

A GAS-PHASE UF₆ ENRICHMENT MONITOR
FOR ENRICHMENT PLANT SAFEGUARDS

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Abstract

An in-line enrichment monitor is being developed to provide real-time enrichment data for the gas-phase UF₆ feed stream of an enrichment plant. The nondestructive gamma-ray assay method can be used to determine the enrichment of natural UF₆ with a relative precision of better than 1% for a wide range of pressures.

1. Introduction

One of the primary strategies for an international safeguards system at a uranium enrichment plant is material accounting on the declared feed, product, tails, and waste streams of the facility. On-line enrichment data combined with on-line weights for the feed, product, and tails cylinders are necessary to implement a real-time dynamic material balance for the plant.¹ The performance of an in-line enrichment monitor for liquid UF₆ product, which has successfully operated at the Goodyear Atomic diffusion plant since 1973, has been previously reported.² However, feed streams at enrichment plants are typically in the gas phase, and the enrichment principle employed in the Goodyear Atomic product monitor is not feasible for most gas-phase enrichment measurements. The gas-phase enrichment is determined by combining measurements of the ²³⁵U and the total uranium concentrations in the gas. A NaI(Tl)-based in-line enrichment monitor is being developed to provide real-time enrichment data for the gas-phase UF₆ feed stream in the feed and withdrawal building of an enrichment plant. The ²³⁵U concentration is determined from a measurement of 186-keV gamma rays from ²³⁵U. The total uranium concentration is determined by measuring the transmission through the UF₆ gas of 60-keV gamma rays from an external ²⁴¹Am source.

2. Experiment

To investigate the physical principles upon which the enrichment monitor is based, a prototype instrument was tested under controlled laboratory conditions. The laboratory enrichment monitor consists of a UF₆ gas-handling system, sample chamber, NaI(Tl) detector, an ²⁴¹Am transmission source, nuclear electronics, pressure and temperature instrumentation, and a data acquisition system. Two sets of measurements have been made, one over the low-pressure range of 10 to 150 torr and the other over the high-pressure range of 175 to 700 torr. The low-pressure data were taken using a gas-handling system operated at ambient temperature. A modified gas-handling system, designed to maintain the sample chamber at a constant temperature of 60°C and to

provide UF₆ pressures in excess of 800 torr, was used to perform the high-pressure measurements. The stainless-steel cylindrical sample chamber used for both low- and high-pressure measurements has a 14.6-cm diameter and is 9.83-cm high. The top and bottom chamber walls through which the various radiations must pass are 0.079-cm thick. A different NaI(Tl) detector was used for each of the two pressure ranges. Both detectors were 12.7 cm in diameter and had a NaI light pipe. The low-pressure measurements were performed with a 2.54-cm-thick detector and the high-pressure measurements with a 2-cm-thick detector. Figure 1 is a schematic of the prototype system showing the orientation of the detector, the sample chamber, and the location of the ²⁴¹Am transmission source. The low-pressure measurements were performed with a 100-μCi ²⁴¹Am source, and a 200-μCi source was used for the high-pressure measurements. The transmission-source strength was increased to compensate for the additional structural and insulating material between the sample chamber and detector required by the heating system. The sample chamber is instrumented with thermocouples and a precision capacitive manometer for temperature and pressure measurements respectively.

Depleted UF₆ (0.2% ²³⁵U) was introduced into the sample chamber by vapor transfer. Measurements were made of the count rates of 186- and 60-keV gamma rays from ²³⁵U and ²⁴¹Am respectively and of rates produced by an electronic pulser. The thermocouple and

