284
24-81
LA-8657-MS

(2)

01. 2260
R-1716

**Conceptual Design for the Field Test and Evaluation
of the Gas-Phase UF₆ Enrichment Meter**

MASTER

University of California

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

CONCEPTUAL DESIGN FOR THE FIELD TEST AND EVALUATION
OF THE GAS-PHASE UF_6 ENRICHMENT METER

by

R. B. Strittmatter, J. N. Leavitt, and R. W. Slice

ABSTRACT

An in-line enrichment monitor is being developed to provide real-time enrichment data for the gas-phase UF_6 feed stream of an enrichment plant. Data from proof-of-principle measurements using a laboratory prototype system are presented. A conceptual design for an enrichment monitor to be field tested and evaluated at the Oak Ridge Gaseous Diffusion Plant is reported.

I. INTRODUCTION

The Los Alamos Scientific Laboratory (LASL), Safeguards Instrumentation Research and Development Group Q-1, as a part of the U. S. Enrichment Plant Safeguards Program, is developing an in-line gas-phase UF_6 enrichment monitor. The monitor is being developed to provide the technology to implement effective real-time material balance accounting at the feed and withdrawal area of uranium enrichment plants.

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 UF_6 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 technology which is being developed to measure UF_6 enrichment in the gas-phase will be applicable to international safeguards as well as process control of enrichment plants. The Union Carbide Corporation, Nuclear Division, through the Oak Ridge Gaseous Diffusion Plant (ORGDP) operations office and the Oak Ridge Enrichment Safeguards Program office have agreed to participate with LASL in a field test and evaluation of the gas-phase enrichment monitor.

II. ASSAY TECHNIQUE

A. Description of the Measurement Technique

The gas-phase enrichment is determined by combining measurements of the ^{235}U and total uranium concentrations in the UF_6 gas. The ^{235}U concentration is determined from a measurement of 186-keV gamma rays emitted from ^{235}U . The total uranium concentration is determined by measuring the transmission through the UF_6 gas of 60-keV gamma rays from an external ^{241}Am source. Figure 1 is a schematic of a prototype system showing the orientation of the NaI(Tl) detector, the sample chamber, and the location of the ^{241}Am transmission source.

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

$$I = R/C \ln T_{60} \quad , \quad (1)$$

where R is corrected for dead-time 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 the enrichment I from Eq. (1) is composed of two parts: the measurement of R , yielding the ^{235}U concentration in the sample chamber, and the measurement of T_{60} yielding the total uranium concentration in the sample chamber. Since the measurement accounts for variations in UF_6 density the measured assay is independent of the UF_6 pressure.

B. Proof-of-Principle Measurements

To investigate the physical principles upon which the enrichment monitor is based, a prototype instrument was tested under controlled laboratory conditions.³ The laboratory enrichment monitor consists of a UF_6 gas-handling system, sample chamber, NaI(Tl) detector, a ^{241}Am transmission source, nuclear electronics, pressure and temperature instrumentation, and a data acquisition system. Two sets of measurements were 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 stainless-steel cylindrical sample chamber used for both low- and high-pressure measurements is 14.6-cm in 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 employed a NaI light pipe between the NaI(Tl) and the photomultiplier. The low-pressure measurements were performed with a 2.54-cm-thick detector and the high-pressure measurements with a 2-cm thick detector.

Data were obtained to demonstrate the accuracy of determining UF_6 densities by the transmission method. The transmission of gamma rays through matter is represented by an exponential function $e^{-\mu\rho x}$ where μ is the mass absorption coefficient for the material being traversed, ρ is the density of the material, and x is the thickness of the material. Since μ is a constant for a particular material and gamma-ray energy, and x is held constant by the sample chamber dimensions, the UF_6 density is proportional to the natural logarithm of the 60-keV gamma-ray transmission.

