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A CONTINUOUS IN-LINE MONITOR FOR UF₆ ENRICHMENT

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A system has been developed to measure continuously the ^{235}U and ^{234}U enrichment in the UF_6 product of a gaseous diffusion plant. The measurements are made on liquid UF_6 prior to withdrawal into product cylinders. The ^{235}U enrichment is measured to a relative accuracy of 0.5% at two sigma by counting the 185.7-keV gamma ray from ^{235}U with an NaI detector. The ^{234}U enrichment is measured with a neutron detector counting (α,n) neutrons from UF_6. The neutron system can measure either ^{234}U enrichment directly or ^{235}U enrichment indirectly (for low-enriched uranium, 2 to 5% ^{235}U, the two enrichments are nearly proportional). The accuracy of the neutron measurement is 2.5% at two sigma. The gamma and neutron measurements are independent and could be used singly if only one isotope were desired. Both are required to measure ^{235}U and ^{234}U enrichment. The system is presently installed at the Goodyear Atomic Corporation gaseous diffusion plant in Piketon, Ohio.

GENERAL DESCRIPTION

The UF₆ enrichment monitor is a system to measure continuously the enrichment of ^{235}U and ^{234}U in the UF₆ product of a gaseous diffusion plant [enrichment is defined here as (mass ^{235}U/ mass U) × 100% and (mass ^{234}U/mass U) × 100%]. The measurements are made on liquid UF₆ prior to withdrawal into product cylinders. The system is presently installed at the Goodyear Atomic Corporation gaseous diffusion plant in Piketon, Ohio. It will be used for process enrichment control and criticality control, to guarantee that highly enriched UF₆ is not withdrawn into a low-enrichment cylinder.

The major gamma radiation from ^{235}U is at 185.7 keV. For a sample of constant volume the intensity of this radiation is proportional to ^{235}U enrichment.[1,2] An NaI scintillator detects the 185-keV gamma rays, and two single-channel analyzers (SCAs) are used to strip these events from the background.

There are two major components of the neutron signal from UF₆. Alpha particles from uranium (mostly ^{234}U) react with fluorine to produce fast neutrons,

$$^{19}F(\alpha, n)^{22}Na \quad .$$
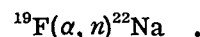
For a fixed mass of UF₆, this signal is proportional to the ^{234}U enrichment. The other neutron source is ^{238}U, through spontaneous fission and (α,n) reactions. The ^{238}U signal is about equal to the ^{234}U (α,n) signal in normally enriched UF₆. This represents a background to the ^{234}U enrichment measurement. The neutron yield of UF₆ can be approximated by the expression[3,4]

$$n/sec \; gU = 5.76 \, I_4 + 0.0285 \quad ,$$

where I_4 is the percent ^{234}U.

For low-enriched uranium (2 to 5% ^{235}U), the ^{235}U enrichment is almost directly proportional to the ^{234}U enrichment.[5] In this region the neutron signal can be used as an indirect measurement of ^{235}U enrichment. At higher enrichments the proportionality breaks down and the neutron signal can only give ^{234}U enrichment. The neutron system can be operated to output either ^{234}U enrichment (the direct measurement) or ^{235}U enrichment (assuming ^{235}U tracks ^{234}U) by a simple change in the calibration constants used for the enrichment computation. At present the system at Goodyear is set up to compute ^{235}U enrichment, since this is the parameter of interest to the withdrawal station

operators. In this mode the two detectors (gamma and neutron) provide duplicate measurements of ^{235}U enrichment. A single detector could provide this measurement. The ^{234}U enrichment is of particular interest for nuclear safeguards, and in the near future the neutron calibration will be changed to compute ^{234}U directly. Both detectors are required to measure the two isotope enrichments.

Figure 1 shows a block diagram of the system. The sample chambers and detectors are located in the extended range product (ERP) withdrawal station at Goodyear. This assembly was connected to the existing withdrawal line, and a control valve was added to direct the UF$_6$ flow through the instrument line. An elevated vapor bypass provides a low-resistance path for vapor bubbles which may form in the UF$_6$. The vapor bypass should normally be empty and the measurement loop must be full in order to get a meaningful answer. Although not indicated in this figure, sufficient valves and vent lines have been added so that the instrument

loop can be isolated from the rest of the system without interfering with product withdrawal.

Pulses from the gamma and neutron detectors are amplified and fed to the control-arithmetic unit. The unit scales these pulses for a set time period of 2, 5, or 10 min. At the end of each count period, it computes the percent ^{235}U and percent ^{234}U from the accumulated count and predetermined calibration constants. Scaler counts, time-of-day, and the computed enrichments are displayed on the front panel and printed as a continuous record on the teletype. The gamma and neutron sections are electrically separate but are contained in one chassis.

