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Nondestructive Assay of Green HTGR Fuel Rods

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

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Nondestructive Assay of Green HTGR Fuel Rods

H. H. Barschall*

M. M. Meier

J. L. Parker

* Los Alamos Consultant. University of Wisconsin, Department of Nuclear Engineering,
Madison, WI 53706

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NONDESTRUCTIVE ASSAY OF GREEN HTGR FUEL RODS

by

H. H. Barschall, M. M. Meier, and J. L. Parker

ABSTRACT

This report describes the nondestructive (NDA) work done at Los Alamos during 1979 and 1980 as part of the New Brunswick Laboratory-sponsored evaluation of NDA of the uranium content of fabricated fuel rods for high-temperature gas-cooled reactors (HTGR). The methods used (delayed neutron and passive gamma ray) are concisely described, and the results are summarized and compared in graphical and tabular form. The results indicate that, with the use of proper physical standards, accuracies within about 1 per cent should be achievable by NDA procedures.

I. INTRODUCTION

A. History and Purpose

In February 1979 the New Brunswick Laboratory (NBL) invited Los Alamos National Laboratory (Los Alamos) to participate in an investigation and evaluation of nondestructive assay (NDA) of the uranium content of fabricated fuel rods for high-temperature gas-cooled reactors (HTGR). This report summarizes the measurements carried out at Los Alamos in response to this request and describes the conclusions that were reached.

The present activity is the third in a series of evaluations of NDA for HTGR fuel. The first effort occurred in 1972 and involved measurements on $(U-Th)C_2$ beads. The results of this study were reported in Reference 1. The

second effort in 1974 involved a greater variety of beads, including BISO and TRISO beads, and the results were reported in Reference 2. The present experiments studied "green" HTGR fuel rods, i.e., rods that have not been fired to remove the hydrogen.

The aim of the study was to compare NDA determinations of ^{235}U content with destructive chemical analysis. Both "relative" measurements that compare similar fuel rods and "absolute" measurements that compare the fuel rods with a uranium standard that contains a known amount of ^{235}U in a homogeneous compound or solution were performed. No effort was made to investigate whether a deliberate substitution of different materials in the fabrication of the fuel rod could be detected.

B. Samples

The fuel rods were cylinders 1.4 cm in diam and 5 cm high. They were sealed in 2-dram, snap-cap polyethylene vials. There were three batches of 48 rods. The three batches differed substantially in ^{235}U content, but within each batch the ^{235}U content was intended to be uniform within 2 per cent.

The rods contained two kinds of "particles," i.e., particles that had "kernels" of either UC_2 or ThO_2 . The uranium was 93.05 per cent ^{235}U according to the manufacturer and 92.9 ± 0.1 per cent according to a mass spectroscopic analysis performed at Los Alamos.

Table I summarizes the information provided by NBL about the approximate composition of the rods. The kernels are coated with TRISO to form particles that are embedded in a matrix of graphite particles and pitch.

C. Difficulties of NDA for Green HTGR Fuel and Associated Procedures

The principal problems in NDA studies of HTGR fuel arise from the heterogeneous distribution of the fissile components and from the hydrogen in the green rods. The heterogeneity results in self-absorption within the kernels, a considerable difficulty in passive γ -ray NDA. Since the previous NDA was carried out with fast-neutron interrogation, the green rods introduced a serious complication by the presence of hydrogen in the fuel. The hydrogen moderates the neutrons. Both the efficiency of the detector and the fission and absorption cross sections of ^{235}U are higher for the moderated neutrons. For an evaluation of the consequences of the moderation, the hydrogen content of the rods had to be determined in a separate experiment if neutron interrogation was to be used.

TABLE I
COMPOSITION OF FUEL RODS

Diameter of Kernels and Particles (μm)

	<u>Kernel</u>	<u>Particle</u>
UC ₂	199	633
ThO ₂	452	786

Weights (g)

<u>Batch No.</u>	<u>U Particles</u>	<u>Th Particles</u>	<u>Matrix</u>	<u>Total</u>	<u>²³⁵U</u>	<u>Th</u>	<u>H</u>
12	1.2	7.4	4.9	13.6	0.16	3.48	0.135
13	0.9	9.9	4.0	14.8	0.12	4.65	0.141
14	2.2	7.4	4.1	13.8	0.29	3.48	0.136

For the particular samples to be analyzed, the use of polyethylene vials made the problem of moderation enormously more difficult because the hydrogen in the vials has a different effect on the fission probability from the hydrogen in the pitch.

