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The Feasibility of Nondestructive Assay Measurements in Uranium Enrichment Plants

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THE FEASIBILITY OF NONDESTRUCTIVE ASSAY MEASUREMENTS IN URANIUM ENRICHMENT PLANTS

by

Roddy B. Walton

ABSTRACT

Applications of nondestructive assay (NDA) methods to a wide range of measurement problems in uranium enrichment facilities are reviewed. These methods are of interest for domestic and international safeguards of enrichment facilities, as well as for process control and efficient materials management. The results of a number of test and evaluation projects that were performed over the last decade at the Oak Ridge Gaseous Diffusion Plant (ORGDP), which is operated by the Union Carbide Corporation--Nuclear Division, and the Portsmouth (GAT) Plant, operated by the Goodyear Atomic Corporation, are presented. Measurements of the residual holdup in the top enrichment portion of the shut-down K-25 cascade were made with portable neutron and gamma-ray detectors, and inventory estimates based on these data were in good agreement with ORGDP estimates. In the operating cascade, the tests showed that portable NaI detectors are effective for monitoring NaF and alumina media for gaseous effluent traps and that gas phase enrichments and inventories, as well as large deposits of uranium, can be detected with portable neutron and gamma-ray instrumentation. A wide variety of scrap and waste materials, including barrier and compressor blades, incinerator ash and trapping

media, and miscellaneous waste, were measured using passive gamma-ray and neutron methods and 14-MeV neutron interrogation. Methods developed for rapid verification of UF_6 in shipping containers with portable neutron and gamma-ray instruments are now used routinely by safeguards inspectors. Passive assay methods can also be used to measure continuously the enrichments of ^{235}U and ^{234}U in the UF_6 product and tails withdrawals of a gaseous diffusion plant. A system that was developed and installed in the extended-range product withdrawal station of the Portsmouth facility measures enrichment with a relative accuracy of 0.5% (2σ). A stand-alone neutron detector has also been successfully evaluated for the measurement of the isotopic abundance of ^{234}U in UF_6 in sample cylinders, an application of potential importance to Minor Isotope Safeguards Technology (MIST). Based on this accumulation of data, recommendations are made on the role of NDA measurements for enrichment plant safeguards, including additional tests and evaluations that may be needed, particularly for advanced uranium enrichment processes.

I. INTRODUCTION

In the past decade considerable progress has been made in applying nondestructive assay (NDA) methods to measurement problems in uranium enrichment plants. The motivation for this work is primarily nuclear safeguards and the economic value of uranium. While the principal measurements for accounting and quality assurance at enrichment facilities are weighing, mass spectrometry, and chemical analysis, NDA methods are well suited for in-line verification and criticality control at product withdrawal stations, verification of UF_6 cylinder contents by inspectors, measurement of scrap and waste and decontamination solutions, and for some process control problems and hold-up evaluations.[1] The role of NDA instrumentation in perimeter safeguards systems for

enrichment plants under IAEA safeguards and in domestic facilities such as the projected centrifuge addition to the Portsmouth Plant is currently under close scrutiny.

The purpose of this report is fourfold: (1) to present results of NDA measurements performed in 1971-72 at the Oak Ridge Gaseous Diffusion Plant (ORGDP), focusing on measurements of hold-up and in-process materials not previously reported, (2) to review briefly in-line and inspection methods for verification of UF_6, (3) to present some concepts involving NDA for centrifuge plant safeguards, and (4) to provide recommendations on the feasibility of NDA measurements for enrichment plant safeguards, including additional tests and evaluations that are needed. These four topics are presented as major sections of this report.

II. MEASUREMENTS AT THE OAK RIDGE GASEOUS DIFFUSION PLANT

A. The MONAL Field Operation

For about six months beginning on October 1, 1971, the Mobil Nondestructive Assay Laboratory (MONAL) was stationed at ORGDP for the purpose of investigating the feasibility of NDA measurements of the enrichment and uranium content of typical materials in the gaseous diffusion plant and the Y-12 highly enriched uranium facility. The MONAL, shown in Fig. 1, was developed by the Los Alamos Scientific Laboratory (LASL) in the early phase of its safeguards program to test and demonstrate NDA techniques at a variety of nuclear facilities in the U. S., with emphasis on uranium and plutonium scrap and waste materials.[2,3] This 3-m by 15-m customized trailer carried a wide range of nuclear assay equipment including a 14-MeV neutron generator and delayed neutron assay system, early models of the Random Driver interrogation unit[4,5] and the Ge(Li) Segmented Gamma Scanning system,[6,7] a thermal neutron coincidence counter for passive measurement of spontaneous fissions from ^{238}U and ^{240}Pu, and portable neutron and gamma-ray instrumentation for measuring holdup and surveying cylinders of UF_6.

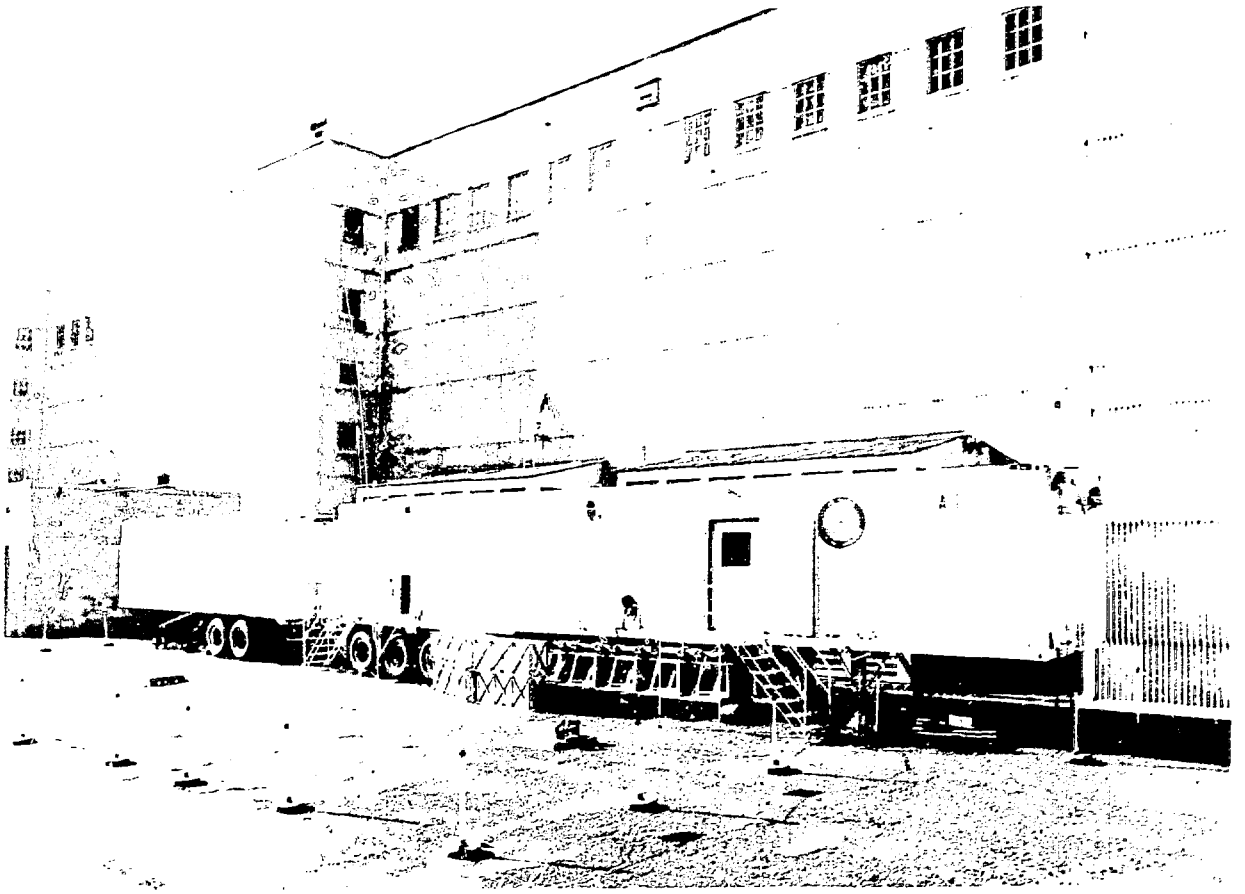

Fig. 1. The Mobil Nondestructive Assay Laboratory (MONAL) stationed at the Oak Ridge Gaseous Diffusion Plant in 1971.

Many of the results of the MONAL field operation at ORGDP have been reported by T. D. Reilly;[8] however, some details of measurements of hold-up and process line materials were not covered in Reilly's report. This last information is described below because it may be relevant to current safeguards planning, and for completeness much of the information given in the previous report is also included.

B. Holdup in Shut-Down Cascade

Measurements of residual holdup in the [illegible] cascade at ORGDP, K-25,* were made using portable [illegible] gamma-ray detectors. This cascade was shut down in [illegible], and during the period of the MONAL visit plans for dismantling [illegible] were being considered. The measurements were undertaken to [illegible] determine the feasibility of NDA instruments for hold-up measurements, 2. provide semiquantitative data for comparison with estimates of holdup based on decontamination of broken parts and laboratory studies of U deposition in this type of equipment, and [illegible] provide a pilot run for criticality safety surveys that might have been needed for dismantling operations. Measurements were attempted in buildings K-305 and K-306 near the top of the original cascade, where the ^{235}U enrichment was in the range 88 - 93%. The process equipment in K-306 had been "treated" to remove chemically active surface layers of contamination.

The plan view of the K-306 portion of the K-25 cascade is shown in Fig. 2. Except for the first and last units in the building, each unit has 14 cells and each cell contains six diffusion stages inside a steel housing. K-305 contains 10 units but otherwise has the

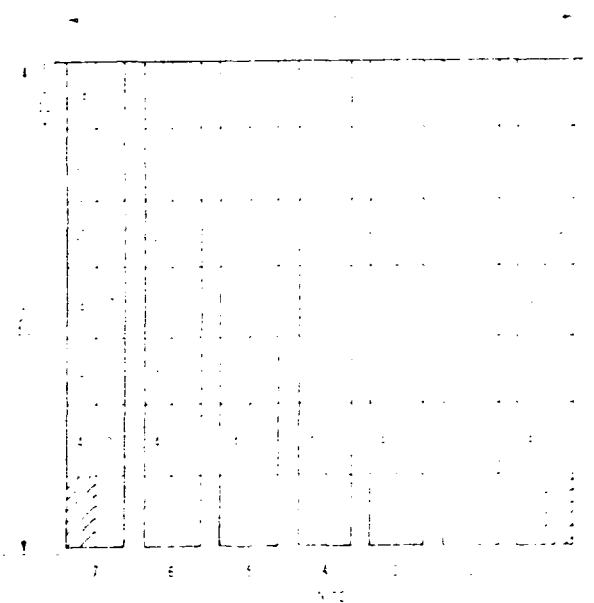

Fig. 2. Plan view of cascade layout in building K-306 of the [illegible] cascade. Each cell, corresponding to a numbered small [illegible], contains six stages of gaseous diffusion equipment.

*Gaseous diffusion building designations for the ORGDP, Portsmouth, and Paducah plants are "K," "X," and "C," respectively.

same layout. A stage comprises one gaseous diffusion converter, one centrifugal pump for the "A" (enriched) stream and one for the "B" (depleted) stream, two pump seals, and many interconnecting pipes. A schematic of the cross section of a cell, approximately to the scale of a 306 cell, is shown in Fig. 3. The approximate dimensions of converters in K-305 and K-306 are 1.2-m diam by 2.0-m long and 1.0-m diam by 1.3-m long, respectively.

The bulk of the measurements were made on seals, pumps, and converters because a preliminary survey showed the larger concentrations of uranium to be in these components. The survey also revealed that gamma-ray assay using portable NaI detectors was

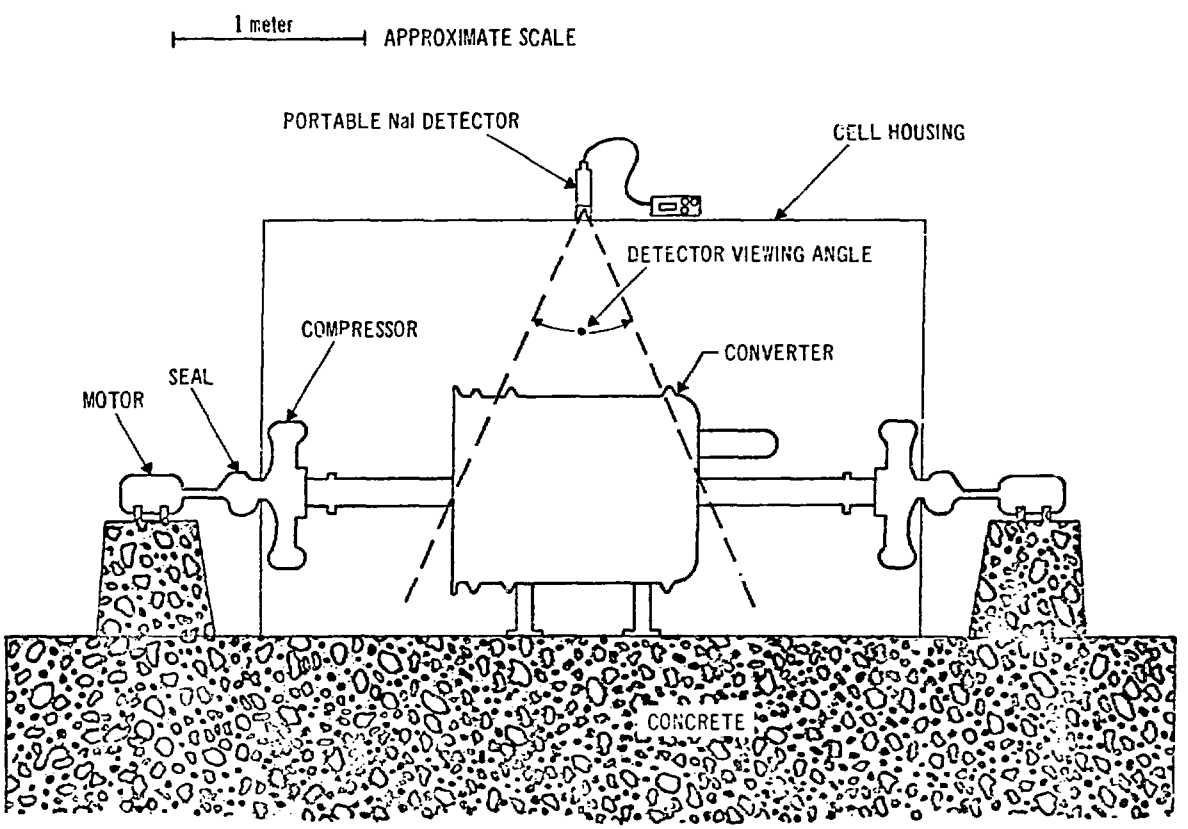

Fig. 3. Schematic cross section of a cell in Building K-306. The detector position used for measuring holdup in converters is also shown.

feasible for pumps and converters but not for the seals. The metal housings for the seals are much too thick for reliable gamma-ray measurements; instead, the seal assays were based on measurements of neutrons from F(α,n) reactions in the residual material, presumably UO_2F_2. Because the dominant source of α activity is ^{234}U, a characteristic $^{235}U/^{234}U$ ratio had to be assumed to derive the ^{235}U assay. Both neutron and gamma-ray detection methods had been used at ORGDP and the Portsmouth Plant to search for UF_6 condensations in operating cascades.