If the computed enrichment goes above or below preset limits, the control unit issues a high or low alarm. These alarms are front panel lights and rear panel switch closures to drive other warning systems and operate valves (e.g., the block valve would be closed on high alarm to stop UF$_6$ withdrawal).
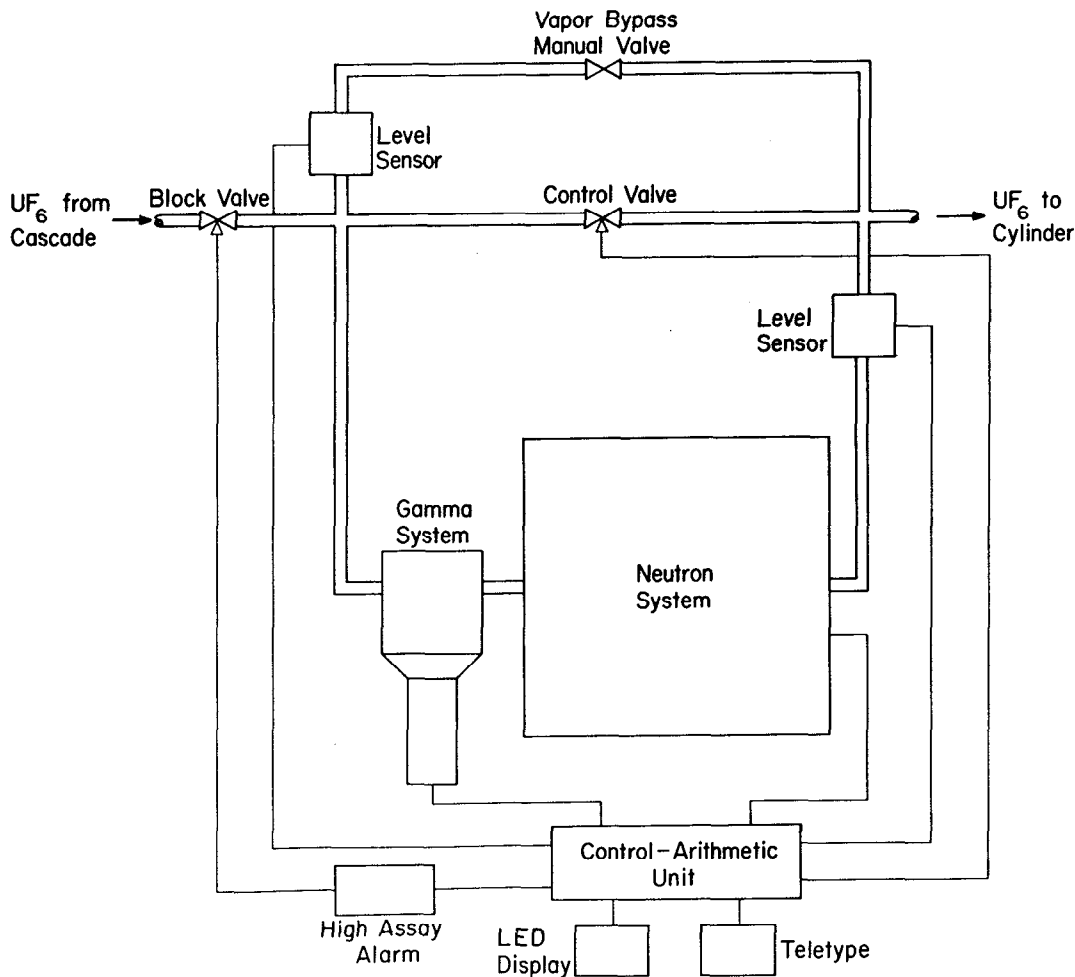

Fig. 1.   UF$_6$ enrichment monitor block diagram.

DETECTORS AND ELECTRONICS

To define the fixed volume required for the gamma-ray measurement, the UF$_6$ flows through a circular chamber viewed by the NaI detector. Figure 2 shows the gamma chamber, detector, and associated lead shielding. The heater and insulation are required to keep the UF$_6$ in the liquid state. The detector is a 5.1-cm-long × 1.3-cm-thick NaI scintillation detector. A 0.23-cm-thick cadmium absorber is used to suppress the undesirable low-energy x-ray portion of the uranium spectrum. A 4-μCi ^{241}Am gamma source is glued to the face of the detector to provide a constant reference peak (59.5 keV) for electronic gain stabilization. The gross rate from this source is ~6000 pulse/sec. The crystal experiences daily temperature variations of 10°C and even greater seasonal variations. A 10°C increase in the detector temperature causes about a 6% drop in gain;

the stabilizer compensates for this change, giving no measurable change in the peak count rate or the computed enrichment.

The activity in the 185.7-keV line is measured with two single-channel analyzers, one set over the peak (130 to 230 keV) and the other set above the peak (240 to 340 keV), to provide a correction for the Compton background from higher energy gamma rays. The activity in these two windows is scaled and analyzed in an arithmetic-control unit designed for this system.[6] It scales the activity and computes the ^{235}U enrichment from the expression