Because of the hydrogen problem for neutron NDA, a complementary study with passive γ -ray NDA was carried out even though the HTGR rods also present significant difficulties for that method.

An effort was also made to modify the neutron interrogation NDA by slowing the primary neutrons in a polyethylene moderator.

D. Relation of Present Experiments to NBL Project

The present report covers only the NDA measurements at Los Alamos. It does not include destructive (wet chemical) analysis to be performed at Los Alamos, nor the assays that have proceeded concurrently at NBL and General Atomic Company (GAC). A summary of all these efforts is expected to be issued in an NBL report.

II. EXPERIMENTAL METHODS

A. Hydrogen Assay

Because of the impact of hydrogen on the response of the detection systems using neutron interrogation, the hydrogen content of the samples has to be determined. The hydrogen analyser is described in Reference 3. In this device a ^{252}Cf neutron source and two ^3He neutron detectors are embedded near the center of a 30-cm steel cube. Samples are introduced into a chamber near the source and detector, and the increase in count rate of the detectors caused by the presence of hydrogen in the sample yields information about the hydrogen content.

Determinations of hydrogen content were relative to standards fabricated at Los Alamos. The standards have the same shape as the fuel rods and contain 100, 200, or 300 mg of ^{235}U , and 100 or 150 mg of hydrogen. The ^{235}U was in the form U_3O_8 , and the hydrogen in the form of dextrose. The remainder of the volume was filled with graphite. The mixture was prepared in a glass vial, which was surrounded by a polyethylene vial to resemble the samples to be analyzed.

In order to evaluate the effect of hydrogen on the neutron-based NDA, the hydrogen content of the vial and of the fuel need to be known separately. The hydrogen content of the vial was determined from the weights of the rod and vial measured at Los Alamos and rod masses provided by NBL. The difference between the hydrogen content of the vial determined from the weights and the total hydrogen content determined as described above yields the hydrogen content of the fuel. The latter showed variations among the rods of 1.6 per cent for batch 12, 2.1 per cent for 13, and 3.3 per cent for 14. The accuracy of the determinations was about 2 per cent.

B. Fast-Neutron NDA

Fast-neutron NDA used the small-sample assay system (SSAS) at the Los Alamos Van de Graaff accelerator. This system is described in Reference 4. Neutrons of energies between 120 and 500 keV are produced by bombarding a 2-mg/cm^2 Li target with 2.35-MeV protons. The proton beam is pulsed at 10-Hz repetition rate and stays on for 35 ms. While the neutron source is off, delayed neutrons from ^{235}U fission in the sample to be assayed were counted with a modified "slab detector"⁵ consisting of 39 ^3He proportional counters in a polyethylene slab.

The interrogating neutrons are energetic enough to penetrate through small samples without much absorption, but are below the ^{232}Th and ^{238}U fission thresholds. Since the fission cross section of ^{235}U increases rapidly towards low neutron energies, low-energy neutrons produced in scattering by protons will strongly influence the fission rate and hence the delayed-neutron production. The effect of the hydrogen in the vial was found to be only 13 per cent of that in the rod itself, but because of the greater abundance of hydrogen in the vial, the correction for the vial was about 12 per cent while the hydrogen in the rod required a 31 per cent correction. The presence of Th did not affect the response of the system, apparently because the effects of Th fission by ^{235}U fission neutrons and of self-absorption cancelled.

C. Moderated-Neutron NDA

The moderated mode of the neutron NDA differed from the fast mode described in Reference 4 only by the insertion of a polyethylene moderator. This moderator degrades the primary spectrum to one that more closely resembles a thermal spectrum. Although the slower neutrons are less penetrating than the fast neutrons, the hope was that the effect of the presence of hydrogen in the sample, and especially the difference between the effects of hydrogen in the vial compared with hydrogen in the rod itself, would be much smaller for the slow neutrons.

D. Gamma-Ray NDA

Passive γ -ray assay is one of the most widely used methods for determining uranium content in special nuclear materials.⁶ Quantitative assays are difficult because of the large absorption of the 186-keV γ rays emitted by ^{235}U , especially in the high-Z elements in the sample. This absorption is usually corrected on the basis of a transmission measurement⁷ with an external source that emits γ rays of energies in the range of those emitted by ^{235}U . Such a measurement determines the linear attenuation coefficient and permits the calculation of a correction factor (CF) that is the ratio of the count from a sample that would have been obtained if the sample had no self-absorption to the actual count. CF has been calculated⁷ for various simple geometries both for point sources in an absorbing medium and for homogeneous samples. Calculations of CF have been performed as a function of the distance between sample and detector.