Portable gamma-ray instrumentation similar to that used for the K-25 hold-up measurements is shown in Fig. 4. The detector

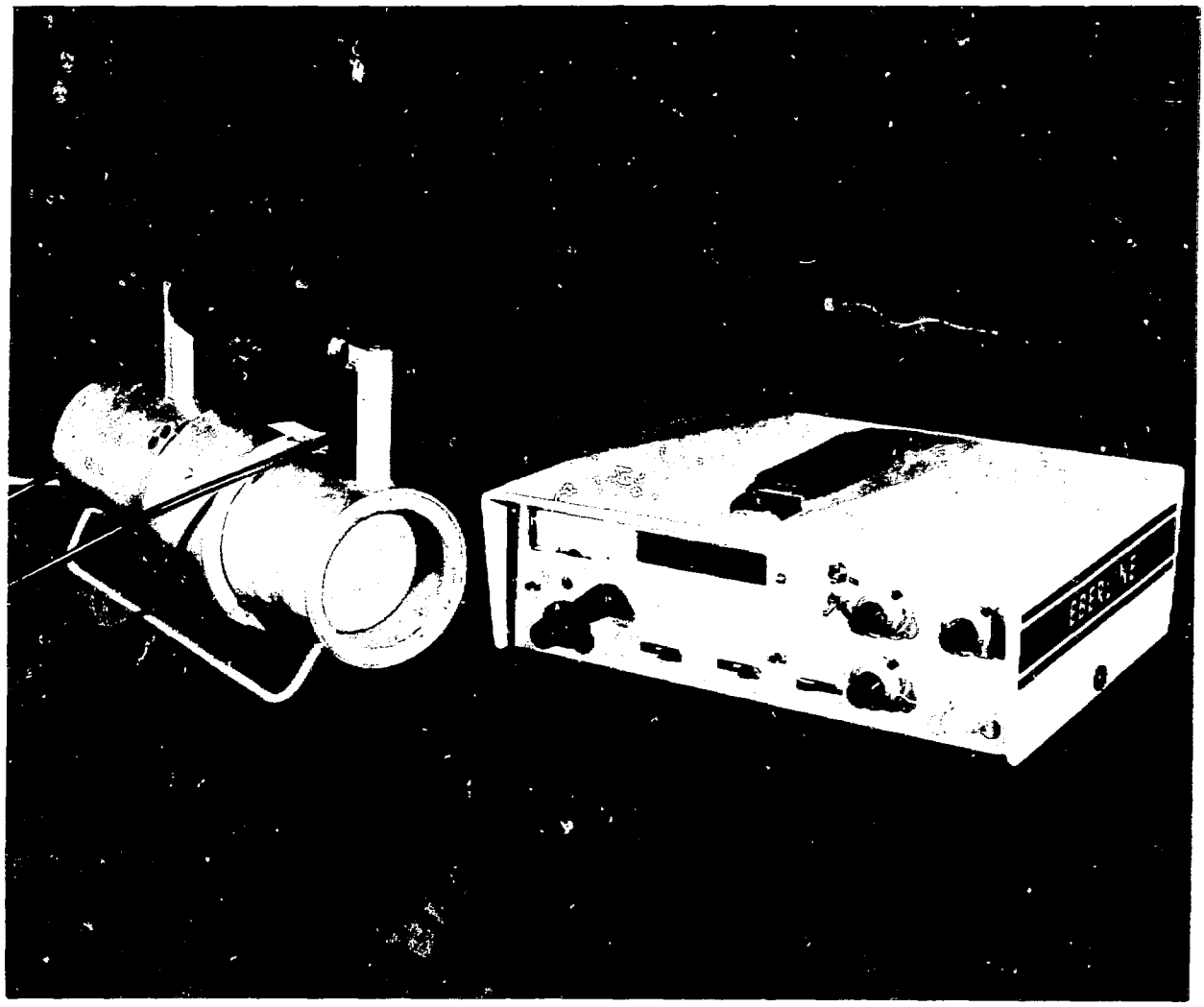

Fig. 4. Portable gamma-ray instrumentation similar to that used to measure ^{235}U holdup in the K-25 shut-down cascade at ORGDP.

consisted of a 5-cm-diam by 1.3-cm-thick NaI crystal with photomultiplier surrounded by a hand-held lead collimator-shield. The position of the detector within the collimator could be varied to select a viewing angle that would encompass a specific piece of equipment. The measurements of pumps and converters were performed with the detector placed on top of the cell housings, as shown in Fig. 3. A cart-mounted system, shown in Fig. 5, was also used for floor-level surveys for hot spots. The cart-mounted detector and electronics were contained in freon evaporative coolers to enable them to be used in the operating cascade, where the temperatures inside cell housings exceed 45°C.

The gamma-ray detector used for the measurements of pumps and converters was calibrated using a thin foil of highly enriched uranium metal having a mass of 0.695 g ^{235}U. The net counting rate for a 50-keV-wide window bracketing the 186-keV peak was approximately 92 cpm for 1 g ^{235}U at a distance of 2 m from the detector, and the background rate was 30 cpm. A combination of transmission measurements and calculations was used to correct the data for gamma-ray absorption. Effective thicknesses of the equipment were obtained by

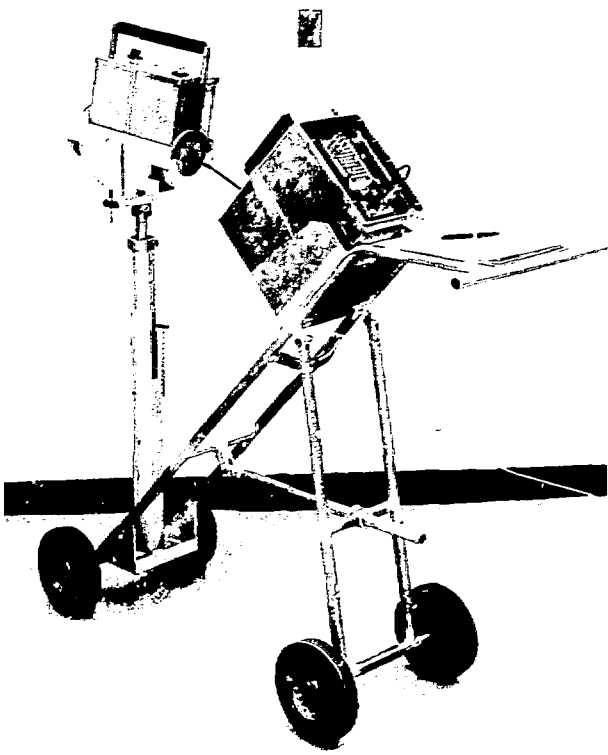

Fig. 5. Cart-mounted gamma-ray assay system for holdup and UF_6 gas phase measurements. The detector and electronics are contained in freon evaporative coolers to allow measurements inside operating cells.

measuring the transmission of 662-keV gamma rays from a ^{137}Cs source placed at various points inside a set of pumps and converter that had been removed from the cascade and partially dismantled. Design details of the pumps were also used in the calculations of 186-keV gamma-ray absorption. The transmission studies and Monte Carlo calculations gave the somewhat surprising result that the gamma-ray absorption correction factors based on the detection of uncollided 186-keV photons had to be reduced by about 70% to account for photons that were Compton-scattered by the matrix and detected in the 50-keV energy window.

The neutron measurements were performed with a hand-held neutron detector attached to a portable electronics unit. The detector consisted of a cadmium-clad assembly of two 2.5-cm-diam by 50-cm-long, 4-atmosphere ^{3}He counters wrapped with 2.5-cm layers of polyethylene. Cylinders of UF_6 with known weights and isotopic compositions, together with the neutron yields from uranium isotopes in UF_6 recommended by Sampson,[9] were used to calibrate this detector. A ^{252}Cf source with a strength of 8.8×10^3 n/s was used as a secondary standard during the measurement campaign. Figure 6 shows the detector response as a function of distance, as measured with the ^{252}Cf source. The distances from the seal axes to the detector were 15 cm and 30 cm for measurements in K-306 and K-305, respectively. Counting rates of 0.25 cpm/g ^{235}U and 0.113 cpm/g ^{235}U for K-306 and K-305 seals, respectively, were obtained assuming a $^{235}U/^{234}U$ ratio of 165 and the chemical composition UO_2F_2 for this material. The respective background rates for buildings 306 and 305 were approximately 30 cpm and 80 cpm. The (α,n) neutron yield from UO_2F_2 is approximately one-third that of UF_6.

These pilot measurements were made in Unit 1, Building 306, denoted by K-306-1, and Unit 7, Building 305, denoted by K-305-7. Table I is a summary of the number of pieces of equipment measured in each of the buildings and the percentage of the total equipment represented by this sample. The project goal was to measure as many pieces of equipment as possible with three persons working

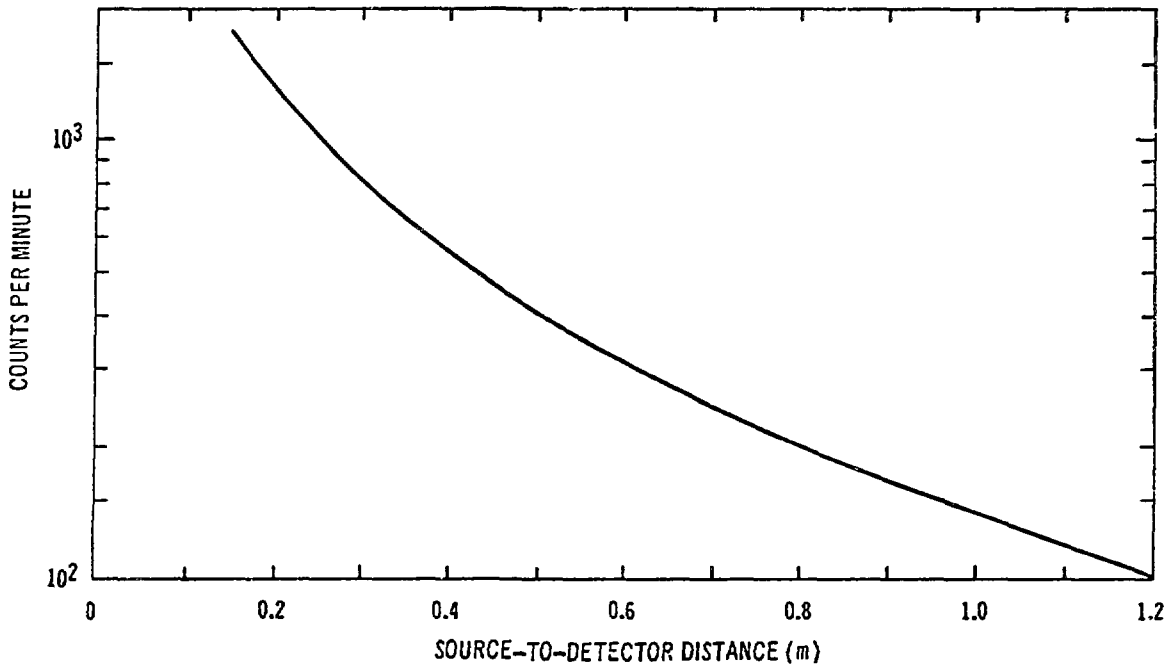

Fig. 6. Counting rate of hand-held polyethylene-moderated ^3He neutron detector as a function of distance from a ^{252}Cf source (8.8×10^3 n/s).

TABLE I

SUMMARY OF NUMBER OF COMPONENTS AND MEASUREMENTS IN K-306 AND K-305

K-306

	Seals	Pumps	Converters
Total components	1152	1152	576
Components measured	114	156	78
% of total measured	10	14	14

K-305

Total components	1200	1200	600
Components measured	108	36	18
% of Total measured	9	3	3

for one week so that estimates of the inventories of these buildings could be made. Consequently, sacrifices in the statistical precision of individual measurements were made. The counting times for individual neutron and gamma-ray measurements were 60 s and 12 s, respectively, with longer background counts interspersed frequently. The sensitivities of these instruments for measuring the ^{235}U content of the different types of equipment are given in Table II.