$$\% \, ^{235}\text{U} = A \cdot C1 - B \cdot C2 \quad , \qquad (1)$$

where A and B are calibration constants and $C1$ and $C2$ are the observed count rates. The calibration constants are determined from mass spectrometer data and are set by front-panel
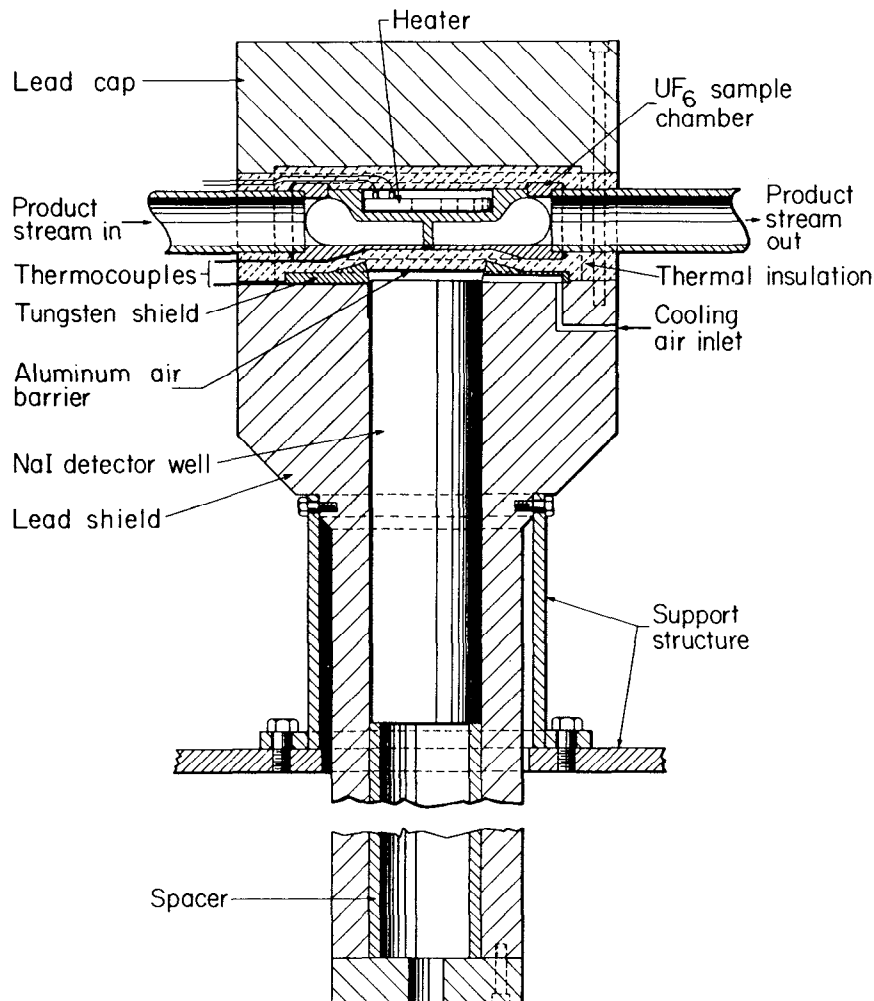

Fig. 2.   Gamma-enrichment meter—schematic diagram of detector and shield assembly.

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thumbwheel switches. The computed enrichment is displayed on front panel LEDs and printed on a teletype.

A block diagram of the system electronics is shown in Fig. 3. All electronics, except the control-arithmetic unit and the preamplifiers, fit in one standard NIM bin. The bin and the control-arithmetic unit are located in a small rack cabinet with covers to limit access to important controls (Figs. 4 and 5). The electronics cabinet and tele-type are in an air-conditioned instrument room near the withdrawal station.

Figure 6 shows a schematic drawing of the neutron sample chamber, the neutron detector, and the shield assembly. The detector surrounds a fixed mass of UF$_6$ defined by a 30-cm-long × 7.6-cm-diam pipe section in the UF$_6$ stream. This detector consists of 16 ^{3}He proportional counters in a polyethylene moderator and is surrounded by ~12.7 cm of polyethylene shielding to reduce the background from the nearby UF$_6$ product cylinder. The efficiency of this detector for neutrons origi-nating in the UF$_6$ sample chamber is ~20%. Neutron pulses from the detector are amplified and scaled in the arithmetic-control unit. The section of the control unit that analyzes the neutron activity scales neutron pulses and computes either ^{234}U enrichment or ^{235}U enrichment from the ex-pression

$$\% \, ^{234}U \text{ or } \% \, ^{235}U = D \cdot C3 - E \quad , \qquad (2)$$

where D and E are calibration constants and $C3$ is the observed neutron activity. The computed en-richment is displayed on LEDs, printed on the teletype, and compared with preset limits to de-termine if an alarm condition exists. The neutron and gamma sections are independent except that they share the same count interval. Figure 7 shows the completed detector assembly prior to installation.

PERFORMANCE AND CALIBRATION

Gamma-Ray System

The ^{235}U enrichment in UF$_6$ in the instrument loop must be accurately known to calibrate this system. The instrument loop cannot be sampled directly, so calibration data come from routine mass spectrometer samples taken from the gas stream ahead of the in-line monitor. If the assay (^{235}U enrichment) of the withdrawal stream is changing, there will be a time delay between the change at the gas withdrawal points and the change at the instrument loop and cylinder pigtail. This delay is a function of the volume of material be-tween the two points and the flow rate; it can be as large as 30 to 60 min. In view of this, calibration data are obtained when the withdrawal assay is running relatively constant so that the gas samples can be expected to reflect accurately the assay of material in the instrument loop.
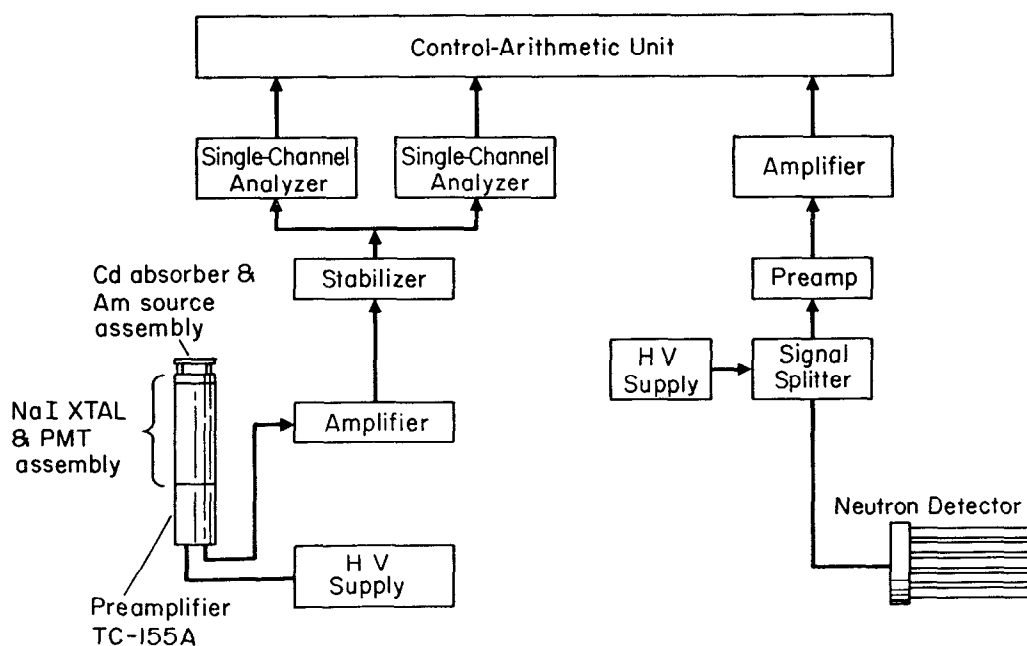