Although the fuel rods have a simple external geometry, i.e., are cylindrical, the heterogeneity of the distribution of the high-Z kernels makes the calculation of CF difficult. Each kernel absorbs ~10 per cent of the radiation emitted inside the kernel. Although there are on the order of 10 000 kernels in a rod, the small diameter of the kernels results in the presence of few kernels along any ray along the diameter of the cylinder. In fact the most probable number of kernels along a diameter is one.

The transmission of a broad beam of 186-keV γ rays through the cylindrical rod is ~25 per cent, while the transmission would be expected to be only ~18 per cent if fuel rods were homogenized. Although the difference between the transmission of the actual rod and a homogenized rod is large, a first approximation is to assume that CF can be calculated in the same way as for a homogeneous rod, and then correct for the self-absorption in the kernel. This turns out to be not a very accurate approximation primarily because the coating of the kernel imposes a minimum distance between kernels, a distance that is an appreciable fraction of the rod diameter. Although an estimate of this effect was included in the corrections, the overall accuracy of the comparison of an HTGR rod with a homogeneous standard is estimated to be about 2 per cent, while the comparison of different HTGR rods with each other could probably be performed with better than 1 per cent accuracy.

E. Standards

The absolute determination of ^{235}U relies on a comparison with standards prepared by the Los Alamos Analytical Chemistry Group. For the neutron assay the standards had to have a hydrogen content similar to that of the HTGR rods. The standards are powders consisting of a mixture of U_3O_8 , dextrose, and graphite. The ^{235}U content of the nine standards varied from about 0.1 to 0.3 g.

For the γ -ray assay where the hydrogen content was not important, solution standards were prepared, since solutions provide samples of greater uniformity, and uniformity is most important for the γ -ray measurements. The ^{235}U content of the three standards varied from about 0.8 g to 4 g.

In addition, one of the HTGR rods of each of three batches was assayed by analytical chemistry for ^{235}U content. Prior to this assay each of these rods had been carefully compared with a rod of the same batch to provide transfer standards that could be used for further comparisons. Comparisons

with transfer standards will be called "relative" measurements, while comparisons with the prepared powder or solution standards will be referred to as "absolute" measurements.

III. RESULTS

A. Fast Neutrons

Preliminary experiments showed that the response of the system was linear with ^{235}U content, independent of Th content, but very sensitive to hydrogen content. The system was calibrated initially with the nine prepared standards and the three transfer standards. Measurements with standards were interspersed with measurements of unknowns to correct for short-term drifts in the neutron source or in the detection system.

The results of the measurements are given in Table II. The overall uncertainty that is given includes the uncertainty from counting statistics, which is a major source of uncertainty in these measurements, and the uncertainty introduced by errors in the hydrogen content of both the vial and the rod. Additional smaller uncertainties are introduced by variations in the height of the cylindrical samples and by the initial calibration. Both absolute measurements against the prepared standards and relative measurements against the transfer standards are tabulated.

B. Moderated Neutrons

Although the hope was that the use of moderated neutrons would reduce the uncertainties introduced by the hydrogen in the sample, preliminary calibration runs showed a large dependence of the response on the hydrogen content of the polyethylene vial. The reason for this dependence is not understood. The effect should be investigated further if moderated neutrons are to be used again with samples in polyethylene vials. The dependence on the hydrogen content of the rods was reduced as expected. The response to ^{235}U content was nonlinear, presumably because of self-absorption of slow neutrons in the sample, and there was a small dependence on Th content, for the same reason.

The count rate for the moderated neutrons was increased by a factor of the order of ten over that for fast neutrons because of the high detection efficiency of the counters.

The results of the measurements are shown also in Table II. The uncertainties caused by counting statistics are a small contribution to the overall