Results of the measurements of individual components in 306-1 are presented in Table III, including the 1-σ statistical uncertainties. It was essential to tally negative numbers, which reflect poor counting statistics, to obtain statistically correct total inventories. These data show that the amount of material in the K-306 converters is generally negligible relative to that in the pumps and seals, with the exception of five converters that contain a total of 226 g ^{235}U. The "treatment," or flushing of the K-306 converters was therefore very effective, and the exceptions probably contain deposits of relatively stable chemicals, such as UO_2F_2, that formed during the operation of the cascade

TABLE II

^{235}U DETECTION SENSITIVITIES[a]

IN THE SHUT-DOWN CASCADE

(g ^{235}U)

Building	Seals[b]	Pumps[c]	Converters[c]
K-306	65	15	5
K-305	235	46	8

[a]Sensitivity: ^{235}U equivalent of three standard deviations (3σ) of the background.

[b]Neutron measurement time: one minute. The source-to-detector distance is about 15 cm in K-306 and 30 cm in K-305.

[c]Gamma measurement time: 12 s. The source-to-detector distance is about 2.2 m.

and prior to the cleaning treatment. The K-305 converters have significantly more residual U than the K-306 converters but also have relatively much less than their seals and pumps. The largest deposit, 1250 g ^{235}U, was found in a seal in K-305.

TABLE III
MEASURED ^{235}U HOLDUP IN K-306-1
(Quantities and Uncertainties in g ^{235}U)

Cell-stage	"A" Seal	±1σ	"A" Pump	±σ	Converter	±σ	"B" Pump	σ	"B" Seal	σ
2-1	42	24	24	8	6	2	26	8		
2-2			19	7	9	2	17	7	21	24
2-3	18	24	17	7	0	1	24	8		
2-4			-10	3	4	2	10	6	42	24
2-5	9	21	17	7	2	2	28	8		
2-6			24	8	1	1	10	6	21	24
3-1	48	24	31	9	-1	1	13	7	12	21
3-2	9	21	1	5	-1	1	21	8	96	29
3-3	30	24	-4	8	-1	1	22	8	42	24
3-4	0	21	8	6	0	1	13	7	60	29
3-5	27	24	10	6	0	1	12	7	60	24
3-6	51	24	134	16	1	1	10	6	0	21
4-1	30	24	12	7	1	1	12	7		
4-2			5	6	0	1	21	8	21	24
4-3	21	24	-6	4	-1	1	15	7		
4-4			1	5	1	1	6	6	48	24
4-5	42	24	-2	5	-1	1	40	10		
4-6			21	8	1	1	28	8	60	29
5-1	-3	21	19	7	1	2	-4	4	60	24
5-2	63	24	-2	5	5	2	28	8	18	24
5-3	12	24	24	8	28	4	49	10	105	30
5-4	48	24	6	6	14	3	68	12	126	30
5-5	21	24	10	6	-1	1	21	8	87	28
5-6	0	21	12	7	0	1	5	6	123	30
6-1	12	24	12	7	3	2	19	7		
6-2			-4	4	0	1	22	8	108	30
6-3	57	18	3	5	-1	1	22	8		
6-4			8	6	-1	1	15	7	18	24
6-5	9	21	17	7	1	1	49	10		
6-6			24	8	1	1	13	7	18	24
7-1	-36	21	10	6	-1	1	12	7	63	24
7-2	36	24	8	6	1	2	40	10	-12	24
7-3	-30	21	24	8	-2	1	15	7	30	24
7-4	-3	21	24	8	28	4	10	6	12	24
7-5	-3	24	199	19	115	8	8	6	63	24
7-6	87	24	6	6	-2	1	44	10	177	33
8-1	-12	21	10	6	2	2	5	6		
8-2			19	7	-3	1	40	10	39	24

TABLE III (continued)

Cell-stage	"A" Seal	±1σ	"A" Pump	±σ	Converter	±σ	"B" Pump	σ	"B" Seal	σ
8-3	66	30	19	7	-1	1	22	8		
8-4			6	6	-1	1	12	7	36	24
8-5	18	24	19	7	0	1	28	8		
8-6			31	9	1	1	10	6	-39	24
9-1	39	30	26	8	-1	1	17	7	90	30
9-2	105	30	2	8	-1	1	31	9	60	29
9-3	18	24	15	7	-2	1	52	11	63	24
9-4	39	24	5	6	2	2	12	7	36	29
9-5	-30	21	42	10	-2	1	36	8	51	24
9-6	30	24	13	7	-1	1	6	6	171	33
10-1	0	21	6	6	2	2	6	6		
10-2			-6	4	2	2	29	9	12	24
10-3	27	24	3	5	-2	1	29	9		
10-4			6	6	-2	1	15	7	96	30
10-5	48	24	-4	4	-1	1	1	5		
10-6		28	12	7	1	1	19	7	57	29
11-1	21		17	7	-1	1	12	7	36	24
11-2	63	24	-2	5	1	1	17	7	96	30
11-3	51	29	26	8	0	1	29	9	60	24
11-4	9	21	8	6	1	2	21	8	18	24
11-5	21	29	22	8	0	1	51	11	42	24
11-6	9	21	3	5	0	1	22	8	48	29
12-1	-3	21	17	7	-1	1	6	6		
12-2			12	7	0	1	21	8	21	24
12-3	39	24	1	5	-2	1	12	7		
12-4			26	8	-2	1	15	7	42	24
12-5	66	29	1	5	0	1	29	9		
12-6			15	7	-2	1	5	6	42	24
13-1	-3	24	28	8	4	2	3	5	3	21
13-2	69	29	12	7	1	1	47	10	-18	24
13-3	12	24	8	6	-4	1	38	9	3	21
13-4	30	24	15	7	0	1	15	7	18	24
13-5	-36	21	29	9	1	1	51	11	42	24
13-6	51	24	13	7	5	2	6	6	-21	21
14-1	21	24	22	8	0	1	13	7		
14-2			12	7	41	5	28	8	18	24
14-3	21	24	1	5	0	1	47	10		
14-4			5	6	-1	1	31	9	36	24
14-5	66	29	22	8	-2	1	24	8		
14-6			36	9	-1	1	15	7	99	30

The average ^{235}U content per component in K-306-1 and K-305-7 based on these measurements is presented in Table IV. These averages and the total number of components given in Table I were

TABLE IV

AVERAGE ^{235}U (g) CONTENT PER COMPONENT

IN K-306-1 AND K-305-7

	Seal	Pump	Converter	Stage
K-306-1	35	19	3.1	111
K-305-7	205	128	20	686

used to derive estimates of the total ^{235}U inventories for these two buildings, presented in Table V. The estimated error of ± 50% in the total inventory is dominated by uncertainties in gamma-ray attentuation corrections and the $^{235}U/^{234}U$ ratio used to interpret the neutron data. The agreement between the present estimate of 417 kg of ^{235}U in the pumps, seals, and converters in K-305 is in good agreement with the ORGDP estimate of 509 kg, considering the NDA measurement uncertainties and the small sampling. Because these materials are spread over the inner surfaces of thousands of pieces of equipment, they would not constitute a practical target for diversion. Furthermore, while the K-25 measurements proved the feasibility of using NDA to verify holdup in those buildings and equipment, these conclusions do not translate directly to the problem of measuring holdup in low-enriched gaseous diffusion equipment because of the much lower enrichment of the material and the larger sizes of the components. It would be much more difficult to obtain quantitative assay results for the latter type of equipment.

TABLE V

^{235}U (kg) CONTENT OF K-306 AND K-305

	Seals	Pumps	Converters	Total
K-306	40	22	1.8	64
K-305	246	154	12.	412
Total	286	176	13.8	476 ± 240

It was also requested that we estimate the amount of normal U in six sublimer-condenser tanks that had been removed from the "feed manufacturing plant" and stored between K-305-8 and K-305-9. These tanks are about 1-m diam by 4.5-m long and contain heat exchanger elements for condensing gaseous UF_6 from process lines. Twelve 1-minute measurements were made with the hand-held neutron detector at representative points on each cylinder. These data were averaged and a material composition of UF_6 was assumed to obtain the total amount of uranium in each tank. These assays, together with axial profiles for each tank, are presented in Fig. 7. Recent discussions indicate that this use of neutron detectors for monitoring sublimer-condensers that are being installed under the Cascade Improvement Program (CIP) may be competitive with load cells, which are now being considered.[10]

C. Measurements in the Operating Cascade

A portable NaI gamma assay unit was set up with a strip chart recorder to demonstrate the feasibility of dynamic monitoring of UF_6 retention in NaF traps. Several of these traps are used in series between the top of the cascade and the effluent stack to filter UF_6 from the gaseous effluent of the ORGDP cascades. NaF is used since it adsorbs UF_6 at one temperature and releases it later under heating. The traps, which are wrapped with heating elements and insulation, are regularly desorbed and rotated. The

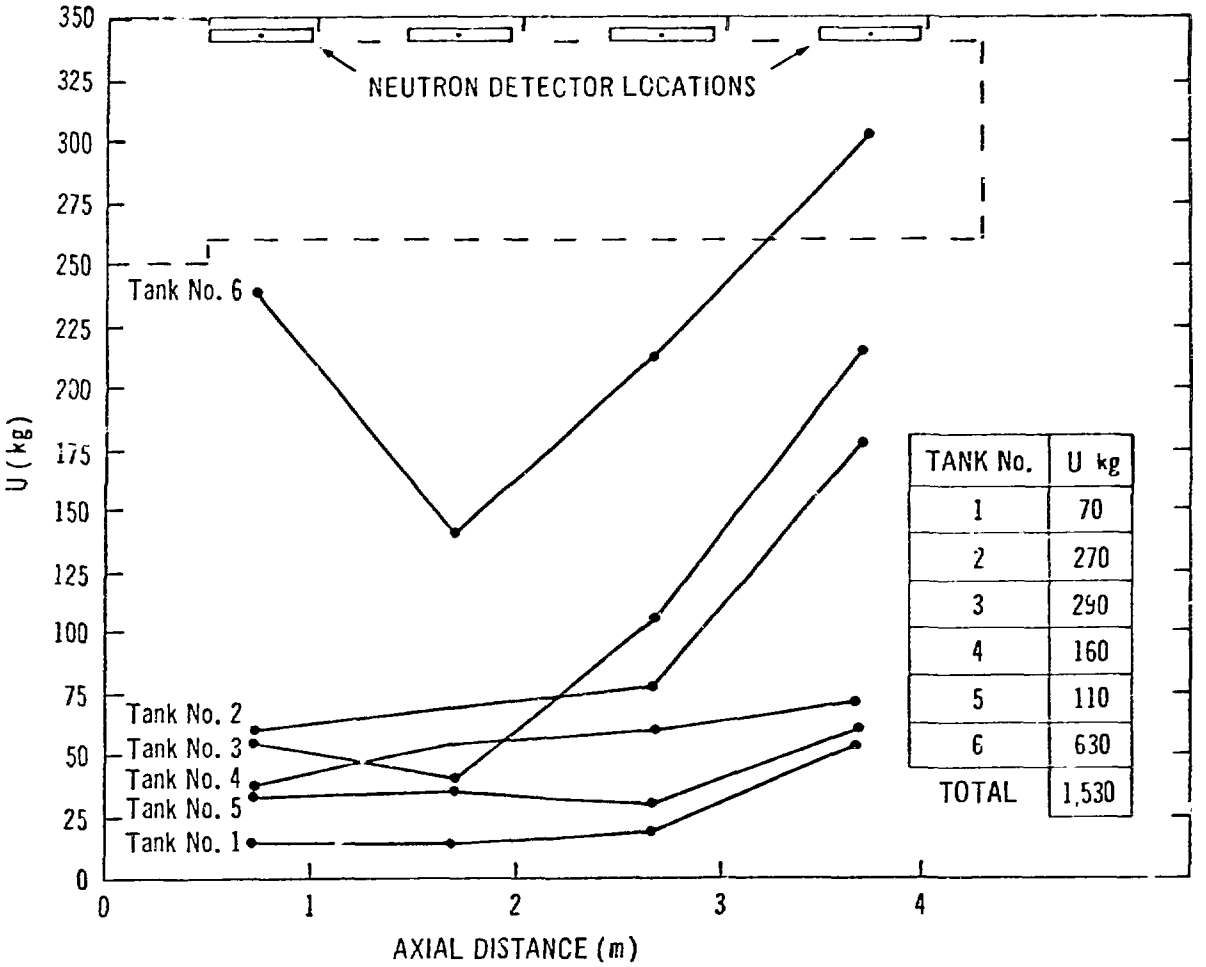

Fig. 7. Axial profiles of neutron measurements of U in sublimer-condenser tanks. The physical profile of a tank and the measurement positions are shown in the upper inset, and the total uranium assays for six sublimer-condensers are given in the lower inset.

detector was set to measure the 186-keV activity as a function of time in one of these traps. Figure 8 is a graph of the strip chart data taken over a two-day period. There was no apparent uranium retention during the period it was operated as the second trap from the input; however, the signal showed a steady rise

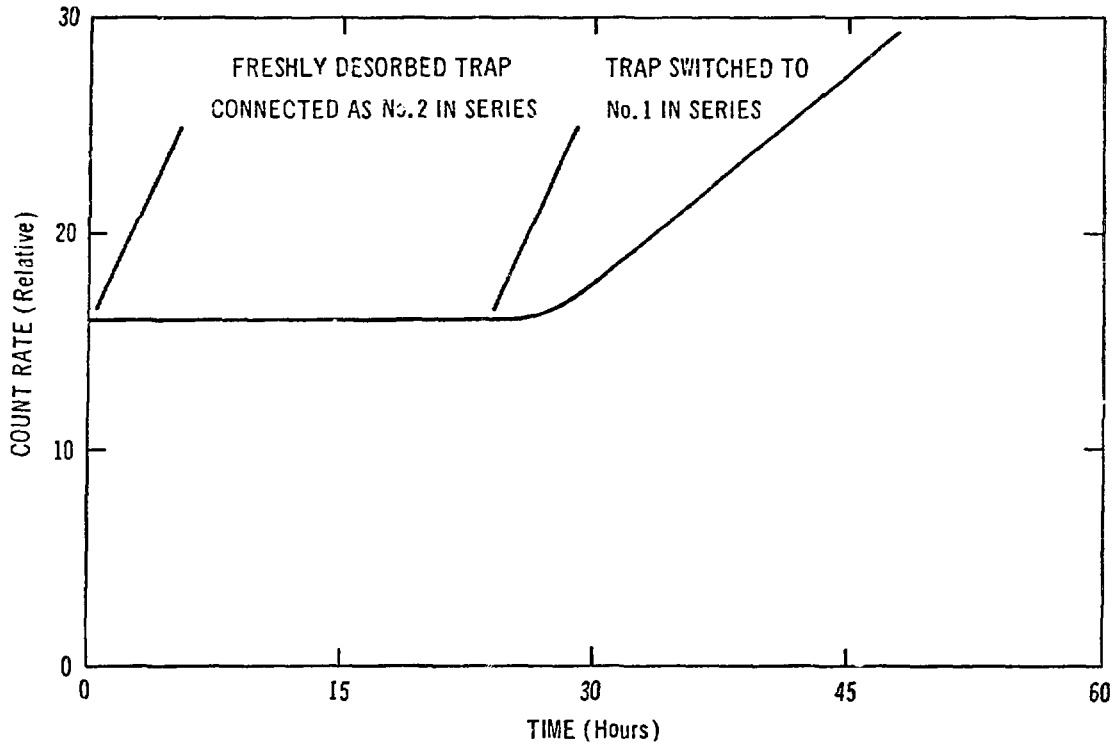

Fig. 8. Dynamic monitoring of a NaF trap for UF_6 at the gaseous effluent stack of the light element cascade ("Ponderosa Pines") of ORGDP. A portable NaI detector was used to monitor the UF_6 absorption in one of the traps.

after it was switched to the first position in the series of traps. The total rise observed in the second 24-hour period was about 2 mg U/cm^3, assuming an enrichment of about 4%. The sensitivity of the measurement was approximately 0.2 mg/cm^3.