Fig. 3.   System electronics block diagram.

The response of the gamma system is assumed to follow Eq. (1). To accurately determine A and B, 20 calibration points were measured over a three-week period (Aug. 11 to Sep. 2, 1973). Each point involved three numbers, the ^{235}U enrichment as determined by the laboratory analysis of a routine UF_6 sample, and values of $C1$ and $C2$ taken as the average of six 10-min counts immediately following the withdrawal of the laboratory sample. These points were least-squares fit to the assay equation to determine the best estimates of the parameters A and B. Using these parameters, the standard deviation of the relative difference between the gamma assay (% ^{235}U) and the laboratory sample analysis for all 67 samples in this three-week period is ~0.25%. This indicates very good agreement between the gamma-enrichment meter and the laboratory sample analysis. Figure 8 shows a graph of gamma enrichment versus laboratory sample for a six-day period. Gamma measurements are plotted every 3 h for legibility; measurements are taken every 10 min. This illustrates the excellent agreement between the two measurements.

The present calibration indicates a net ^{235}U signal of ~277 counts/sec per % ^{235}U. The background is ~160 counts/sec but is variable. Most of this background comes from a plating of nonvolatile ^{238}U daughter products on the walls of the gamma chamber. The gamma-ray activity results from the decay of ^{234m}Pa which follows ^{234}Th in the ^{238}U decay chain. The UF_6 alone presents a very clean
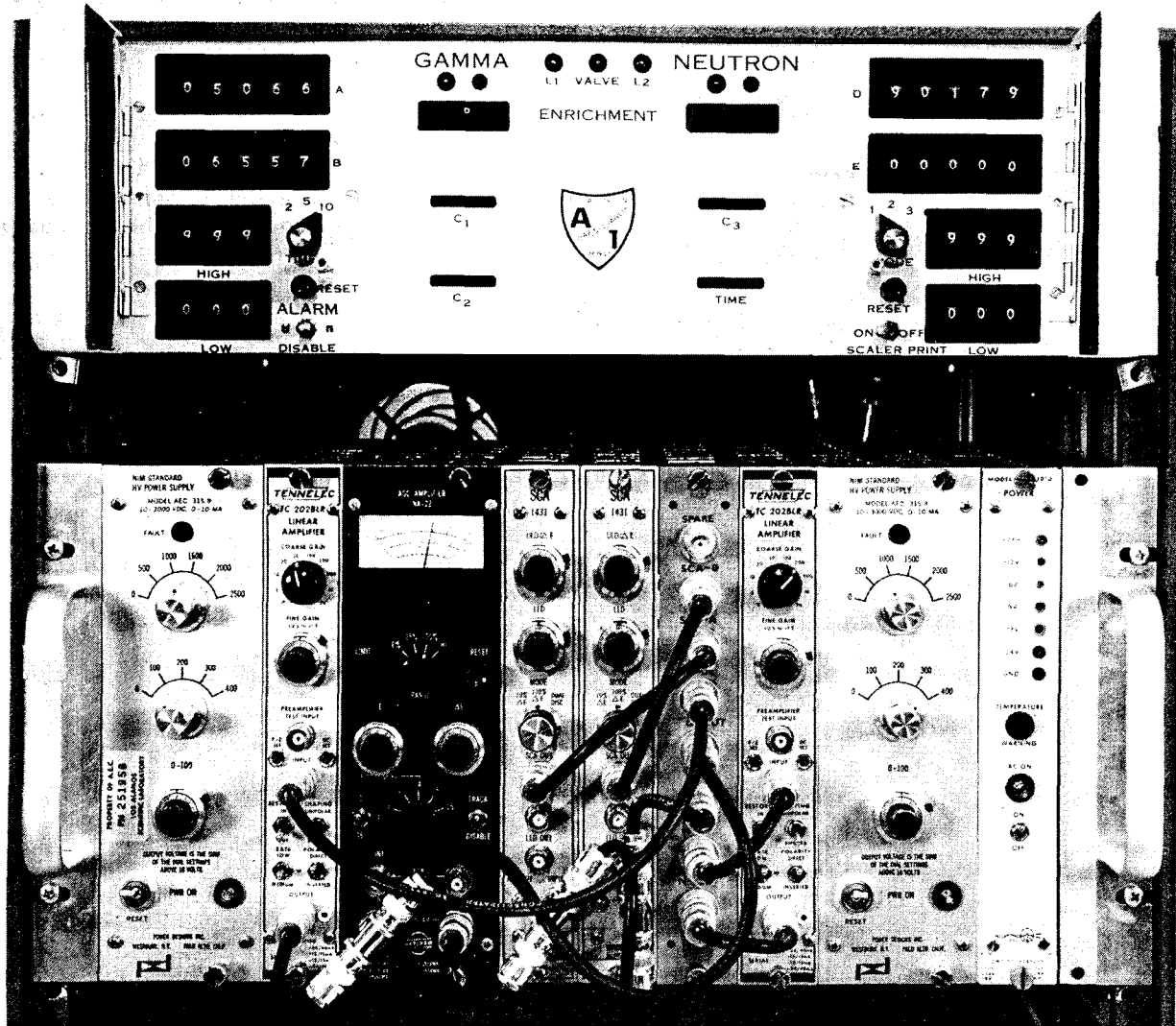