TABLE II
NEUTRON ASSAY RESULTS

BATCH 12

Rod No.	Absolute Fast-n		Absolute Mod.-n		Relative Fast-n		Relative Mod.-n	
	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)
1201	0.162	2.88	0.165	3.12	0.161	1.56	0.164	2.21
1202	0.160	2.89	0.161	2.28	0.159	1.55	0.160	1.17
1203	0.162	2.87	0.162	2.22	0.162	1.55	0.160	1.13
1204	0.161	2.87	0.163	2.36	0.160	1.56	0.162	1.26
1205	0.161	2.89	0.166	3.12	0.160	1.56	0.165	2.21
1206	0.160	2.87	0.163	2.66	0.159	1.55	0.162	1.64
1207	0.160	2.86	0.169	3.17	0.160	1.63	0.168	2.30
1208	0.163	2.85	0.166	2.84	0.163	1.57	0.165	1.88
1209	0.158	2.93	0.167	3.88	0.157	1.57	0.165	3.10
1210	0.159	2.87	0.162	2.38	0.159	1.57	0.161	1.27
1211	0.160	2.90	0.164	2.99	0.160	1.56	0.163	2.04
1212	0.160	2.89	0.161	2.32	0.159	1.55	0.160	1.20
1213	0.161	2.87	0.160	2.38	0.160	1.56	0.159	1.26
1214	0.162	2.87	0.167	2.95	0.162	1.56	0.166	2.02
1215	0.160	2.89	0.162	2.70	0.159	1.56	0.161	1.67
1216	0.164	2.87	0.168	3.66	0.163	1.56	0.167	2.86
1217	0.161	2.86	0.161	2.29	0.161	1.55	0.160	1.17
1218	0.162	2.86	0.163	2.66	0.161	1.56	0.162	1.63
1219	0.159	2.91	0.165	3.89	0.158	1.57	0.164	3.10
1220	0.160	2.88	0.165	3.39	0.159	1.56	0.164	2.53
1221	0.160	2.86	0.164	3.03	0.160	1.56	0.163	2.10
1222	0.161	2.90	0.167	3.64	0.161	1.56	0.166	2.82
1223	0.162	2.89	0.169	3.92	0.162	1.56	0.168	3.15
1224	0.164	2.87	0.164	2.91	0.163	1.54	0.163	1.92
1225	0.160	2.89	0.161	2.46	0.159	1.56	0.160	1.37
1226	0.159	2.91	0.164	3.18	0.158	1.58	0.163	2.28
1227	0.163	2.87	0.164	2.70	0.162	1.57	0.163	1.69
1228	0.164	2.85	0.162	2.26	0.163	1.56	0.161	1.15
1229	0.162	2.91	0.164	2.89	0.161	1.57	0.163	1.94
1230	0.166	2.83	0.165	2.31	0.165	1.56	0.164	1.22
1231	0.160	2.89	0.165	3.38	0.159	1.60	0.164	2.52
1232	0.161	2.91	0.167	3.96	0.160	1.58	0.166	3.12
1233	0.162	2.87	0.165	2.35	0.162	1.58	0.163	1.26
1234	0.162	2.89	0.165	3.08	0.161	1.58	0.164	2.17
1235	0.160	2.89	0.163	2.57	0.159	1.59	0.162	1.53
1236	0.162	2.88	0.165	3.11	0.161	1.59	0.163	2.20
1237	0.161	2.87	0.164	2.34	0.160	1.58	0.163	1.24
1238	0.161	2.88	0.164	2.67	0.160	1.59	0.162	1.66
1239	0.159	2.88	0.161	2.21	0.158	1.58	0.160	1.12
1240	0.163	2.85	0.164	2.42	0.162	1.58	0.163	1.35
1241	0.163	2.84	0.163	2.26	0.162	1.58	0.162	1.15
1242	0.165	2.84	0.166	2.38	0.164	1.57	0.164	1.31
1243	0.162	2.87	0.164	2.57	0.161	1.57	0.163	1.54
1244	0.163	2.86	0.164	2.70	0.163	1.57	0.163	1.69
1245	0.164	2.86	0.167	3.31	0.163	1.59	0.166	2.45
1246	0.163	2.87	0.169	3.41	0.163	1.58	0.168	2.57
1247	0.161	2.89	0.167	3.17	0.161	1.58	0.165	2.28
1248	0.163	2.87	0.168	2.96	0.163	1.57	0.167	2.03
AVERAGE	0.162		0.164		0.161		0.163	