Using the laboratory prototype enrichment monitor 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 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

$$A = 1.812 \times 10^3 T^{-3}. (\text{Ref. 4}) \quad (3)$$

The UF_6 density calculated at 760 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 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 a low-pressure range of 10 to 150 torr. 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 pulser 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 correction factor attenuation is less than 7%. Figure 3 shows the relative enrichments determined according to Eq. (1) normalized to the weighted average of the measured values. 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 shaded area represents the limits of a $\pm 1\%$ relative standard deviation.

The gamma-ray background for these measurements comes from two primary sources, the build up of ^{238}U daughter products in the sample chamber, and 1.3 kg of UF_6 in the sample 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 .

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 systematic 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 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 rays 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.

There are various sources of background radiation which can affect the precision and accuracy of the measurement technique. These sources include

1) high-energy gamma rays from the daughter products of ^{238}U , 2) extraneous 186-keV gamma rays from ^{235}U not in the gas-phase, 3) 208-keV gamma rays from the ^{241}Am source, and 4) 60-keV gamma rays from U daughter products. The first three sources affect the determination of the valid 186-keV peak area while sources 1 and 4 affect the 60-keV peak area determination. Measurements are on-going to develop methods to minimize these effects.

III. FIELD TEST AND EVALUATION

An agreement has been reached between LASL and ORGDP to install a gas-phase enrichment monitor in the Paducah product feed station of the ORGDP. At this station UF_6 from the Paducah Gaseous Diffusion plant, having enrichments ranging from 0.9 to 2.0%, is fed into the Oak Ridge plant for the production of UF_6 assays in the 2.5 to 4.0% range. The Paducah product feed station provides a site at which a range of assays from 0.9 to 2.0% can be measured and compared to accountancy data obtained from mass spectrometric analysis.

A schematic of the feed station is presented in Fig. 5. Uranium hexafluoride is removed from 10-ton feed cylinders by vaporization transfer. A feed pressure at the feed cylinder of 75 psia is maintained by placing the cylinder in a steam heated autoclave at 107°C . The UF_6 is transported via 3 inch schedule 10S steam traced Monel pipe to the valve labeled PV 3056. At this valve the pressure is reduced to 10 to 16 psia. Immediately downstream of valve PV 3056, a bypass will be installed to accommodate the gas-phase monitor. Although the pressure can range from 10 to 16 psia the short-term pressure fluctuations are typically maintained to within $\pm 5\%$ of the average pressure. Flow rates in the 3-inch-diameter pipe vary from 450- to 1000- lbs UF_6/hr .

The system of one-inch diameter pipe indicated in Fig. 5 originates from an autoclave No. 2. This one-inch line is used to empty near-empty cylinders and does not represent a major flow of material. The UF_6 , which originates from autoclave No. 2, will not pass through the enrichment monitor and, consequently, will not affect the comparison between the assay measured by the enrichment monitor and the accountancy data for the UF_6 cylinder of autoclave No. 1.

Site conditions which must be considered in the detailed design of the instrument include:

- 1) The ambient temperature in the feed area ranges from 13 to 35°C with daily fluctuations of 10°C in the summer and 5°C in the winter.
- 2) Relative humidity in the area reaches 100%.
- 3) Available electric power is not of high quality and will require conditioning.
- 4) The radiation background, consisting primarily of high-energy gamma rays from ^{238}U daughter products in the feed cylinders was measured at the installation area. The results indicate that the background originating from the feed cylinders was less than the contribution from the ^{238}U daughter products in the feed pipe itself.

Process specifications and design criteria which affect the design of the instrument include the following:

- 1) All gas-handling system components are to be designed to withstand a pressure of 200 psia.
- 2) All connections in the gas-handling system are to be welded. Consequently the material of construction must be compatible with welding to Monel.
- 3) The gas handling system is to be heat traced at a nominal temperature of 93°C.