Fig. 4.   Electronics cabinet with protective covers removed.

signal for analysis, since most of the daughter products have been removed in the diffusion process. The background is fairly large and varies with time. In particular, it increases significantly whenever the withdrawal stops and a UF$_6$ freezeout occurs in the instrument loop (some freezeout seems to occur whenever the UF$_6$ flow is stopped). This daughter product plating occurs on all system piping. It represents a minute amount of material, ~10^{-9} g ^{234}Th/month. During the first three months of operation the background count rate rose from <10 counts/sec to an equilibrium value of about 160 counts/sec (the time scale is determined by the 24-day half-life of ^{234}Th). While the background may increase considerably during periods of interrupted flow, it returns to the equilibrium value when flow is resumed. This varying background does not affect the computed ^{235}U enrichment as it is subtracted out by the high energy SCA. Once calibrated, the gamma system has operated very stably with little need for recalibration.

Neutron System

The response of the neutron counter is assumed to follow Eq. (2). As explained earlier the neutron system can output either ^{234}U enrichment (the directly measured quantity) or ^{235}U enrichment (assuming ^{235}U is directly proportional to ^{234}U). Goodyear has chosen the latter mode of operation at present. To date, the calibration and evaluation of the neutron measurement have been based on a correlation of the neutron signal with ^{235}U enrichment, as determined by mass spectrometry of routine UF$_6$ samples. Nevertheless, because of the near proportionality of ^{235}U and ^{234}U, the direct ^{234}U measurement will exhibit similar accuracy to the indirect ^{235}U measurement.