TABLE II (cont)
NEUTRON ASSAY RESULTS
BATCH 13

Rod No.	Absolute Fast-n		Absolute Mod.-n		Relative Fast-n		Relative Mod.-n	
	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)
1301	0.118	3.39	0.122	3.78	0.122	1.87	0.127	2.54
1302	0.120	3.32	0.122	2.97	0.124	1.89	0.127	1.70
1303	0.125	3.29	0.125	2.75	0.129	1.86	0.129	1.51
1304	0.118	3.40	0.118	2.74	0.122	1.88	0.123	1.45
1305	0.121	3.37	0.122	3.54	0.125	1.86	0.127	2.30
1306	0.123	3.26	0.123	2.51	0.127	1.87	0.128	1.30
1307	0.121	3.32	0.121	2.16	0.125	1.86	0.125	1.26
1308	0.125	3.25	0.124	2.19	0.130	1.88	0.129	1.20
1309	0.116	3.42	0.119	3.82	0.120	1.88	0.124	2.56
1310	0.119	3.35	0.120	2.67	0.123	1.89	0.125	1.40
1311	0.125	3.22	0.123	2.12	0.129	1.89	0.128	1.32
1312	0.121	3.29	0.125	3.27	0.126	1.90	0.130	2.03
1313	0.123	3.25	0.124	2.25	0.127	1.92	0.129	1.19
1314	0.118	3.32	0.121	2.45	0.123	1.92	0.126	1.25
1315	0.122	3.34	0.127	3.64	0.126	1.89	0.132	2.42
1316	0.121	3.25	0.122	2.11	0.126	1.89	0.127	1.56
1317	0.121	3.28	0.122	2.57	0.125	1.90	0.127	1.33
1318	0.124	3.23	0.126	2.60	0.129	1.89	0.131	1.38
1319	0.118	3.29	0.117	2.17	0.122	1.90	0.121	1.32
1320	0.123	3.28	0.123	2.41	0.127	1.89	0.127	1.23
1321	0.125	3.26	0.125	3.18	0.130	1.89	0.130	1.94
1322	0.123	3.31	0.125	3.32	0.127	1.89	0.129	2.07
1323	0.123	3.31	0.123	2.69	0.126	1.87	0.128	1.44
1324	0.122	3.31	0.124	2.46	0.127	1.87	0.129	1.27
1325	0.121	3.33	0.121	2.12	0.125	1.89	0.125	1.52
1326	0.117	3.36	0.117	2.55	0.121	1.90	0.121	1.29
1327	0.125	3.23	0.128	3.70	0.129	1.86	0.133	2.50
1328	0.120	3.29	0.122	2.94	0.125	1.90	0.126	1.67
1329	0.117	3.43	0.119	3.88	0.121	1.90	0.123	2.62
1330	0.123	3.28	0.123	2.22	0.128	1.88	0.128	1.19
1331	0.123	3.28	0.123	2.12	0.128	1.89	0.128	1.28
1332	0.123	3.24	0.122	2.23	0.128	1.91	0.126	1.19
1333	0.120	3.31	0.122	2.50	0.124	1.95	0.127	1.27
1334	0.124	3.27	0.128	3.04	0.128	1.91	0.133	1.81
1335	0.125	3.28	0.126	2.94	0.129	1.88	0.131	1.69
1336	0.120	3.33	0.122	2.77	0.124	1.87	0.126	1.50
1337	0.124	3.30	0.124	2.96	0.128	1.89	0.129	1.70
1338	0.121	3.27	0.123	2.37	0.126	1.88	0.127	1.21
1339	0.124	3.25	0.128	2.47	0.128	1.87	0.132	1.29
1340	0.125	3.22	0.127	2.72	0.129	1.88	0.132	1.50
1341	0.119	3.33	0.120	2.43	0.123	1.88	0.124	1.22
1342	0.125	3.24	0.126	2.71	0.129	1.88	0.131	1.46
1343	0.120	3.33	0.125	3.67	0.124	1.85	0.130	2.44
1344	0.125	3.25	0.127	2.66	0.130	1.88	0.132	1.43
1345	0.122	3.26	0.125	2.39	0.127	1.87	0.130	1.22
1346	0.118	3.32	0.122	3.06	0.122	1.87	0.126	1.78
1347	0.116	3.41	0.121	3.79	0.120	1.87	0.126	2.54
1348	0.118	3.30	0.126	3.07	0.122	1.94	0.128	1.81
AVERAGE	0.121		0.123		0.126		0.128	