The same instrument was used to measure vertical profiles of this trap after it had been operated for 24 hours as the first in the series of traps. These measurements were made with a 50-keV window bracketing 186 keV, called channel "A," and a 50-keV window centered at 250 keV, channel "B." This last channel provides a measure of aged deposits of U because it detects a portion of the spectrum of high-energy gamma rays from ^{234m}Pa, a nuclide in the

^{238}U decay chain that is in secular equilibrium with ^{234}Th (24.1 d half-life). The daughters of ^{238}U are nonvolatile and therefore cannot independently reach the traps via the gas stream. Channel "A" is also sensitive to freshly deposited uranium. Figure 9 shows the measured vertical profiles. The difference in the shapes of data for "A" and "B" is indicative of the freshly absorbed U. Results of similar measurements made on a different trap that was freshly desorbed are shown in Fig. 10. The profiles for channels "A" and "B" closely correspond, indicating permanently retained uranium deposits. Results obtained with this instrument showed it could be used as a continuous monitor to check trap operation and to indicate when to switch or desorb a given trap; to provide a rapid warning of cascade system failure; to provide a measure of the uranium removal as a trap is recharged; and to indicate when to change trapping media.

Alumina traps are used within the cascade to remove uranium from gaseous effluents. In contrast to NaF, alumina cannot be recycled. Vertical profile data for two alumina traps, one partially expended and the other totally expended, are shown in Figs. 11 and 12, respectively. The data were taken with the portable NaI unit, with a 30-s count for each point. Such measurements can reliably determine when alumina traps should be changed.

Measurements were also made to test the feasibility of using portable instrumentation to detect and assay material holdup in the operating cascade. The instruments used included a small NaI detector similar to that used for the trap measurements, the ^{3}He neutron detector described earlier, and a collimated 12.7-cm by 12.7-cm NaI crystal coupled to a portable electronics unit. This last device was called the "Elephant Gun," and is pictured in Fig. 13. It was used to measure 767- and 1001-keV gamma rays from the ^{238}U daughter ^{234m}Pa. (Subsequent to the MONAL operations at ORGDP, this device was used for the assay of low-enriched uranium waste.)[11] As a test, ORGDP personnel hid nine 12A UF_6 cylinders, containing from 68 to 205 kg of UF_6 within a portion of the operating cascade. The large NaI detection system was pushed

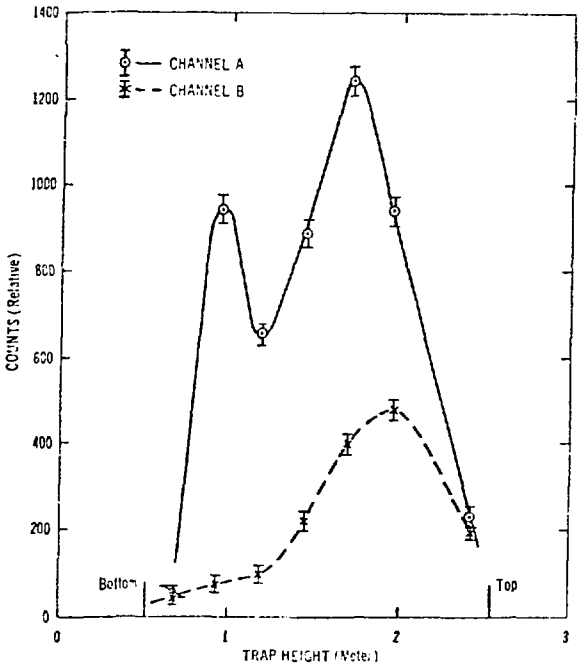

Fig. 9. Vertical profiles of gamma rays from a NaF trap after a 24-hour operation as first in the series of traps at the gaseous effluent stack of ORGDP. Channel "A" includes 186-keV gamma rays from both freshly absorbed U and permanently fixed deposits. Channel "B," a higher energy detection window, detects only the permanently fixed, well-aged deposits.

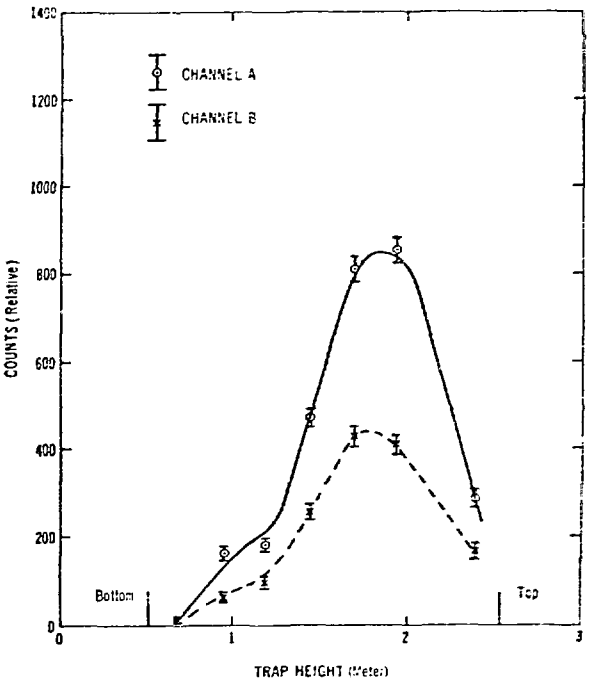

Fig. 10. Vertical profiles of gamma rays from a freshly desorbed (recycled) NaF trap that is one of a series of traps at the gaseous effluent stack of ORGDP. Channel "A" includes 186-keV gamma rays from ^{235}U and Channel "B" measures higher energy gamma rays from ^{238}U daughters.

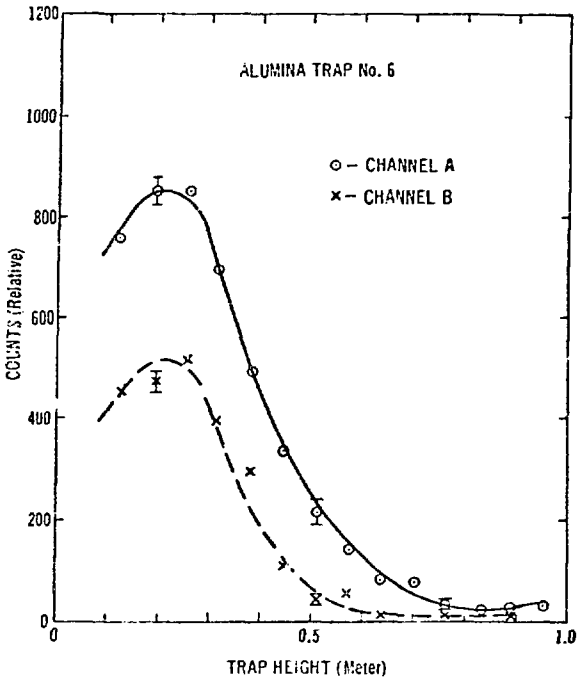

Fig. 11. Vertical profiles of gamma rays from an alumina trap that was partially expended. Channel "A" includes 186-keV gamma rays and channel "B" is a higher energy counting window.

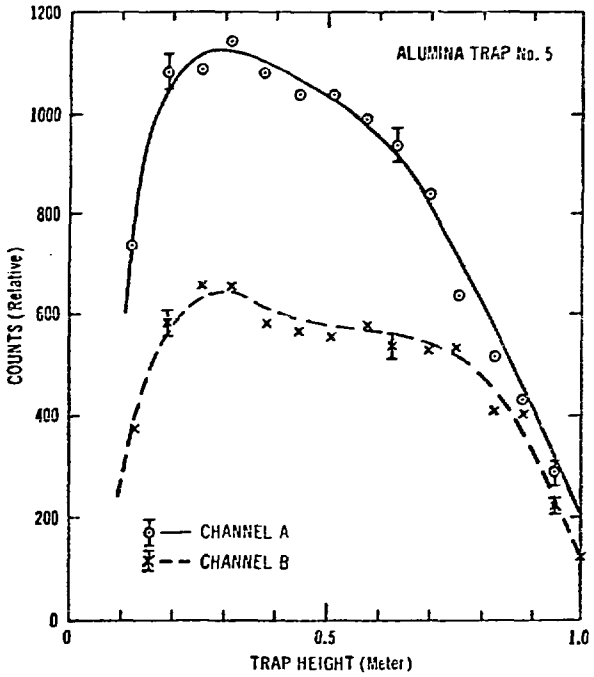

Fig. 12. Vertical profiles of gamma rays from an alumina trap that was almost fully expended. Channel "A" includes 186-keV gamma rays and channel "B" is a higher energy counting window.

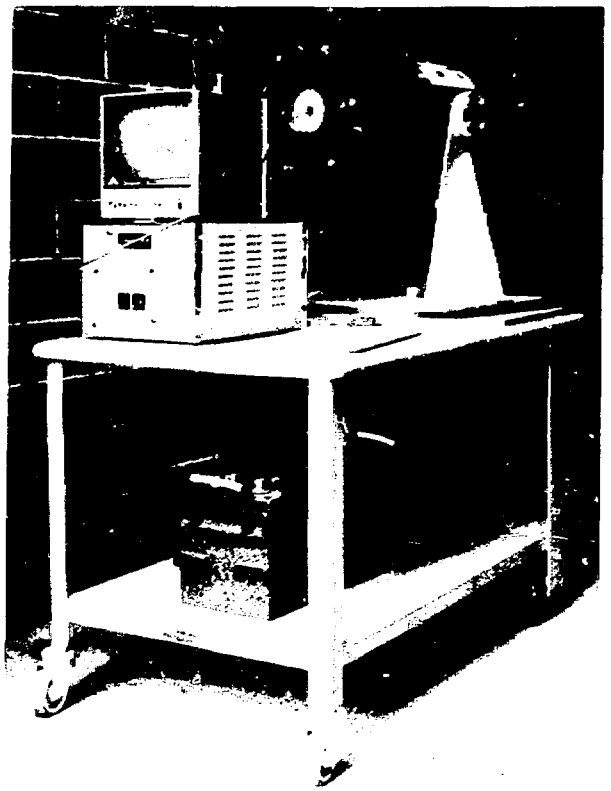

Fig. 13. A large cart-mounted, collimated NaI detector used to measure in-process UF_6 gas and ^{238}U deposits in the K-33 operating cascade of ORGDP. High-energy gamma rays from ^{234m}Pa daughters of ^{238}U were detected.

around the plant looking for gamma rays from the ^{238}U daughters; the majority of the cylinders were located in a one-hour hunt. The device could unambiguously detect a 200-kg cylinder at a distance of 40 m with a 30-s count. For finding condensations in the UF_6 gas pipes, the hand-held neutron and gamma-ray detectors were as effective and more practical for use by operating personnel.