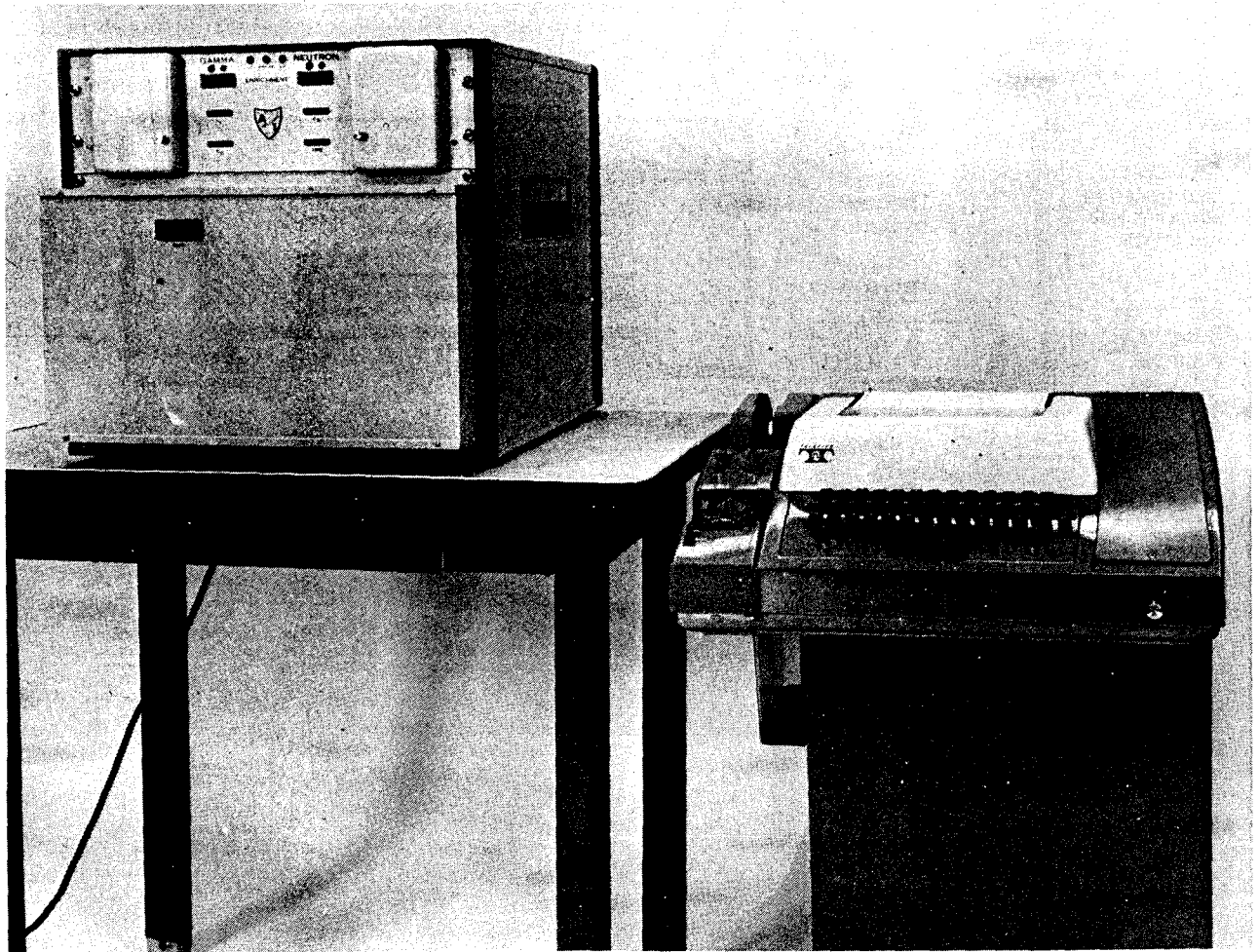The assay equation can be rewritten

Fig. 5.   UF$_6$ enrichment monitor—electronics cabinet and teletype printout.

$$\% \,^{235}\text{U} = D \cdot (C3 - E/D) \quad ,$$

where E/D represents the ambient room background and the background from ^{238}U in the chamber. This number is independent of ^{235}U and should remain constant except for a variation in the ambient background with cylinder fill weight. In the present location the detector is about 3 ft from the filling product cylinder which is the major source of ambient background. As the cylinder fills, the background varies from ~650 to 1750 counts/10 min. This variation represents an uncertainty in the neutron assay of ~±0.03% ^{235}U. The total neutron signal from 3% enriched material is ~70 000 counts/10 min.

Over 100 calibration points were determined during a four-week period in June and July 1973. These points and selected subsets thereof were least-squares fit to the assay equation to determine values of D and E. It was assumed that the large number of points included measurements at all stages of cylinder filling so that the fit gave an average over variations in the ambient background. For short periods ($\cong 1$ week), the standard deviation of the relative difference between the neutron assay and the laboratory sample was observed to be ~1.3%. This is consistent with the limit set by counting statistics and the variability of the ambient background (~1.1%). For longer periods, the agreement is not as good because the assumption of direct proportionality between ^{235}U and ^{234}U is not strictly correct. The ratio, $^{235}\text{U}/^{234}\text{U}$, will vary slightly as the operating parameters of the cascade change. In a one-month period, this ratio was observed to drift over a range of 7%. Based on available information, even larger changes can be expected. The neutron calibration (for the indirect ^{235}U measurement) must be checked and adjusted frequently to account for such changes. Obviously, the direct ^{234}U enrichment measurement does not suffer from this problem.
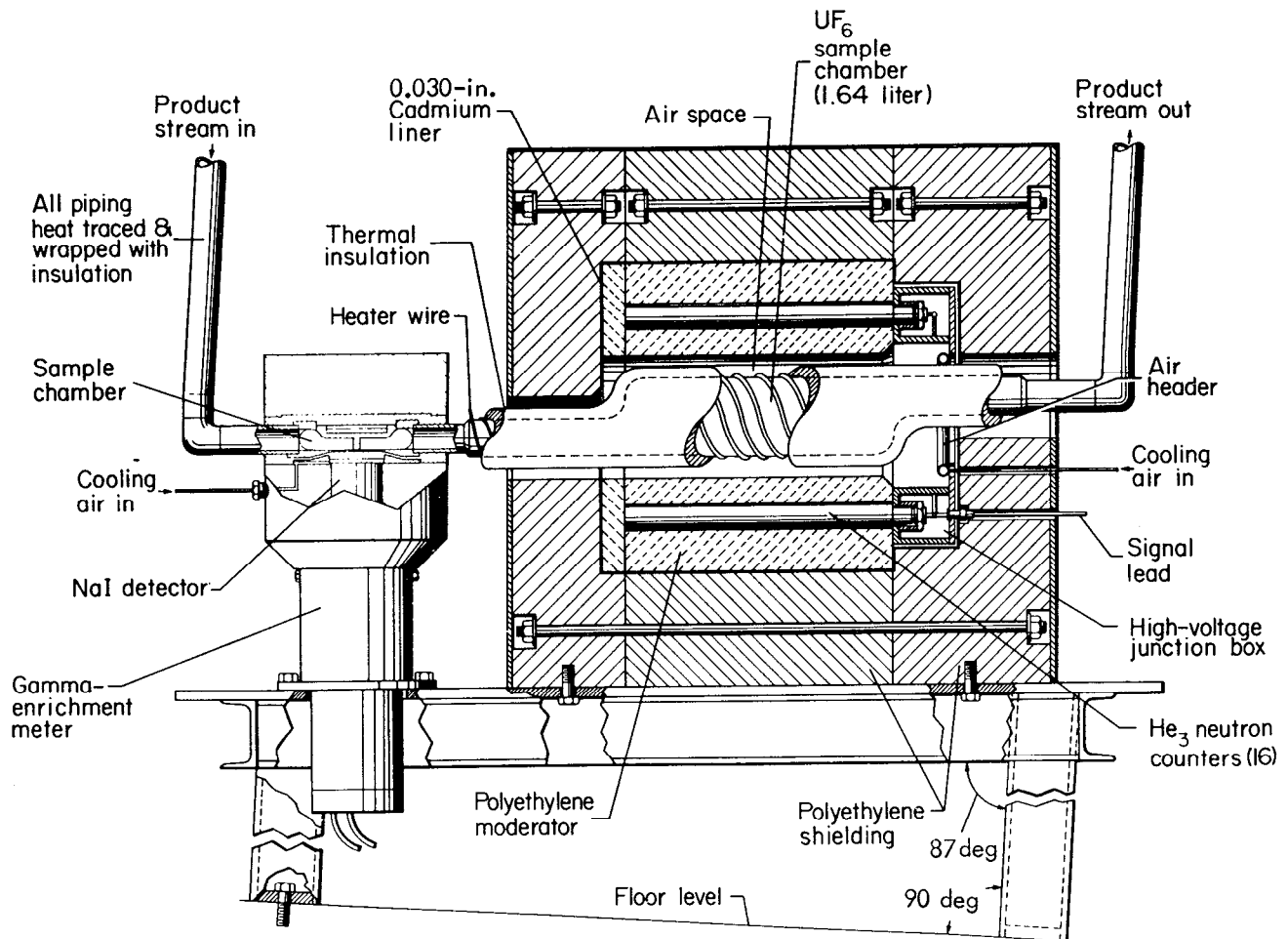
The assay expression, Eq. (2), ignores any