TABLE II (cont)
NEUTRON ASSAY RESULTS
BATCH 14

Rod No.	Absolute Fast-n		Absolute Mod.-n		Relative Fast-n		Relative Mod.-n	
	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)	Mass ²³⁵ U (g)	Overall Rel. St. Dev. (%)
1401	0.282	2.23	0.288	2.04	0.280	1.36	0.282	1.43
1402	0.281	2.22	0.286	2.05	0.280	1.37	0.280	1.44
1403	0.285	2.23	0.294	2.40	0.283	1.37	0.288	1.94
1404	0.282	2.23	0.287	2.15	0.280	1.37	0.281	1.60
1405	0.272	2.29	0.287	2.68	0.270	1.37	0.281	2.27
1406	0.278	2.28	0.289	2.64	0.276	1.36	0.283	2.23
1407	0.286	2.26	0.293	2.50	0.284	1.36	0.287	2.06
1408	0.277	2.27	0.294	3.27	0.275	1.38	0.288	2.97
1409	0.273	2.30	0.283	2.54	0.271	1.37	0.277	2.09
1410	0.277	2.26	0.286	2.11	0.275	1.37	0.280	1.54
1411	0.276	2.22	0.294	2.75	0.274	1.41	0.288	2.37
1412	0.281	2.20	0.294	2.27	0.279	1.39	0.288	1.78
1413	0.281	2.23	0.291	2.47	0.279	1.37	0.285	2.02
1414	0.273	2.24	0.288	2.65	0.271	1.39	0.282	2.24
1415	0.281	2.21	0.293	2.42	0.279	1.37	0.287	1.97
1416	0.282	2.22	0.290	2.41	0.280	1.38	0.284	1.94
1417	0.278	2.26	0.285	2.23	0.277	1.36	0.279	1.69
1418	0.281	2.26	0.292	2.47	0.279	1.36	0.286	2.02
1419	0.276	2.26	0.286	2.84	0.274	1.38	0.280	2.46
1420	0.286	2.26	0.297	2.78	0.284	1.37	0.291	2.41
1421	0.273	2.35	0.282	2.14	0.271	1.52	0.276	1.57
1422	0.276	2.35	0.287	2.89	0.274	1.51	0.281	2.52
1423	0.282	2.31	0.290	2.58	0.280	1.51	0.284	2.16
1424	0.278	2.30	0.291	2.48	0.276	1.53	0.285	2.04
1425	0.281	2.32	0.293	2.72	0.279	1.51	0.287	2.33
1426	0.272	2.34	0.283	2.38	0.270	1.52	0.277	1.89
1427	0.275	2.35	0.281	2.32	0.273	1.51	0.275	1.81
1428	0.284	2.31	0.293	2.29	0.282	1.50	0.286	1.80
1429	0.261	2.38	0.278	2.08	0.259	1.51	0.272	1.46
1430	0.282	2.30	0.293	2.81	0.281	1.51	0.287	2.44
1431	0.279	2.35	0.297	3.36	0.277	1.51	0.291	3.07
1432	0.281	2.30	0.292	2.40	0.279	1.51	0.286	1.93
1433	0.271	2.43	0.287	3.13	0.269	1.50	0.281	2.80
1434	0.278	2.38	0.287	2.27	0.276	1.50	0.281	1.76
1435	0.278	2.37	0.290	2.80	0.277	1.53	0.284	2.43
1436	0.283	2.30	0.299	3.43	0.281	1.50	0.293	3.16
1437	0.275	2.44	0.282	2.39	0.273	1.50	0.276	1.91
1438	0.277	2.36	0.281	2.04	0.275	1.51	0.275	1.42
1439	0.281	2.33	0.293	2.49	0.279	1.49	0.287	2.05
1440	0.274	2.39	0.291	3.28	0.272	1.50	0.284	2.97
1441	0.280	2.35	0.286	2.04	0.279	1.51	0.280	1.44
1442	0.275	2.39	0.288	2.82	0.273	1.50	0.282	2.44
1443	0.278	2.34	0.286	2.58	0.276	1.50	0.280	2.15
1444	0.284	2.29	0.293	2.11	0.282	1.50	0.286	1.55
1445	0.285	2.31	0.298	2.71	0.283	1.50	0.292	2.33
1446	0.276	2.39	0.292	3.32	0.274	1.50	0.285	3.03
1447	0.277	2.36	0.284	2.51	0.275	1.50	0.278	2.05
1448	0.283	2.31	0.293	2.33	0.281	1.51	0.286	1.85
AVERAGE	0.279		0.289		0.277		0.283	

uncertainty. The largest uncertainty is that caused by the uncertainty in the hydrogen content of the vial. Again both absolute and relative determinations are tabulated.

C. Gammas

In the γ -ray assay only a third of the rods, i.e., 16 of each batch, were assayed. The 185.7-keV γ rays from ^{235}U were detected in 4 Ge-Li detectors that were placed at 90° intervals around the rod. Transmissions were measured with a ^{169}Yb source.