The large NaI detector was also used to look for plating or holdup in the main gas pipes. The viewing cone of the detector corresponded to a 1.5-m-diam circle at 2.4 m. At this distance, it could detect 166 g ^{238}U in a 30-s count, assuming the ^{238}U to be well aged, i.e., the ^{234m}Pa daughters to be in secular equilibrium. Figure 14 shows data collected along a main gas transfer corridor in the K-33 cascade, viewing two large pipes. At the time of these measurements, the top enrichment of this cascade was about 3%. Except for large, real deposits in the evacuation booster station valves, the "average" amount of ^{238}U in the two pipes was $\sim$ 240 g/m. This was the measure of gas phase inventory, appropriately time-averaged by the 24.1-d half-life of ^{234}Th over recent operating history. Real holdup in the pipes, i.e., U plated on the inner

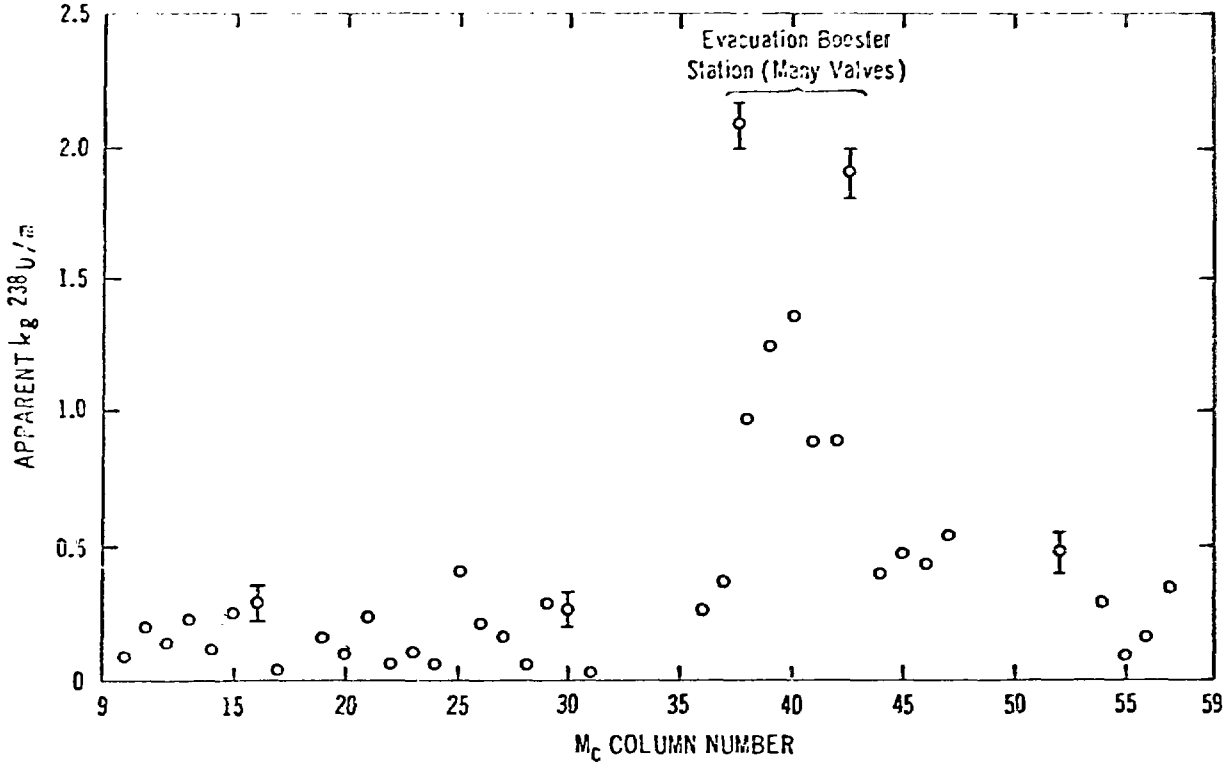

Fig. 14. ^{238}U content of two pipes along a main gas transfer corridor ("5th Avenue") of the K-33 cascade. The amount of ^{238}U was derived from measurements of ^{234m}Pa daughters of ^{238}U. Except for real U deposits in the valves, the results provide a measure of time-averaged gas phase inventory.

surfaces of the pipes, was very much smaller, as was later verified by measurements of similar pipes in a portion of K-33 that had been shut down for several months.

The feasibility of performing quantitative measurements of ^{235}U in the gas phase with small NaI instruments, such as shown in Fig. 5, had been demonstrated previously at the Portsmouth plant,[12] so similar measurements were not attempted by LASL in K-33. Later, J. C. Baily made extensive studies of the feasibility of measuring the gas phase in a diffusion plant using NaI to detect ^{235}U gamma rays.[13]

The feasibility of using a small hand-held NaI detector to find localized deposits in the large converters of K-33 was investigated in a cell that had been shut down. Some of the converters were thought to contain deposits as a result of in-leakage of air. Data taken for 30 s at selected points on the converters in this cell are indicated by the numbers in the circles shown in Fig. 15. "A" and "B" refer to the enriched and depleted flow paths, respectively. Clearly, some of these converters contained appreciable deposits, but quantitative measurements were not attempted because of time constraints on this study.

A survey of areas inside and near the K-33 plant was conducted with the portable neutron detector described above to determine whether this technique could provide a qualitative measure of the operating status of the cascade. An increase in the neutron field relative to background should be proportional to the quantity of ^{234}U in the UF_6 gas phase in the vicinity of the measurement. The results are presented in Table VI. Data taken on the second floor of K-33, where the enrichment equipment is located, show that the neutron counting rates, when corrected for a background rate of about 30 cpm, are approximately proportional to the number of operating units in the vicinity of the measurements. These results

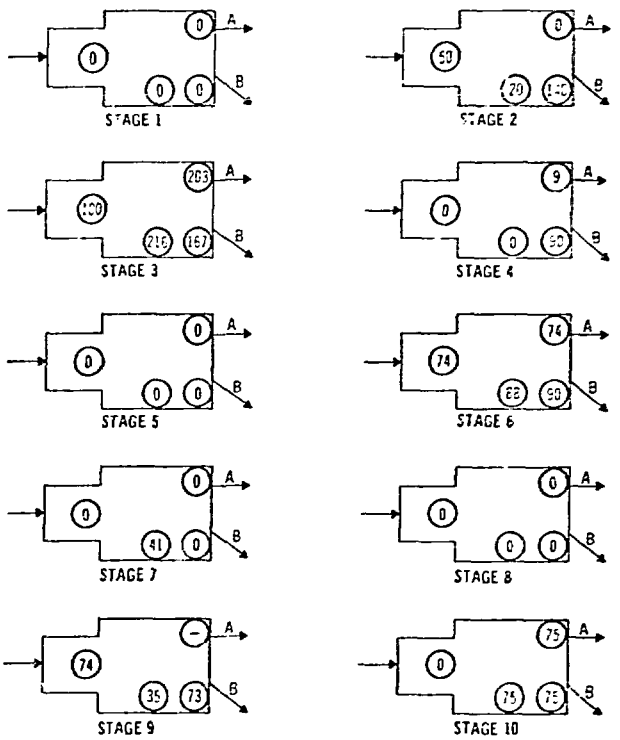

Fig. 15. Results of a survey in the converters in one inoperative cell in K-33. The numbers in the circles are a relative measure of the deposits or "plugs," "A" and "B" refer to enriched and depleted streams, respectively.

TABLE VI

NEUTRON SURVEY INSIDE AND NEAR THE K-33 OPERATING CASCADE

Measurement Location	Neutron Counting Rate (cpm)[a]
Outside K-33 in parking lot	29
First floor, K-33, in operations office	30
Second floor, K-33, Pillar M_b26, no units running	47
Second floor, K-33, Pillar M_b41, 1 unit running	73
Second floor, K-33, Pillar M_b58, 4 units running	153

[a]All counts taken for 5 minutes.

have important implications on the feasibility of using neutron detectors inside a centrifuge plant to monitor for enrichments exceeding declared limits, as elaborated later in this report.

D. Scrap and Waste Measurements

Passive neutron and gamma-ray assay methods and 14-MeV neutron interrogation were used to measure the wide variety of ORGDP scrap and waste materials, which, together with a summary of assay results, are listed in Table VII. The types of containers for these materials are given in Table VIII. Measurement data for a wide variety of scrap and waste materials from Y-12 are not included in the present summary but have been described by Reilly.[8]

TABLE VII

K-25 SCRAP MEASUREMENT SUMMARY

Description	Number of Items	U(kg)	^{235}U(kg)
NaF trap media	79	200	8.4
Alumina trap media	34	37	1.2
Miscellaneous cylinders	238	524	8.1
Incinerator ash and impure oxide	21	65	6.0
Barrier	18	0.2	-
Compressor blades	4	0.7	-
Misc.-UNH,MgF_2, Fuel rods	9	17	0.7
Totals	403	844	23.7

TABLE VIII

K-25 CONTAINER TYPES

55-, 30-, 20-, 10-gallon drums
13-cm-diam x 61-cm-high trap residue cans
15-cm-diam x 38-cm-high incinerator ash cans
Miscellaneous cylinders: 20 different sizes
Miscellaneous containers

The miscellaneous cylinders and containers, which were stored in a small warehouse, were quite old, in poor physical condition, and poorly characterized because their condition precluded sampling and analytic techniques. The plant wished to clean out this building and dispose of the contents but required better information to proceed. Rapid survey measurements of these items were performed with a Ge(Li) spectrometer set up in the warehouse. These assays were based on measurements of 186-keV gamma rays from

^{235}U, 1001-keV gamma rays from ^{234m}Pa (^{238}U daughter), and 662-keV gamma rays from ^{137}Cs (present in many of the items). Corrections for gamma-ray absorption were obtained from both calculations and transmission measurements. With these survey results ORGDP personnel proceeded to clear this warehouse.

The incinerator ash generated by ORGDP operations contains low enriched U with a weight concentration generally less than 30%. The material is in 15-cm-diam by 38-cm-high cans. A study was made on 16 of these cans to compare the NDA with analytical assay (blending, sampling, and analysis). ^{238}U and ^{235}U contents of these cans were measured with a segmented gamma scanning system. The materials were well aged so that the ^{234m}Pa gamma rays provided a measure of the ^{238}U. In addition, the ^{238}U content was measured with a thermal-neutron coincidence detector that counted ^{238}U spontaneous fissions. The total amount of ^{235}U in these cans measured by gamma-ray assay agreed with the chemical analysis to within 0.5%, but the total uranium measured by gamma-ray assay was 5% lower than the analytic assay.[8] Table IX shows a comparison of ^{238}U measurements of 10 of the 16 cans that were also measured with the neutron coincidence counter. The agreement between the neutron coincidence assay and the gamma-ray assay is noteworthy because these detectors operate on grossly different principles and were calibrated with different standards. It was concluded that NDA is superior to sampling/analytic assay due to the difficulty in obtaining a representative sample for chemical assay.

Used barrier and compressor blades, contained in 20-, 30-, and 55-gal drums, were measured with the MONAL 14-MeV neutron interrogation/delayed neutron assay system. The barrier material had been processed to remove most of the residual uranium, and the compressor blades were usually washed before loading into drums. It is difficult if not impossible to obtain representative samples of these materials for chemical analysis. Representative NDA results obtained for these materials are presented in Table X. Based on this limited sampling, we concluded that the barrier

TABLE IX

INCINERATOR ASH: SAMPLING/ANALYTIC-NONDESTRUCTIVE ASSAY COMPARISON[8]

^{238}U (Grams)

ID #	Sampling/ Analytic	Gamma Ray Assay	Neutron Coincidence
8449	542 ± 3	509 ± 10	450 ± 54
8447	1485 ± 30	1461 ± 6	1550 ± 92
8450	995 ± 19	929 ± 13	870 ± 63
8453	934 ± 36	877 ± 9	890 ± 72
8451	925 ± 11	865 ± 23	860 ± 63
8452	2282 ± 21	2283 ± 42	2230 ± 81
8444	934 ± 7	930 ± 13	960 ± 13
8441	1361 ± 10	1327 ± 13	1340 ± 33
8442	343 ± 6	302 ± 1	280 ± 35
8443	616 ± 16	585 ± 11	560 ± 53
Totals	10,417 ± 60	10,068 ± 56	9,990 ± 200
Ave.Dev.(%)	2.2	2.3	

materials contained much less uranium than expected; the compressor blades contained much more than expected; and these materials are not amenable to representative sampling for chemical analysis.

In response to the interest of personnel at Oak Ridge in monitoring liquid effluents and holding ponds, information was provided on a continuous in-line sewer monitor developed and used at Lawrence Livermore Laboratory.[14] The system is based on the so-called FIDLER, a 12.7-cm-diam by 16-mm-thick NaI detector that measures L-x rays and low-energy gamma rays from radioactive high-Z elements. This device has sensitivities in the sub-MPC region for all major radioactive isotopes of interest; examples are given in Table XI. During the MONAL operation, a FIDLER device was demonstrated on water samples at Oak Ridge.

TABLE X

NEUTRON INTERROGATION RESULTS FOR USED BARRIER AND COMPRESSOR BLADES

Material	Size (gal)	Weight (kg)	Tag Value (ppm U)	NDA[a] (ppm U)	NDA[a] (g U)
Barrier	20	182	Unknown	0 ± 28	0 ± 5
"	"	"	"	"	"
"	"	"	"	"	"
"	"	"	260	"	"
"	"	"	80	"	"
"	"	"	225	"	"
"	"	"	230	"	"
"	"	"	230	"	"
"	"	"	200	"	"
"	"	"	180	"	"
"	55	457	910	430	194 ± 50
Comp. Blades (Unwashed)	∿ 30	92	71	2440	224 ± 50
Comp. Blades (Washed)	∿ 30	74	32	5670	417 ± 100

[a]The minimum level of uranium detectability is approximately 2-3 gU, and the uncertainty at the 5-gU level is approximately ± 25% (1σ).

TABLE XI
SENSITIVITIES FOR THE DETECTION OF SELECTED ISOTOPES IN WATER[8]

Isotope	MPC (Public Water) $\mu Ci/cm^3$	MPC (Public Water) $\mu g/cm^3$	Sensitivity[a] (MPC)
^{235}U	3×10^{-5}	14	0.02
^{238}U	4×10^{-5}	120	0.04
U (Natural)	2×10^{-5}	29	0.03
^{239}Pu	5×10^{-6}	8×10^{-5}	0.4
^{241}Am	4×10^{-6}	1×10^{-6}	0.04

[a]Sensitivity for a 200-s count at the 3σ level.

Subsequently, portable models were supplied for evaluation to the AEC-ORO and AEC-Atlantic Regulatory Offices.[15]

III. NDA MEASUREMENTS OF UF_6 CYLINDERS, PRODUCT, AND TAILS

A. Measurements of UF_6 Cylinders with Portable Instruments

Three techniques using portable instruments have been developed and tested for rapid nondestructive verification of UF_6 enrichment in cylinders: gamma-ray counting with NaI or germanium detectors combined with ultrasonic measurement of cylinder wall thickness,[16-18] passive neutron counting,[18] and neutron interrogation with thermal neutrons from a radioactive source.[18,19] The accuracies and limitations of these techniques were obtained by testing them on a large number of UF_6 cylinders, including type 30 (2½ ton), the standard shipping container for power reactor feed material, and type 5A, which is used for shipping highly enriched uranium. Most of these evaluations were conducted at ORGDP and at the Portsmouth plant. Photographs of typical portable

instruments are shown in Figs. 16 and 17. Since a complete report of this work has already been published[18] and the passive gamma-ray and neutron methods are used routinely by domestic and IAEA inspectors and plant personnel, only a summary of the methods and their accuracies will be given here.