Fig. 6.   UF$_6$ enrichment monitor—schematic diagram of gamma spectrometer and neutron detector with shield assemblies. Though provisions were made for air cooling the detectors, tests later indicated that this was not necessary. The diameter of the neutron shield assembly is 61 cm.

multiplication effects in the counter. These were estimated with a Monte Carlo neutron transport code. This analysis gave the ratio of neutron counts from 5% (^{235}U) material to that from 1% material as 5.33 ± 0.06, showing the magnitude of the error made by assuming a linear equation for percent ^{234}U. This effect can be greatly reduced (with a loss in detection efficiency) by adding a thin sheet of cadmium between the detector and the neutron sample chamber.

CONCLUSIONS

In summary, both parts of the in-line UF$_6$ monitor function reliably and exhibit precisions consistent with counting statistics. The accuracy is good as is illustrated in Table I. This shows a comparison of laboratory samples (mass spectrometry) with the corresponding in-line measurements (gamma and neutron) during a six-day period. The laboratory analysis was taken to be the true ^{235}U concentration, and the fractional standard deviation between this and the measured enrichment was computed. If the neutron portion of the system is set to compute ^{234}U enrichment, a similar (or slightly better) accuracy will obtain.

As stated earlier both parts of the system are required to measure both isotopes. If, however, only one isotope were desired, a single detector could be used (gamma for ^{235}U or neutron for ^{234}U).
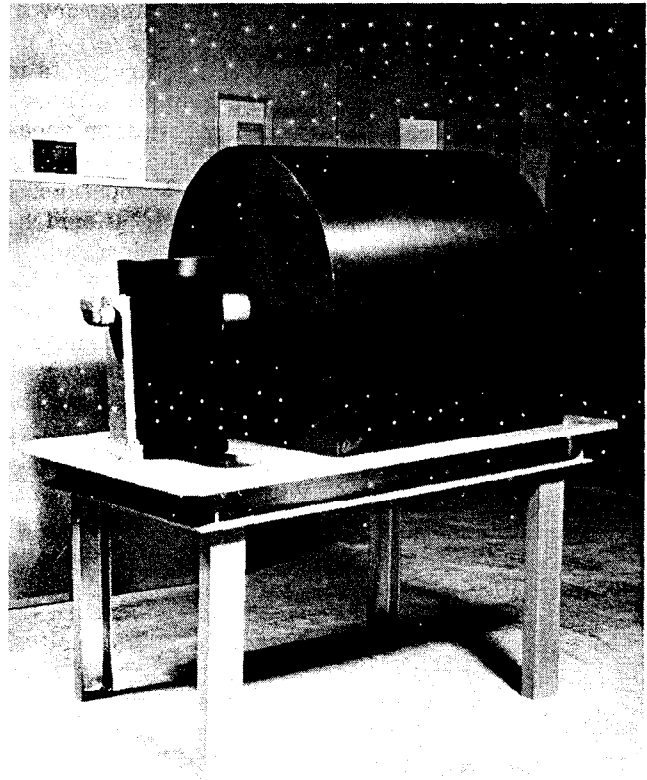

