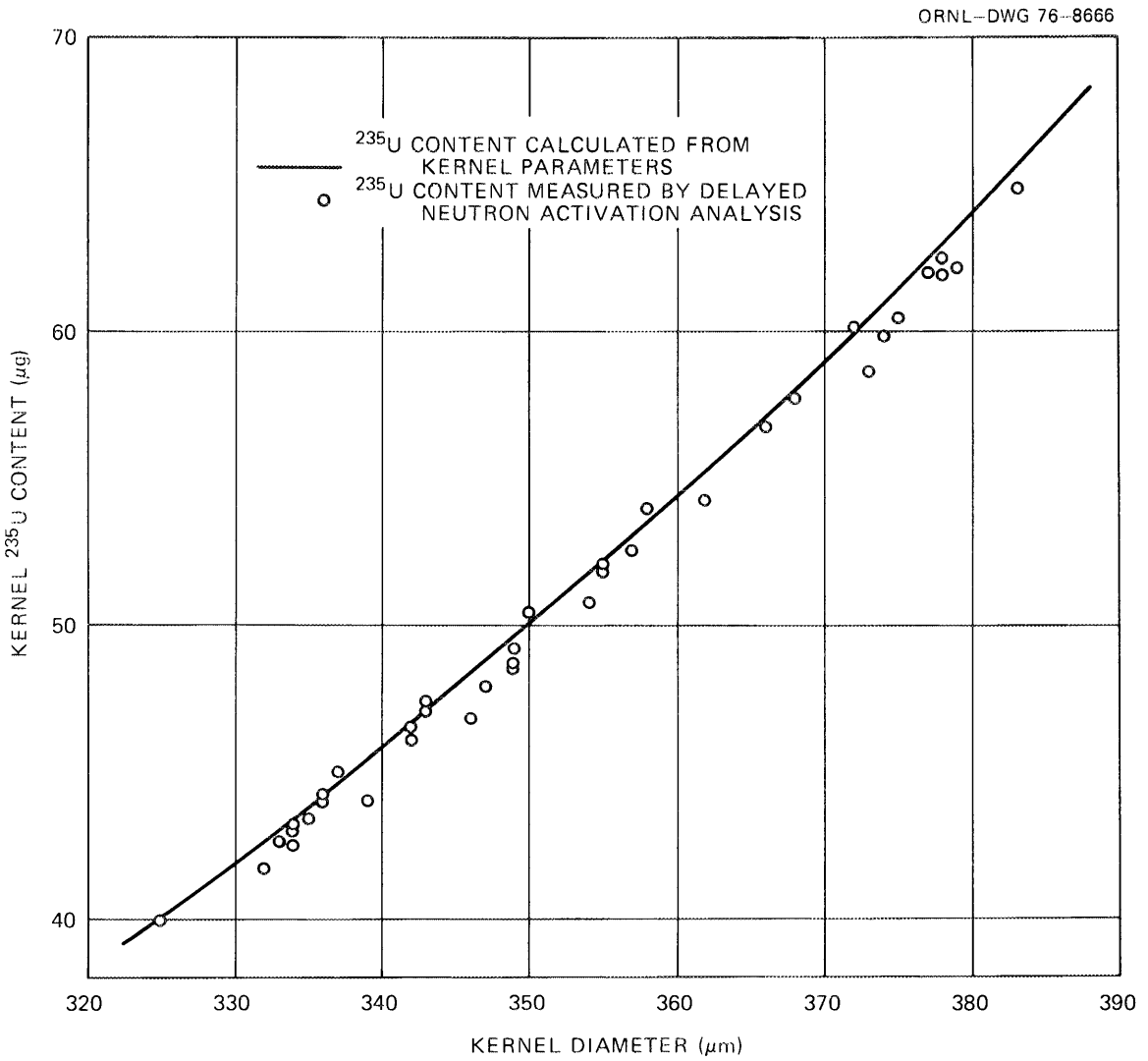

Fig. 8. Comparison of measured ^{235}U content with calculated values for sample A601.

To form a basis of comparison for these absolute measurements, the ^{235}U content of each particle was also calculated as follows using data derived from chemical analysis of the kernel batches:

$$U = 10^{-6}\ \rho V x\ ,$$

where

U = micrograms of ^{235}U in the kernel,

ρ = kernel density, g/cm^3 (see Table 1),

x = weight fraction of ^{235}U in kernel,

V = kernel volume = $(\Pi/6)D^3$, where D = kernel diameter, μm.

The ^{235}U content of the kernels calculated in this manner is given by the curves in Figs. 7 and 8. Comparison of the measured with the calculated ^{235}U content shows good agreement between the delayed-neutron method and the indirect calculation. The average difference between W_i and the corresponding calculated value in either sample is 0.70 μg of ^{235}U per particle, or about 1.3%. This small difference indicates that the absolute delayed-neutron method gives reasonable results despite the correction factors that were applied to the particles and standards.

CONCLUSIONS

The relative measurements showed that the uranium concentration in both the A611 and A601 kernels is uniform and does not vary significantly with kernel size. The close agreement of the measured absolute uranium content with the calculated uranium content of the kernels showed that the measurement technique is quite accurate in spite of the uncertainties in the correction factors and in the temporal neutron flux variations.

However, before this technique can be used to build suitable calibration standards, its accuracy would have to be improved somewhat, to about ±0.5%. The largest drawback to the method is physical limitations in the irradiation facility which allow only one particle to be assayed at a time. Because a calibration standard contains several thousand fissile particles, assaying even a fraction of these particles in this facility would be very time consuming. Hence, the delayed-neutron method would be suitable for building calibration standards only if the irradiation facility permitted the assay of many particles in a single run.

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