Two types of measurements were performed: absolute measurements in which the γ -ray emission from the unknown was compared with that of a solution standard of known ^{235}U content, and relative measurements in which the comparison was made with transfer standards. The transfer standards had been compared with a rod whose ^{235}U content has been determined by analytical chemistry. The absolute and relative measurements on all the rods differ by a factor of 1.015, the absolute measurements being larger.

For absolute measurements the principal source of uncertainty is the correction for the heterogeneity of the rods. For relative measurements the principal uncertainty is in the knowledge of the ^{235}U content of the transfer standards. The estimated uncertainty in the chemical determinations ranged from about 0.1 per cent to 0.6 per cent, and there is an additional uncertainty introduced by the comparison between the transfer standard and the rod that underwent chemical analysis.

Table III lists the results for the 48 rods that were assayed both in the relative and in the absolute measurements. The uncertainty given in the table is based on counting statistics only and does not contain any estimate of uncertainty due to systematic or calibration error.

D. Consistency

For all 144 rods the results of the two neutron assays may be compared. Figure 1 shows a histogram that gives the number of cases in which these two relative measurements differed by the percentage marked along the axis of abscissae. The differences are shown separately for each of the three batches and, at the top of the figure, for all the rods.

Gamma-ray assays were performed for only a third of the rods. Figure 2 shows a histogram comparing relative γ -ray assays with relative fast-neutron assays, while in Fig. 3 relative γ -ray assays are compared with relative moderated-neutron assays.

TABLE III
GAMMA-RAY ASSAY RESULTS

BATCH 12

Rod No.	Absolute Gamma		Relative Gamma	
	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)
1203	0.1673	0.30	0.1648	0.30
1206	0.1665	0.30	0.1641	0.30
1209	0.1671	0.30	0.1646	0.30
1212	0.1649	0.30	0.1624	0.30
1215	0.1628	0.30	0.1604	0.30
1218	0.1670	0.30	0.1645	0.30
1221	0.1652	0.30	0.1627	0.30
1224	0.1659	0.30	0.1635	0.30
1227	0.1632	0.30	0.1608	0.30
1230	0.1677	0.30	0.1652	0.30
1234	0.1657	0.30	0.1632	0.30
1236	0.1650	0.30	0.1626	0.30
1239	0.1638	0.30	0.1614	0.30
1242	0.1694	0.30	0.1669	0.30
1245	0.1668	0.30	0.1644	0.30
1248	0.1677	0.30	0.1652	0.30
AVERAGE	0.1660		0.1635	

BATCH 13

Rod No.	Absolute Gamma		Relative Gamma	
	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)
1303	0.1282	0.40	0.1263	0.40
1306	0.1273	0.40	0.1255	0.40
1309	0.1209	0.40	0.1191	0.40
1312	0.1267	0.40	0.1248	0.40
1315	0.1290	0.40	0.1271	0.40
1318	0.1271	0.40	0.1252	0.40
1321	0.1265	0.40	0.1247	0.40
1324	0.1273	0.40	0.1254	0.40
1327	0.1280	0.40	0.1261	0.40
1330	0.1274	0.40	0.1255	0.40
1333	0.1258	0.40	0.1239	0.40
1336	0.1231	0.40	0.1212	0.40
1340	0.1297	0.40	0.1278	0.40
1342	0.1272	0.40	0.1253	0.40
1345	0.1286	0.40	0.1267	0.40
1348	0.1253	0.40	0.1234	0.40
AVERAGE	0.1268		0.1249	

^a The relative standard deviation given here is based on counting statistics only. It does not contain any estimates of systematic or calibration error.

TABLE III (cont)
GAMMA-RAY ASSAY RESULTS

BATCH 14

Rod No.	Absolute Gamma		Relative Gamma	
	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)	Mass ²³⁵ U (g)	Rel. St. Dev. ^a (%)
1404	0.2958	0.25	0.2914	0.25
1406	0.2876	0.25	0.2833	0.25
1409	0.2792	0.25	0.2751	0.25
1412	0.2908	0.25	0.2865	0.25
1415	0.2928	0.25	0.2884	0.25
1418	0.2876	0.25	0.2833	0.25
1421	0.2799	0.25	0.2758	0.25
1424	0.2931	0.25	0.2888	0.25
1427	0.2856	0.25	0.2814	0.25
1430	0.2926	0.25	0.2883	0.25
1433	0.2777	0.25	0.2736	0.25
1436	0.2945	0.25	0.2902	0.25
1440	0.2868	0.25	0.2826	0.25
1443	0.2828	0.25	0.2786	0.25
1445	0.2912	0.25	0.2869	0.25
1448	0.2902	0.25	0.2860	0.25
AVERAGE		0.2880	0.2838	

^a The relative standard deviation given here is based on counting statistics only. It does not contain any estimates of systematic or calibration error.