Of the three methods investigated, passive-neutron counting is the fastest, simplest, and least expensive for verifying low-enriched UF_6. In UF_6 there are two sources of neutrons, $F(\alpha,n)$ reactions and ^{238}U spontaneous fissions. For enriched uranium, the dominant source is (α,n) reactions from the ^{234}U activity,

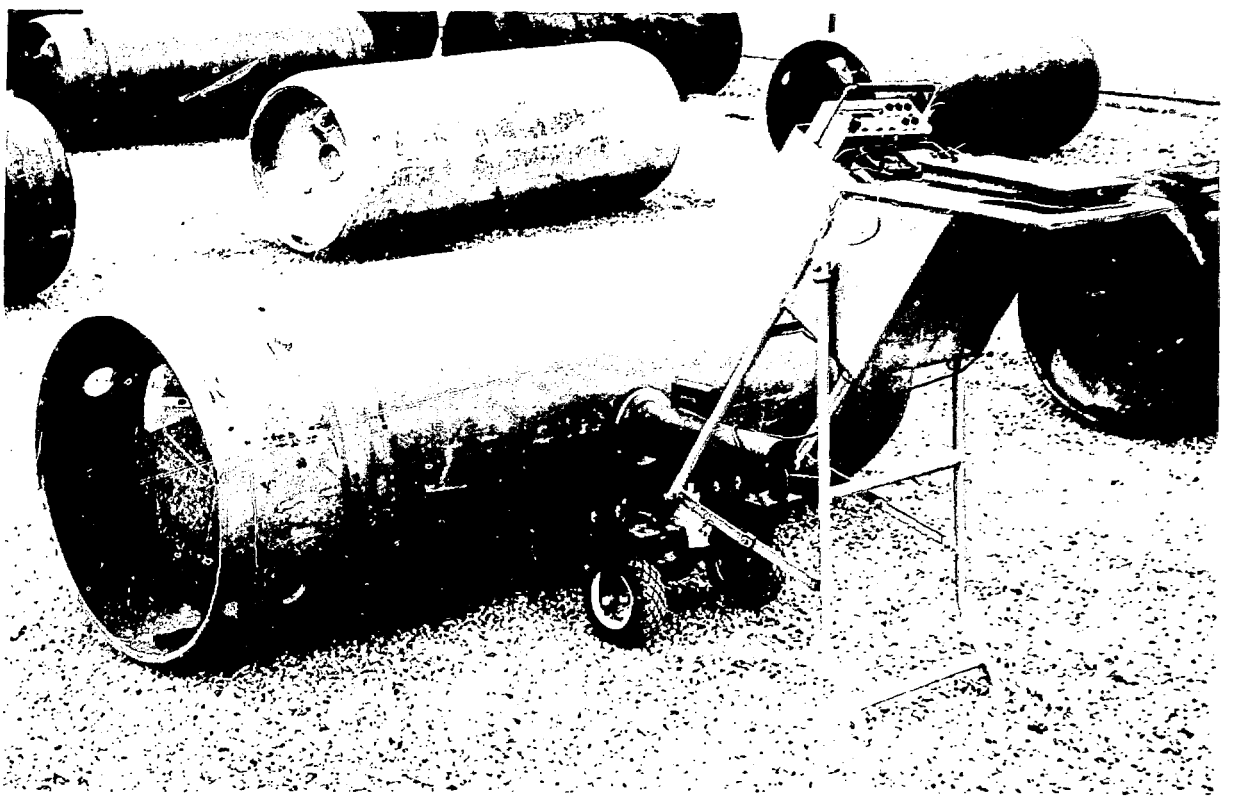

Fig. 16. Field unit for measurement of enrichment of UF_6 in 2-1/2-ton cylinders. The system consists of collimated NaI detector and an ultrasonic thickness gauge.

Fig. 17. Passive neutron assay of a 2-1/2-ton cylinder of UF_6.

and, for natural uranium, ^{238}U and ^{234}U contribute about equally to the neutron yield of UF_6.[9] Use of passive neutron counting for ^{235}U assay requires knowledge of the $^{235}U/^{234}U$ isotopic ratio, which may vary by as much as a factor of two over the complete range of enrichment but for low-enriched uranium (< 5%) is nearly constant. The standard deviation of passive-neutron assay results obtained at ORGDP and the Portsmouth plant for 110

type-30 cylinders (enrichments less than 4%) was 6%, confirming that the $^{235}U/^{234}U$ ratio was almost constant and that the method was generally free of serious systematic errors. If either the specific neutron yield of the UF_6 or the ^{234}U isotopic abundance could be included in the cylinder specification, passive neutron counting would provide a more accurate verification procedure. The specific neutron yield could be obtained at the originating product station by counting neutrons from either a sample of UF_6, as described later in this report, or the actual cylinder.

To better understand the passive-neutron assay method, neutron leakage spectra and fluxes were calculated with the Monte Carlo code, MCN.[20] The results showed that variations in the neutron leakage per source neutron caused by large changes in enrichment and cylinder loadings were surprisingly small.[18] Optimum detector positions were also obtained from these studies.[21]

The NaI gamma enrichment meter, together with the ultrasonic thickness gauge, can be used to measure the enrichment of 2½-ton cylinders to an accuracy of $\sim$ 5% (1σ); however, based on results obtained for 107 cylinders, this method fails (relative error > 16%) about 15% of the time. An outlier is easy to recognize because measurements at different points on the cylinder give significantly different results. Information on cylinder heels, cylinder fill dates, and gamma spectra, together with some calculations, indicate that outlier results are probably caused by the background radiation from ^{234}Th-^{234m}Pa deposited on the cylinder walls when the cylinder was emptied of its previous UF_6 fill. The shape of this background spectrum should differ from that of ^{234m}Pa uniformly distributed in the UF_6 and would therefore cause an error in the simple two-window analysis that was used in this study. Use of a high-resolution germanium detector should obviate this problem and greatly improve the reliability. For higher enrichments of UF_6, stored in 5A cylinders, no problems were encountered with the NaI system, and the standard deviation of results obtained for 19 cylinders was 2%. This reliability and

accuracy was expected for the higher enrichments because of the inherently larger signal relative to background. A few isolated problems have subsequently been found for U that has been irradiated in reactors.[10]

Active neutron interrogation, using thermal neutrons to generate fissions in UF_6 and ^4He counters to detect prompt-fission neutrons, is a promising method for measuring low-enrichment UF_6 but it still needs more development to reach a fully operational status. A schematic drawing of the system designed by Caldwell[19] is shown in Fig. 18. The response of the system varies almost linearly as a function of enrichment up to 2.5% and then effectively saturates at 4%. Analysis of the data showed that the response is sensitive to relative locations of the neutron source, the UF_6 cylinder, and the neutron detectors; consequently, these parameters may be used to optimize the technique for specific applications.[18]

These three basic techniques provide a capability for the verification of UF_6 product that is based completely on portable instrumentation and minimal calibration. Measurement accuracies can be improved with further development.

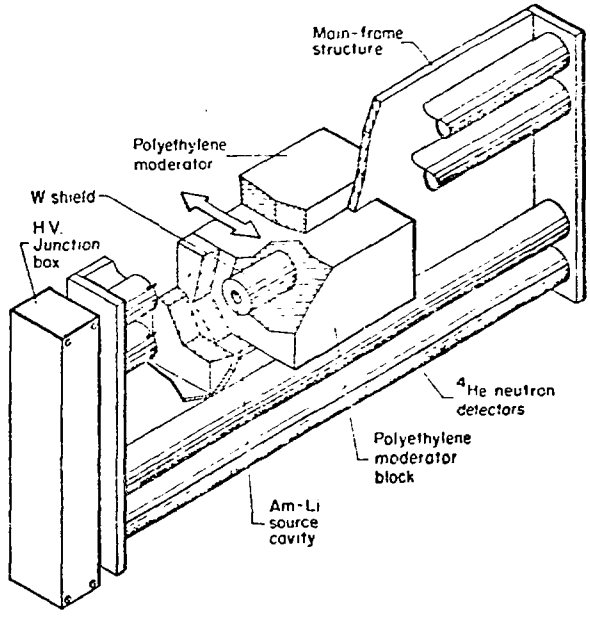

Fig. 18. Portable neutron interrogation unit.[19] Thermal neutrons from the source-moderator assembly produce fissions in UF_6 and the prompt fission neutrons are detected by the ^4He counters.

B. Product and Tails Measurements

In-Line Monitor--Passive-neutron and gamma-ray assay methods can be used to measure

continuously the enrichment of ^{235}U and ^{234}U in the UF_6 product and tails of a gaseous diffusion plant. The measurements are made on liquid UF_6 prior to withdrawal into product cylinders. In 1973 such a system was developed and installed in the extended-range product withdrawal station of the Portsmouth gaseous diffusion plant. The system is used for process enrichment control and criticality safety control to guarantee that highly enriched UF_6 is not withdrawn into a low-enrichment cylinder. Details of the design and performance of this system have been reported.[22,23]

Figure 19 shows an assembly drawing of the in-line enrichment monitor. The liquid UF_6 product stream was directed through the

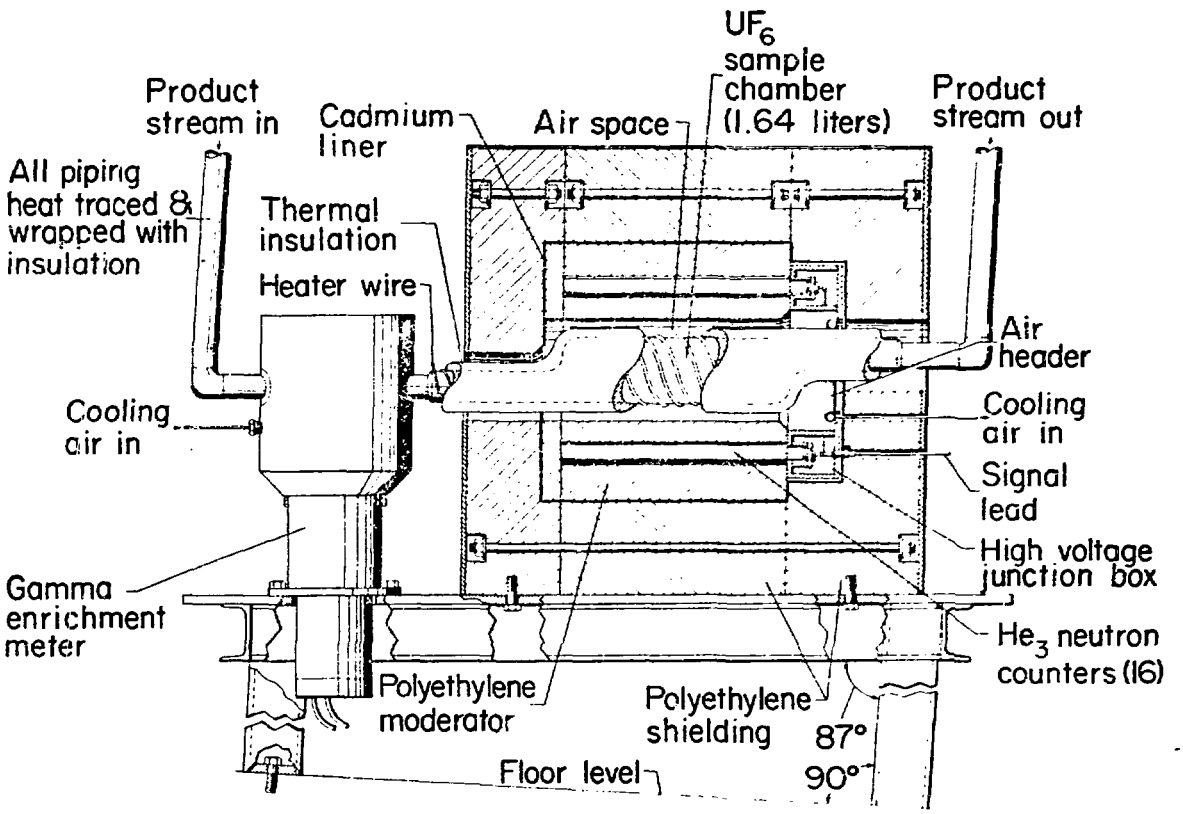

Fig. 19. UF_6 enrichment monitor--schematic diagram of gamma spectrometer and neutron detector with shield assemblies. The diameter of the neutron shield assembly is 61 cm.

assay device with a heated bypass line. Figure 20 shows the installation. An automatic data acquisition system, located in an adjacent room, continuously displays scaler counts, time-of-day, and computed enrichments, and also issues an alarm if the enrichment falls outside preset limits.