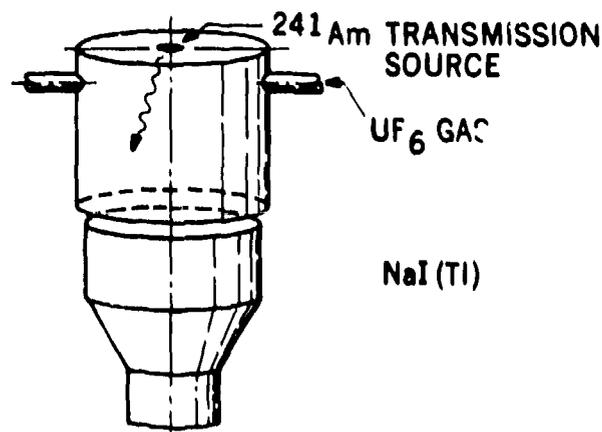


Fig. 1. NaI(Tl)-based gas-phase UF₆ enrichment monitor. The intensity of 186-keV gamma rays emitted by ²³⁵U and the transmission through the UF₆ gas of 60-keV gamma rays from an external ²⁴¹Am source were measured and the data were used to determine the ²³⁵U enrichment.

manometer readings were also recorded. A fraction of the sample gas was removed by cryo-pumping, and the measurements were repeated.

3. Results

The enrichment, I , is related to the measured count rate of 186-keV gamma rays, R , by

$$I = R/C2nT_{60} \quad (1)$$

where R is corrected for deadtime losses and attenuation in the gas, T_{60} is the transmission through the UF_6 of 60-keV gamma rays, and C is a calibration constant. The determination of enrichment I from Eq. (1) can be separated into two parts; the measurement of R , yielding a ^{235}U concentration in the sample chamber, and the measurement of T_{60} yielding the total uranium concentration in the sample chamber.

The data were first analyzed to demonstrate the accuracy of determining UF_6 densities, and consequently total uranium concentrations, from the transmission of 60-keV gamma rays. The UF_6 gas density determined from the natural logarithm of the 60-keV gamma-ray transmission was compared to the UF_6 gas density determined from the temperature and pressure measurements. The gas density was calculated from the temperature and pressure data using the equation of state

$$PV(1 + AP) = nKT \quad (2)$$

where P is the pressure in units of torr, T is the temperature in degrees kelvin, k is the Boltzman constant, n is the number of molecules, and A the second virial coefficient

$$A = 1.812 \times 10^3 T^{-3}. \text{ (Ref. 3)}$$

The UF_6 density calculated at 700 torr and 60°C using Eq. (2) is 3.7% greater than predicted by the ideal gas law. The normalized ratios of the density of UF_6 determined from the temperature and pressure measurements to

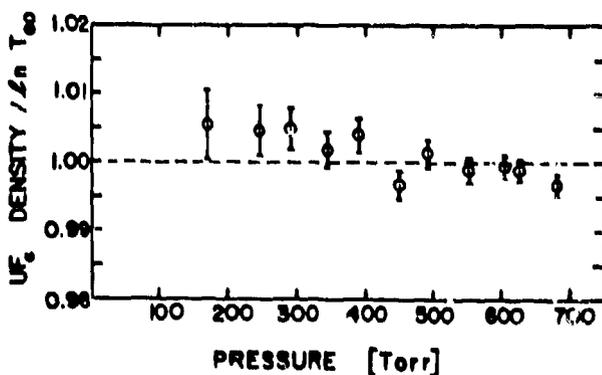


Fig. 2. Ratios of the UF_6 gas density determined from the temperature and pressure measurements to the natural logarithm of the 60-keV gamma-ray transmission. The values have been normalized to the weighted average of the ratios. The indicated pressures are for a gas temperature of 60°C.

the natural logarithm of the 60-keV gamma-ray transmission are presented in Fig. 2 for the pressure range of 176 to 680 torr. A similar plot was obtained for the data taken over the low-pressure range. From these data it was concluded that the relationship between the natural logarithm of the transmission and the gas density is linear within the accuracy of the temperature and pressure measurements over UF_6 pressures of 10 to 680 torr. The slight slope indicated by the data in Fig. 2 can be attributed to the uncertainty in the absolute temperatures and pressures.