IV. CONCEPTUAL DESIGN OF ASSAY SYSTEM

A. System Design

The primary design goal for the field test and evaluation system is to provide the necessary data required to evaluate the described assay method, and to do so in a manner which is safe and unobtrusive to the diffusion plant operations. Secondary design goals are to include features appropriate for application of the device to IAEA safeguards. These features include ruggedness, compactness, minimization of operator intervention, the ability to perform system verification, tamper indication and data protection. A moderate effort will be made to incorporate the secondary design goals into the system when practical.

A schematic of the system configuration is shown in Fig. 6. The major components include the bypass plumbing, the evacuation and sampling station, the mechanical portion of the assay device and the system electronics.

B. Bypass Plumbing

The bypass plumbing provides a means by which a normal UF_6 flow can be delivered to the diffusion plant if for any reason flow through the measurement chamber is not possible. Flow through the chamber will be shut off for calibration measurements, during which the chamber is evacuated and a UF_6 standard is introduced into the chamber. The double valves shown in the bypass plumbing schematic are required for safety reasons if the feed station is operated while the sample chamber is not installed. Bypass plumbing will consist of heat traced 3 inch schedule 10S Monel pipe. All valves and plumbing will satisfy the Union Carbide Corporation Technical Specification P-1.1 subsection 137.

C. Evacuation and Sampling Station

The evacuation and sampling station will provide the flexibility to perform calibration and background measurements necessary for a thorough test and evaluation. Experience gained during the test and evaluation may indicate that the evacuation and/or sampling stations are not necessary for future instrument designs. The evacuation station will be based on a mechanical vacuum pump and chemical trap.

The amount of UF_6 which is involved in the system evacuation depends on the system volume, pressure and frequency of evacuation. The density of UF_6 at 16 psia is approximately 15 g/l. Assuming a 5-liter volume for the sample chamber and piping between valves, 75 g UF_6 will be removed per evacuation. A reasonable estimate for the maximum frequency of evacuation is once per month during the early stages of testing. This represents a total of 900 g UF_6 per year. If the feed system pressure is reduced before evacuation, the amount of UF_6 trapped would be reduced in proportion to the pressure.

D. Mechanical Design of the Assay Device

The conceptual design of the mechanical portion of the assay device is presented in Fig. 7. All major components of the design are included in the drawing, however, actual dimensions have not been finalized. The instrument

consists of a sample chamber, a NaI(Tl) detector, a ^{241}Am source, a motorized wheel for verification and secondary calibration measurements, gamma-ray shielding, heat tracing, insulation and support structure.

The cylindrical sample chamber is constructed of Monel. It defines the dimensions of the UF_6 volume that is measured. The height and radius will be determined by an optimization of the measurement precision and accuracy. The estimated dimensions are 10 to 15 cm in height and 13 cm in diameter. The top and bottom chamber walls will have windows milled thinner than the rest of the walls to reduce the attenuation by the walls of the gamma rays to be measured. The window thickness and area are to be designed to satisfy the 200 psia design pressure specification. Preliminary calculations and hydraulic pressure test measurements indicate it will not be difficult to achieve a suitable conservative window design.

The NaI(Tl) detector will be a standard 12.7-cm diameter by approximately 2-cm thick crystal with a NaI light pipe. The crystal will be coupled to a 12.7-cm diameter photomultiplier.

The ^{241}Am source used to provide 60-keV gamma rays for the transmission measurement will be a commercially available source. The source strength will be in the 5 mCi range. Americium-241 has a 432-year half life and consequently will not require replacement after the installation. The primary radiations from ^{241}Am are alpha-particles and 60-keV gamma rays so that only a minimal amount of shielding will be required to satisfy health physics requirements.