The in-line system was calibrated against mass spectrometer analysis of samples. The net ^{235}U signal in the gamma system is approximately 277 cps $(\%^{235}U)^{-1}$. Three per cent enriched material yields 4.93×10^5 counts in a 10-min count period, and the

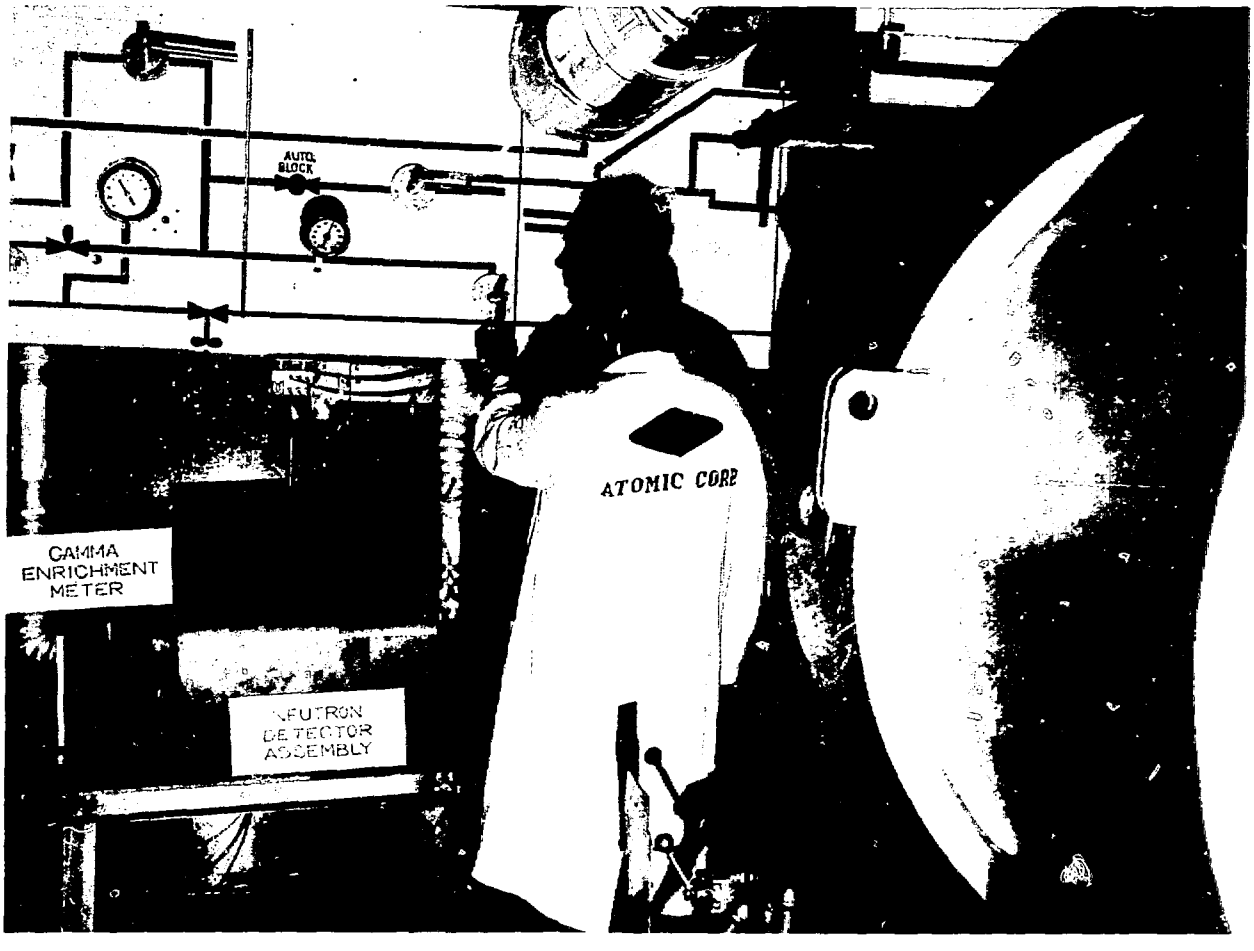

Fig. 20. In-line UF_6 enrichment monitor installation at the extended range product withdrawal station of the Portsmouth (GAT) gaseous diffusion plant.

equilibrium background (three months after startup) was ∿ 160 cps or 1.01×10^5 in 10 min. This background is almost entirely due to plating of ^{234}Th (24.1 d half-life) on the walls of the gamma chamber, with most of the gamma activity coming from its short-lived daughter ^{234m}Pa. The UF_6 from the cascade is nearly devoid of all daughter product activity and presents a very clean gamma signal for analysis; however, some of the daughter products produced in the stream are not carried along but rather plate out on the walls. Although the equilibrium background level does not interfere with the measurements in the 2 - 5% range, it limits the accuracy of the gamma-ray system at lower enrichments, particularly the assay of tails (∿ 0.3% ^{235}U) with NaI.

The net neutron signal from 3%-enriched material is approximately 70 000 counts in a 10-min period, 80% of which are from ^{234}U α activity. The background varies from ∿ 650 to 1750 counts per 10 min as the cylinder is filled.

Both the neutron and gamma-ray enrichment monitors perform reliably. The gamma spectrometer exhibits an accuracy of 0.5% (2σ), which is sufficient for process control and criticality safety requirements. The neutron spectrometer exhibits an accuracy of about 1.5% (2σ) for the measurement of ^{234}U enrichment. Figure 21 shows a comparison of gamma-ray and neutron enrichment measurements with the ^{235}U enrichment obtained from mass spectrometer analysis of samples. For this comparison a constant $^{235}U/^{234}U$ ratio was assumed to convert the neutron count to ^{235}U enrichment.

In-line monitoring of product and tails withdrawals are also of interest for perimeter safeguards of enrichment plants. While the design discussed above is quite satisfactory for monitoring product, it would have to be modified for the assay of tails. The question of whether a germanium detector is needed instead of NaI to overcome the background from ^{238}U daughters plated in the measurement cell must be resolved. This can be accomplished with some laboratory measurements and analysis of long-term data available at the Goodyear Atomic Corporation (GAT). The principal

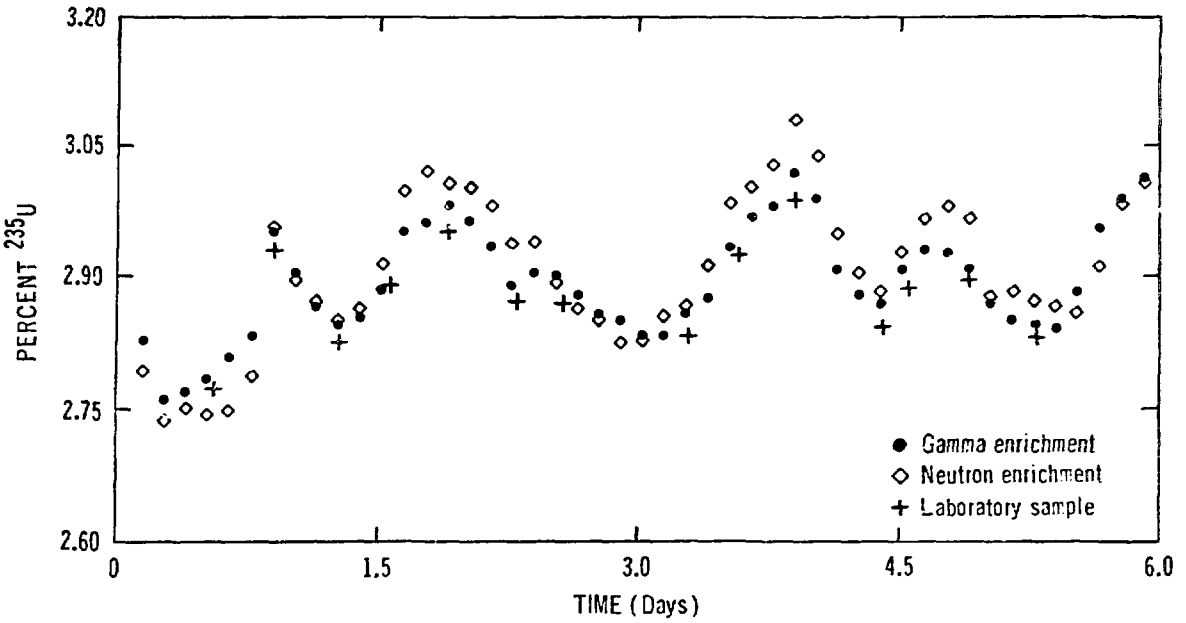

Fig. 21. Comparison of enrichments obtained with the Portsmouth (GAT) in-line gamma-ray and neutron assay systems with data from mass spectrometer analysis of samples. A constant $^{235}U/^{234}U$ ratio was assumed to derive ^{235}U enrichment from the neutron counting data.

modifications needed to adapt the neutron detector for assay of tails are the following: a. increase the number and length of the detectors, b. increase the size of the UF_6 sample chamber. Active neutron interrogation methods should be explored as an alternative to provide a direct assay of the ^{235}U in tails, namely the method described earlier for the assay of UF_6 cylinders.

<u>Neutron Measurements of ^{234}U Isotopic Abundance in UF_6 Samples</u>-- Neutron counting of UF_6 was further explored as a rapid method for determining the ^{234}U isotopic abundance with accuracy comparable to that of mass spectrometry. ^{234}U isotopic measurements are of interest for application of the minor isotopes tracer technique (MIST) for nonintrusive safeguards monitoring of ^{235}U, as

well as for process control, in enrichment facilities. The $^{234}U/^{235}U$ ratios observed for feed and product may be used to verify that the enrichment cascade has operated as declared; however, for this application the ^{234}U must be measured to an accuracy of a few per cent or better.

To provide a capability for the study of a wider range of ^{234}U enrichments than available at the Portsmouth (GAT) in-line station, we designed a 4Π well counter comprising 14 ^{3}He counters imbedded in a polyethylene cylinder with a central cavity for 1 S and 2 S UF_6 samples.[24]

This detector, shown in Fig. 22, was used to measure ^{234}U isotopic abundances of 19 2S and 5 1S UF_6 cylinders with ^{235}U isotopic abundances in ranges from 0.3 to 4.0% and 80 to 97.7%. (Typical net UF_6 weights for 1 S and 2 S cylinders are 800 g and 1700 g, respectively.) These results and comparisons with data obtained with a high-precision two-stage mass spectrometer have been reported by Stieff et al.[25] For counting times of 10^3 s, the neutron method achieved accuracies for the ^{234}U isotopic abundance better than 2.5% (1σ) for depleted and low-enriched UF_6 and better than 0.5% (1σ) for the high-enriched UF_6. Data for the depleted and low-enrichment ranges are shown in Fig. 23. Concerns that the presence of small amounts of ^{232}U, which has a high specific activity, would limit the application of this method, were confirmed by W. T. Mullins[26] in a subsequent evaluation of the LASL detector. The neutron results for the ^{234}U isotopic abundance in samples with as little as 30 ppb of ^{232}U per part of ^{235}U were ∿ 20% too high.

Detailed analysis of neutron yields in these studies indicates the need for better F(α,n) yield data for ^{232}U and ^{236}U in order to refine the passive-neutron counting method for analysis of UF_6.

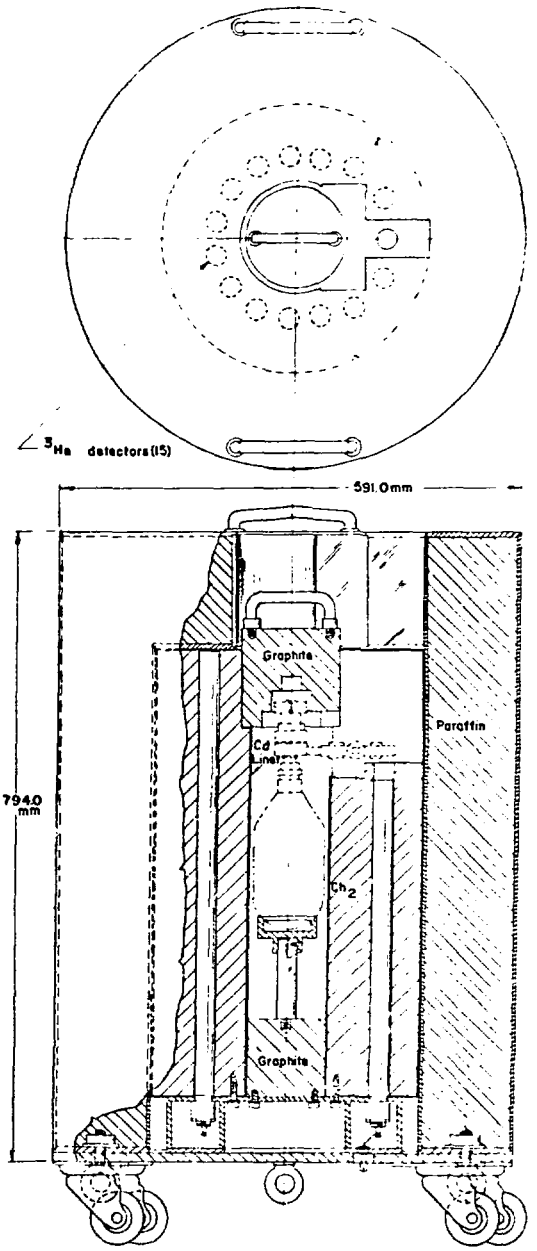

Fig. 22. Schematic of 4Π neutron detector for ^{234}U isotopic analysis of UF_6 in sample cylinders.