To demonstrate that the enrichment can be measured by the described method, Eq. (1) was used to determine a relative enrichment from the data. The gamma-ray peak areas were corrected for deadtime losses using an electronic p lser method. The count rate of 186-keV gamma rays was corrected for attenuation losses in the sample using the expression $\ln T/(T - 1)$, where T is the transmission of 186-keV gamma rays through the UF_6 . T was determined from the measured transmission at 60 keV raised to the 0.21 power, which is the ratio of attenuation coefficients in UF_6 for 60- and 186-keV gamma rays. Although the attenuation-correction factor $\ln T/(T - 1)$ is not expected to be accurate for close-coupled geometries, it is adequate when the correction factor is small. For the UF_6 pressures measured, the attenuation correction factor for 186-keV gamma rays is less than 7%. Figure 3 shows the relative enrichments determined according to Eq. (1) normalized to the weighted average of the measured values. A separate normalization was calculated for the low- and high-pressure measurements. The enrichment values represent the results of 4000-s measurements of depleted (0.2% ^{235}U) UF_6 over the range of UF_6 densities plotted as the pressure at 60°C. The error bars represent the statistical precision of the measured values. The shaded area in Fig. 3 represents the limits of a $\pm 1\sigma$ relative standard deviation.

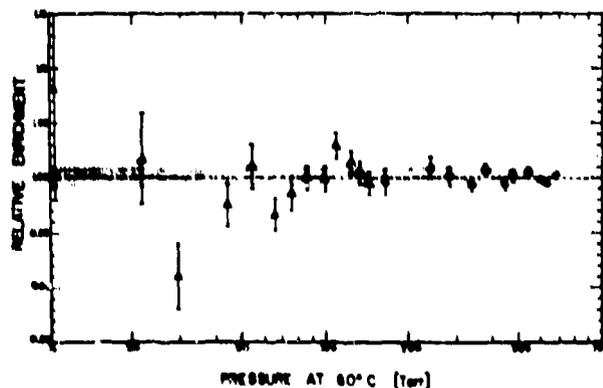


Fig. 3. Results of 4000-s measurements of the relative enrichment of depleted (0.2%) UF_6 . The low-pressure data (Δ) were measured at 27°C; the high-pressure data (\circ) were measured at 60°C. The error bars represent the statistical precision of the measured values. The UF_6 densities were converted to pressures at 60°C.

The gamma-ray background for these measurements comes from two primary sources, the buildup of ^{238}U daughter products in the sample chamber and the 1.3 kg of UF_6 in the feed bottle. The low-pressure data were measured after about 1 day of integrated exposure of the chamber at 100 torr. Consequently, essentially none of the background was associated with the buildup of ^{238}U daughter products in the sample chamber. The background under the 186-keV peak, most of which originated from the 1.3 kg of UF_6 in the feed bottle, resulted in a signal-to-background ratio of 0.9 at 100 torr. The high-pressure data were taken with a background equal to that associated with an equilibrium concentration of ^{238}U daughter products from 100 torr of depleted UF_6 . The background caused by an equilibrium concentration of ^{238}U daughter products was measured after a 100-day exposure to 100 torr UF_6 . The measured equilibrium value results in a signal-to-background ratio of about 0.5 for depleted UF_6 , dependent on the integration region of the gamma-ray peak but independent of the operating pressure.

The low-pressure data were used to normalize a calculation of the expected precision of a 1000-s enrichment measurement for pressures up to 10 000 torr at 155°C for natural and 3% enriched UF_6 . The effect of the background from an equilibrium concentration of ^{238}U daughter products and an external 1.3-kg UF_6 source was included in the calculation. The results of this calculation are presented in Fig. 4.

The plating of uranium on the chamber walls was investigated as a possible source of system-

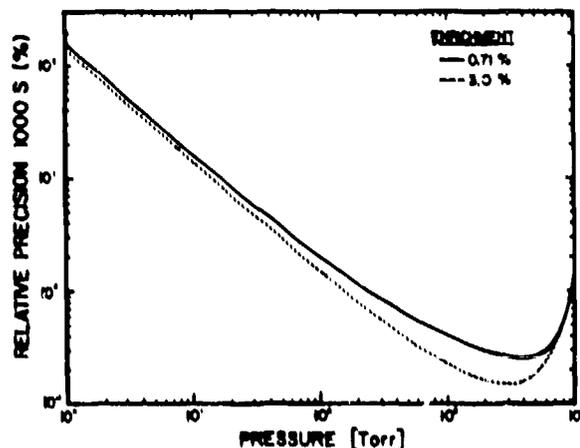


Fig. 1. Relative precision of a 1000-s enrichment measurement calculated for natural (solid curve) and 3% enriched (dashed curve) UF_6 as a function of pressure at 155°C. The gamma-ray background includes an estimate of the background associated with an equilibrium concentration of ^{238}U daughter products in the sample chamber and an external 1.3-kg UF_6 source.