The motorized wheel will be used to provide system-verification and secondary-calibration data. The wheel will have four positions. In the normal operating mode a hole in the wheel provides an unobstructed path for the ^{241}Am 60-keV gamma rays. The three other positions provide the verification and calibration data. In position two a lead disk is placed between the ^{241}Am source and the detector to allow the measurement of the background under the 60-keV peak. For measurements in the third and fourth position the sample chamber will be evacuated. The third position will contain a disk of low-atomic-number material with a transmission for 60-keV gamma rays similar to that of the normal operating UF_6 pressure. This disk will provide data to check the UF_6 density measurement. The fourth position will contain a small ^{235}U sample to provide a measurement of the 186-keV gamma ray intensity from a standard source. The four wheel positions should provide the data required to verify the performance of the assay device.

The system shielding is to be designed to reduce the background from Compton scattered high-energy gamma rays from ^{238}U daughter products external to the measurement chamber. An expected source of background is the feed cylinders in the feed area. Background measurements at the planned installation area indicate that a 2.54-cm thick Pb shield will be adequate for this purpose. Heat tracing, thermal insulation and support structure will be provided but are not discussed in this document.

E. Electronics Design

The electronics system for detector operation, signal processing, data acquisition, data manipulation and system control is outlined in the Figure 8 block diagram. The electronics design is based on the NIM standard using a combination of commercial and custom designed components. For ease of maintenance commercial NIM modules will be used when practical. To satisfy the secondary design requirements of compactness, minimization of operator intervention, tamper indication and data protection the data acquisition, data manipulation and system control operations will be provided by a LASL designed electronics unit. The components outlined by the dashed line in Fig. 8 comprise the LASL electronics.

Processed detector signals will be converted and stored by a 1024-channel 20-bit ADC and memory. The data will be manipulated by a set of algorithms preprogrammed in ROM utilizing an arithmetic chip. A calendar clock will provide the necessary timing functions for data acquisitions. The resulting assay value will be output via a LED display and a serial output device as protected data. Data protection will be provided by means of the standard NBS encryption algorithm. Communication to the device will be limited to system verification and calibration operations and will be implemented through a 20-key pad. The key pad will be provided with physical and/or software protection. Since the need for spectral display is limited to system verification and calibration operations, the display device will be provided as a separate unit. The serial output line will carry the information to be recorded on a separate mass storage device for archival reference. The real-time assay result will also be displayed at a remote location for use by the diffusion plant process operations personnel.

References

1. J. W. Tape, M. P. Baker, R. Strittmatter, M. Jain, and M. L. Evans, "Selected Nondestructive Assay Instruments for an International Safeguards System at Uranium Enrichment Plants," Proc. 20th Annual Mtg. Inst. of Nuclear Materials Management, Albuquerque, N.M., 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. R. B. Strittmatter and J. W. Tape, "A Gas-Phase UF_6 Enrichment Monitor for Enrichment Plant Safeguards," Proc. 2nd Annual Symposium on Safeguards and Nuclear Material Management, Edinburgh, Scotland, March 1980.
4. S. Villani, Ed., Uranium Enrichment (Springer-Verlag, New York, 1979), p. 123.

Figure 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.

Figure 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.

Figure 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 .

Figure 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.

Figure 5. Schematic of the Paducah product feed station at ORGDP, and the proposed location of the enrichment monitor. The normal flow of material will be through the enrichment monitor. The one-inch line from autoclave No. 2 is used to empty near-empty UF_6 cylinders.

Figure 6. Pictorial of the conceptual design of the enrichment monitor system to be installed at the Paducah product feed station of the ORGDP.

Figure 7. Conceptual design of the mechanical portion of the enrichment monitor. The assay instrument consists of a sample chamber, NaI(Tl) detector, a ^{241}Am source, a motorized wheel for verification and secondary calibration measurements, gamma-ray shielding, heat tracing, insulation and support structure.

Figure 8. The block diagram of the LASL designed data-acquisition system for the gas-phase UF_6 enrichment monitor. The system, designed to fit the NIM standard, will provide all data acquisition, control, and computational functions necessary to produce a UF_6 assay result.