The comparisons presented in Figs. 1-3 show only whether the three NDA methods are consistent. They do not give information about the accuracy of the ²³⁵U content determination, since all NDA data presented in the figures are relative measurements based on the same transfer standards.

E. Accuracy

The uncertainties given in Tables II and III are estimates of the NDA errors and do not include possible errors in the analytical chemistry or mass spectroscopy. NDA comparisons of the transfer standards showed differences outside the errors expected on the basis of the uncertainties in relative NDA measurements and in chemical methods. Similar differences were observed in NDA comparisons of prepared solution standards. In addition, as noted in Section IB, there was a difference between the manufacturer's analysis and the Los Alamos mass spectroscopic uranium isotopic analysis. Since the determination of ²³⁵U content by analytical chemistry depends on a knowledge of the isotopic composition, any uncertainty in this composition affects the results.

Fig. 1. Comparison of relative moderated-neutron assays with relative fast-neutron assays.

Fig. 2 Comparison of relative fast-neutron assays with relative γ -ray assays.

Fig. 3 Comparison of relative moderated-neutron assays with relative γ -ray assays.

IV. COMPARISON OF NDA METHODS

For neutron measurements the presence of hydrogen in the samples is the principal problem. The presence of extraneous hydrogen in the vial prevented the present study from being a fair test of neutron-based NDA because the hydrogen in the vial has a different effect from the hydrogen in the rod, and hence introduced unnecessary uncertainties. In principle, neutron NDA has the advantage over γ -ray NDA in that the neutrons are more penetrating than the ^{235}U γ rays so that heterogeneities in the sample and variations in the random arrangement of the kernels in the rod should have little influence on the assay.

Moderating the neutrons increases the count rate of the detector by an order of magnitude and hence reduces assay time for a desired precision. Evidence was obtained that moderation of the neutrons reduces dependence on hydrogen content. Moderation may, however, introduce a nonlinear dependence on ^{235}U content and a dependence on Th content because the moderated neutrons are less penetrating. Therefore the advantage of higher precision obtained with the moderated neutrons and the advantage of less dependence on hydrogen content are partly offset by their more rapid absorption.

Gamma-ray measurements are not affected by the presence of hydrogen, but they are affected by the self-absorption in the rod. Because of the difficulty of calculating the effects of the heterogeneities, the greatest problem arises if comparison with a homogeneous standard is desired. It appears likely that rearrangement of the particles in a rod will affect the number of γ rays that are absorbed in the rod both in the emission and in the transmission measurement. An estimate of the magnitude of this effect is difficult, but it may introduce an uncertainty of as much as 0.5 per cent in the mass determination. There is an additional 0.5 to 1 per cent uncertainty introduced if the comparison of an HTGR rod is made with a homogeneous standard.

The time required to obtain 1 per cent precision was about 800 s in the fast-neutron assay, 200 s in the moderated-neutron assay, and 1000 s in the γ -ray assay. These times could be shortened by increasing the number of detectors or by other experimental changes.

Operation of a pulsed neutron source based on a Van de Graaff accelerator requires more highly skilled staff than that needed for the γ -ray assay. The neutron source is more expensive and more subject to breakdown than the equipment needed for γ -ray measurements. On the other hand, at least in principle,

the neutron measurements should be less affected by the heterogeneous nature of the samples.

On the basis of the present measurements an uncertainty of 1 per cent should be achievable with the NDA procedures. Whether uncertainties of less than 1 per cent could be reached by either neutron or γ -rays NDA appears doubtful, unless there is a major development effort.

V. RECOMMENDATIONS

For neutron NDA, rods should be encapsulated in hydrogen-free material. For γ -ray NDA, HTGR rod standards with accurately known ^{235}U content should be developed.

If another evaluation of this type is carried out, we suggest that fewer samples be used. This would permit attainment of higher precision in a given time period. We would suggest further that, at an early stage of the investigation, ratios of the fissile content of both transfer and prepared standards be compared by NDA and analytical chemistry in order to eliminate inconsistencies before a large number of measurements are performed.

Results obtained at different laboratories could be compared in a more meaningful way, if the standards used for NDA measurements were interchanged between the laboratories. This would elucidate whether differences in results are due to NDA methods or to standards.

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