atic error in the enrichment measurement. A measurement of uranium plating was performed after a 110-day integrated exposure of the stainless-steel chamber walls to 100 torr UF_6 at 20°C and a 3-day exposure to an average of 300 torr UF_6 at 60°C. The measurement was performed by evacuating the chamber and counting the 186-keV gamma rays from ^{235}U . The count rate of 186-keV gamma ray measured when the chamber was evacuated was $0.2 \pm 0.5\%$ of the count rate from 100 torr UF_6 . It was concluded that a negligible amount of plating occurs on stainless steel when kept in a clean vacuum environment.

4. Conclusions

A prototype NaI(Tl)-based enrichment monitor has been developed and tested under controlled laboratory conditions. The results of measurements on depleted UF_6 demonstrate that the method can be used to determine the enrichment of depleted to highly enriched UF_6 over a wide range of pressures. Within the statistical precision of the reported measurements, no systematic error in the relative enrichment as a function of pressure was observed. Calculations based on the reported data indicate that the enrichment can be determined for natural UF_6 with a relative precision of better than 1% for pressures ranging from 250 to 9000 torr at 155°C.

The ratio of UF_6 densities determined from temperature and pressure measurements to densities determined from gamma-ray transmission measurements was shown to be constant within the accuracy of the temperature and pressure measurements. The gamma-ray transmission method of measuring UF_6 densities offers a precise alternative to conventional density or pressure measurements.

Sufficient information has been obtained from the laboratory measurements that a detailed design of a prototype field system has been initiated. A field test and evaluation of a gas-phase UF_6 enrichment monitor in a production enrichment facility will provide further test information not available in the laboratory.

5. References

1. J. W. Tape, M. P. Baker, N. Strittmatter, M. Jain, and M. L. Evans, "Selected Nondestructive Assay Instrumentation for an International Safeguards System at Uranium Enrichment Plants," Proc. 20th Annual Mtg. Inst. of Nuclear Materials Management, Albuquerque, NM, July 1979.
2. T. D. Reilly, E. R. Martin, J. L. Parker, L. G. Speir, and R. B. Walton, "A Continuous In-Line Monitor for UF_6 Enrichment," Nucl. Technology 23, 318 (1974).
3. S. Villani, Ed., Uranium Enrichment (Springer-Verlag, New York, 1979), p. 123.

Figures

1. NaI(Tl)-based gas-phase UF_6 enrichment monitor. The intensity of 186-keV gamma rays emitted by ^{235}U and the transmission through the UF_6 gas of 60-keV gamma rays from an external ^{241}Am source were measured and the data were used to determine the ^{235}U enrichment.
2. Ratios of the UF_6 gas density determined from the temperature and pressure measurements to the natural logarithm of the 60-keV gamma-ray transmission. The values have been normalized to the weighted average of the ratios. The indicated pressures are for a gas temperature of 60°C .
3. Results of 4000-s measurements of the relative enrichment of depleted (0.2%) UF_6 . The low-pressure data (Δ) were measured at 27°C ; the high-pressure data (O) were measured at 60°C . The error bars represent the statistical precision of the measured values. The UF_6 densities were converted to pressures at 60°C .
4. Relative precision of a 1000-s enrichment measurement calculated for natural (solid curve) and 3% enriched (dashed curve) UF_6 as a function of pressure at 155°C . The gamma-ray background includes an estimate of the background associated with an equilibrium concentration of ^{238}U daughter products in the sample chamber and an external 1.3-kg UF_6 source.