Fig. 7.   Detector assembly prior to installation. Gamma detector is on left, neutron detector on right.
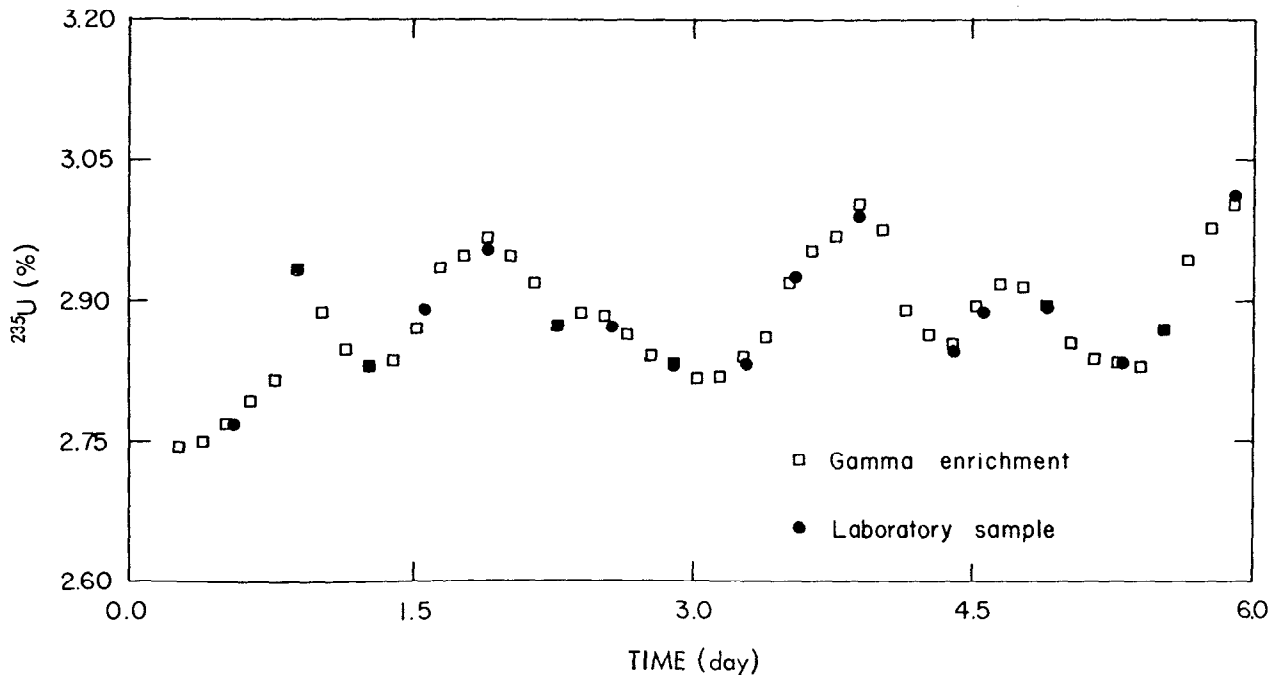

Fig. 8.   Comparison of gamma-enrichment measurements with percent ^{235}U as determined from routine laboratory samples. The points which appear as black squares indicate the two measurements overlap.

The gamma spectrometer exhibits an accuracy of ~0.5% (relative) at the 95% confidence level. This is sufficient for process control requirements in addition to criticality safety requirements. Though it has not been checked directly, the neutron spectrometer should measure ^{234}U enrichment with an accuracy of better than 2.5% at the 95% confidence level. The indirect measurement of ^{235}U enrichment with this device has a similar accuracy if the instrument is recalibrated often to account for changes in the ^{235}U/^{234}U ratio. This ratio has been observed to vary over 7%, and larger variations can be expected. Because of wide variations in the ^{235}U/^{234}U ratio for ^{235}U enrichments above 5%, neutron counting cannot be used for the quantitative assay of ^{235}U. It can be used over the complete range of ^{234}U enrichment. While the gamma system is optimized for low-enriched materials, it can cover the full enrichment range with very few modifications.

The gamma system can be used to measure the ^{235}U concentration (mass ^{235}U/volume of solution) of many uranium process solutions. Such a system is being designed for in-line measurements at the Los Alamos Scientific Laboratory uranium recovery plant. In the present application, the gamma spectrum is very simple. In other poten-tial applications, interferences may exist which prohibit the use of NaI detectors [e.g., at a fuel reprocessing plant ^{237}U (208 keV) in the product UF$_6$ will mask the ^{235}U (185 keV) signature]. In such situations the use of a high-resolution Ge(Li) detector will alleviate the problem and should result in performance equal to that exhibited here. The accuracy exhibited by the gamma system is somewhat unique at this state in the development of nondestructive assay instrumentation. It approaches or equals that available in many destructive analytic techniques, and it offers a continuous measurement of the entire product output.

TABLE I

Comparison of Mass Spectrometer Sample Analysis (% ^{235}U) with In-Line Measurement (gamma and neutron) During One Six-Day Period

Sample	Gamma	(Gamma − Sample)/Sample	Neutron	(Neutron − Sample)/Sample
2.869	2.871	0.0008	2.824	−0.0157
3.013	3.009	−0.0011	2.982	−0.0103
2.790	2.789	−0.0002	2.759	−0.0111
2.898	2.900	0.0006	2.862	−0.0124
2.911	2.910	0.0003	2.894	−0.0058
2.919	2.921	0.0006	2.911	−0.0027
2.946	2.940	−0.0020	2.972	0.0088
2.969	2.974	0.0016	2.975	0.0020
2.964	2.965	0.0003	2.953	−0.0037
2.817	2.813	−0.0015	2.765	−0.0184
2.819	2.825	0.0020	2.767	−0.0184
2.833	2.824	−0.0031	2.776	−0.0201
2.870	2.862	−0.0028	2.874	0.0014
2.888	2.891	0.0010	2.868	−0.0069
2.905	2.904	−0.0004	2.911	0.0021
3.176	3.181	0.0014	3.230	0.0170
3.072	3.079	0.0024	3.127	0.0179
3.073	3.076	0.0009	3.130	0.0185
		mean = 0.00004		mean = −0.003
		sigma = 0.0016		sigma = 0.013

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