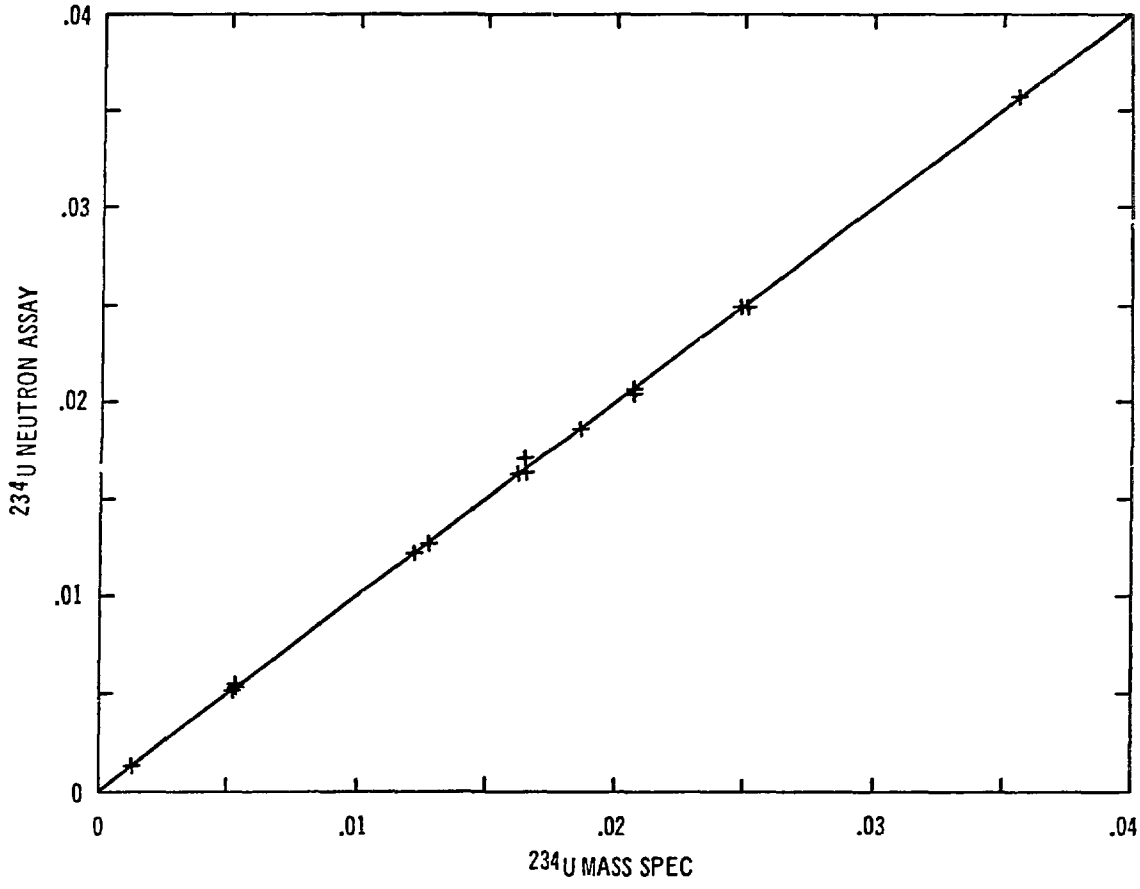

Fig. 23. Comparison of ^{234}U isotopic analysis by neutron counting with mass spectrometer results.

IV. SOME POTENTIAL APPLICATIONS OF NDA FOR CENTRIFUGE PLANTS

A. Minor Isotopes Safeguards Techniques (MIST)

In addressing the development of a safeguards system for containment and surveillance of enrichment plants, Hammond and Stieff[1] have noted the potential of MIST in a perimeter safeguards system where inspections are intermittent and inspector

access to the cascade is limited. Blumkin and Von Halle have performed some calculations of the sensitivity of the flows and isotopic compositions of feeds, product, and tails for corroborating material accounting results for a cascade.[27-30] While the calculations and some measurements[31,32] gave encouraging results for the MIST method, its application to a gaseous diffusion plant is complicated by the number of feed and withdrawal points, the length of the cascade, and the long residence time of the material in the cascade. The method may have a higher sensitivity when applied to a centrifuge cascade because of the great differences in the characteristics of centrifuge and diffusion cascades. Should further analyses show this to be the case, the in-line and at-line neutron and gamma-ray systems for the assay of ^{234}U and ^{235}U, described above, could be implemented in a perimeter safeguards system with little additional development, providing that UF_6 in solid or liquid form is available for the measurements.

B. Neutron Conservation

Neutron counting may also be used as a simple monitoring technique for perimeter safeguards because the total number of neutrons from the UF_6 crossing the perimeter should be conserved. Essentially this concept is a generalization of MIST. Two simplified examples of neutron and mass balance are presented in Table XII. The first case is for the generation of 3% enriched product from normal uranium feed, holding the tails fixed at 0.2% and assuming no diversion or plant holdup. The second example shows a slight adjustment of tails and product to obscure a diversion of 7.8% of the input mass of ^{235}U in the form of 93% enriched material. (In both cases, the ratio of $^{235}U/^{234}U$ was assumed to be 125.) It is evident that neutron measurements of input, tails, and product might be feasible to check for diversions of this magnitude, but for smaller fractional diversions, neutron counting would be useful only for the direct detection of the highly enriched material. Five hundred grams of uranium (93% ^{235}U), as UF_6, should produce a counting rate of $\sim$ 10 cps in a 1-m by 1-m neutron detector located 2 m from the material.

TABLE XII

ENRICHMENT MATERIAL BALANCES

	Feed	Tails	Product	Diverted Product
3% Enriched Product				
No diversion				
U mass	1000 kg	814.2 kg	185.6 kg	0
Enrichment	0.72%	0.20%	3.00%	
Neutrons/s[a]	5.14×10^4	2.60×10^4	2.54×10^4	0
2.95% Enriched Product				
Diversion				
U mass	1000 kg	823.9 kg	175.5 kg	0.563 kg
Enrichment	0.72%	0.182%	2.95%	93.0%
Neutrons/s[a]	5.14×10^4	2.58×10^4	2.37×10^4	1.97×10^3

[a]Calculated assuming constant $^{235}U/^{234}U$ ratio = 125.

C. Monitoring the Gas Phase in Centrifuge Plants

If access to centrifuge process equipment is allowed and if the gas pressures are sufficiently high, it may be possible to use gamma-ray detectors positioned at representative key points in the cascade to provide real-time data on the enrichment and pressure of the gas phase. J. Tape is currently investigating the limits of sensitivity of conventional gamma-ray and x-ray assay of enrichment of the UF_6 gas phase.[33]

Measurement of neutron fields with BF_3 or ^3He neutron counters placed at nonintrusive points within the facility may provide a simpler method for monitoring the gross gas phase inventory and enrichment. Neutron monitoring would not require close access to process equipment and, conceivably, could be implemented with access to the roof of the centrifuge facility. The results of the neutron survey in the K-33 operating plant (presented in

Table VI) showed the neutron field in a fully operating region of K-33, where the enrichment was approximately 3%, to be a factor of five higher than the background. Although this factor almost certainly will not compensate for the difference in gas pressures in diffusion and centrifuge process equipment, the space-averaged gas phase inventory of a centrifuge plant may be comparable to that of a gaseous diffusion plant because of the higher "packing" density of equipment in the former. A recently published photograph of centrifuges at the Almelo enrichment facility[34] appears to support this conjecture.

As a corollary to the neutron monitoring method, it may also be feasible to measure the "time-averaged" ^{238}U gas phase inventory by detecting the penetrating gamma rays from ^{234m}Pa.

D. Hold-Up Measurements in Centrifuges

Based on information from gaseous diffusion plants, uranium is expected to be retained preferentially on centrifuge components where the gas impacts with high velocities and where there may be air interfaces to the UF_6 process stream. Additionally, the chemically inactive layers on the inside of the process equipment may depend on the material type of the equipment. Some information about the design of the centrifuges and their accessibility would be required to make any meaningful prognosis about the feasibility of NDA for the measurement of holdup in centrifuge facilities.

A better approach may be to use NDA to measure broken centrifuge components and compare with the amount of uranium recovered from the decontamination of these parts. Historical data of this type should provide a reliable estimate of holdup with minimal impact on plant operations. Large localized deposits in the plant can be found with portable neutron and gamma-ray instruments, as is currently done in the gaseous diffusion plants.

V. CONCLUSIONS AND RECOMMENDATIONS

A. Conclusions

A host of NDA applications, together with their advantages and limitations, has been described for measurements of uranium at enrichment facilities. Essentially all of the information reviewed was based on tests performed at U. S. enrichment facilities with the participation of representatives of these facilities and the cognizant DOE field offices. These results should constitute a useful data base to project the future role of NDA measurement methods for advanced uranium enrichment processes, as well as gaseous diffusion.

The major conclusions of this review are the following.

1. Hold-Up Measurements--Residual uranium in the top enrichment portion of the shut-down K-25 cascade was successfully measured with portable neutron and gamma-ray instruments with accuracies of approximately ± 50%. Inventory estimates based on these measurements were in good agreement with original estimates by ORGDP. Attempts to measure holdup in the operating low-enrichment (∿3%) K-33 cascade proved that, except for large localized deposits, the radiation from the in-process gas phase inventory precludes detection of bonafide holdup. The ^{235}U 186-keV gamma rays do not have sufficient penetrability for quantitative assay of holdup in the large compressors and converters used for low-enriched uranium (K-33 converter dimensions are 4-m diam by 5.5-m long). Additionally, the neutron yield per g U would be lower relative to that for the K-25 cascade because of the enrichment differences and geometry factors. With respect to achievable assay accuracy, the detection of gamma rays from ^{234m}Pa is probably the best means for quantitative assay of low-enriched U holdup; however, a "cooling" period of several months would be required to allow the daughters of the last process flows to decay.

2. Applications for the Operating Cascade--Data obtained with portable gamma-ray instruments showed that they can be used for dynamic monitoring of U accumulation in NaF traps for the main

gaseous effluent stack of the diffusion plant and to indicate when both NaF and alumina traps are expended. Three per cent enriched U in trapping media can be readily detected for concentrations greater than 0.2 mg U/cm^3. In operating diffusion cascades, portable gamma-ray instruments detecting 186 keV gamma rays can be used to measure the gas density of ^{235}U in interstage piping for the complete spectrum of enrichments. Detection of high-energy gamma rays from ^{234m}Pa provides a measure of the ^{238}U gas phase inventory appropriately time-averaged by the 24.1-d half-life of ^{234}Th over recent operating history. Surveys of the neutron fields in the corridors of K-33, performed with a portable neutron detector, showed this technique could provide a qualitative measure of the operating status of the cascade, the signal being a factor of five above background in a fully operating portion of the cascade.

3. Enrichment Plant Scrap and Waste--Passive neutron and gamma-ray assay methods and 14-MeV neutron interrogation were used to measure over 400 items of ORGDP scrap and waste including used NaF and alumina trap media, incinerator ash, miscellaneous cylinders and used barrier and compressor blades. It was concluded that for most of these materials NDA measurements were superior to chemical anlysis because of the sampling problem. A NaI photon detection system with sensitivities in the sub-MPC region for all major isotopes of interest was successfully demonstrated for monitoring liquid effluents and holding ponds at ORGDP.

4. Measurements of UF_6 Cylinders, Product, and Tails--Passive neutron and gamma-ray counting with portable instruments are now widely accepted methods for verifying the enrichment of cylinders of UF_6. The relative accuracy of the gamma-ray method is $\sim$ 5% for type-30 cylinders of low-enriched (< 5%) UF_6 and $\sim$ 2% for 5-A cylinders of highly enriched UF_6, and the accuracy of the passive neutron method is $\sim$ 6% for type-30 cylinders. An in-line neutron and gamma-ray assay system has operated since 1973 at the extended-range product withdrawal station at the Portsmouth (GAT) gaseous diffusion plant. The relative accuracy of the NaI gamma-ray system for measuring the ^{235}U

enrichment of liquid UF_6 in the enrichment range 2 - 5% is 0.5% (2σ), and the neutron detector measures the ^{234}U enrichment to a relative accuracy of $\sim$ 1.5% (2σ). This type of in-line system, originally developed for criticality safety, may be useful for perimeter safeguards systems where feed and withdrawal points can be monitored. The measurement of neutrons from UF_6 with stand-alone detectors provides a rapid, accurate means for measuring the ^{234}U isotopic abundance, an important capability for implementation of MIST in enrichment plant safeguards.

B. Recommendations

As a consequence of extensive safeguards test and evaluation programs, several NDA methods have already been implemented in enrichment plants; however, the present review has also identified additional studies and tests that should be considered, particularly for centrifuge facilities. The following specific recommendations are offered.

1. A comprehensive systems analysis of the materials measurement and accounting system should be performed for a specific facility to better define the role and effectiveness of materials measurement and management in an integrated safeguards design. Similar studies for other fuel cycle facilities have been reported.[35,36] The results of a facility specific analysis should provide important guidance for defining measurement points, instrumentation, and performance requirements for enrichment facilities using the same generic processes.

2. The practicality and effectiveness of the MIST method for domestic and international safeguards should be better and more specifically defined in the context of a total materials management system for enrichment facilities, particularly for centrifuge facilities. Measurement methods could then be developed to meet indicated requirements. The neutron balance concept, which is generically related to both MIST and mass balancing, may provide a nonintrusive corroborative safeguards method, depending on results of the analysis of MIST and material accounting systems.

3. An in-line enrichment monitor for continuous measurement of the enrichment of tails withdrawals should be developed and tested at a host enrichment facility because of its potential for perimeter safeguards, criticality safety control, and possibly MIST.

4. The feasibility of using neutron detectors placed at non-intrusive points within a centrifuge plant for monitoring of gross gas phase inventory and enrichment should be further explored, preferably with tests conducted at an operating centrifuge facility.

5. Methods for measuring the enrichment of UF_6 in centrifuge gas transfer pipes should be developed for potential safeguards and process control applications. NDA gas phase enrichment measurements would be evaluated by comparing them with routine temperature, pressure, and mass spectrometer measurements at selected key points in an operating facility.

6. It should be determined if better estimates of holdup in large gaseous diffusion components (used in the low-enriched segment of the cascade) can be obtained by refining the procedures used to sample and measure the U in solutions that are used to decontaminate components that have been removed from the cascade. Correlation of the uranium assay of the decontamination solutions with the sizes and types of equipment should provide good data on typical holdup. Currently it is difficult to obtain reliable differential data because of problems in drawing representative samples from the several tanks in which the solution is stored.[10] An in-line version of the Uranium Solution Assay System (USAS),[37] developed for the LASL uranium recovery facility, is a potentially cost-effective means for implementing such a measurement program. This approach to hold-up estimates should also be considered for centrifuge plants.

7. The merits of using neutron counting to measure UF_6 in condenser-sublimers that may be added to U. S. diffusion plants under the CIP program should be investigated as an alternative to weighing with load cells.

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