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IAEA Verification Experiment at the Portsmouth Gaseous Diffusion Plant: Report on the Cascade Header Enrichment Monitor

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I. Abstract

We describe the Cascade Header Enrichment Monitor (CHEM) for the Portsmouth Gaseous Diffusion Plant at Piketon, Ohio, and present the calibration and measurement results. The United States government has offered excess fissile material that is no longer needed for defense purposes for International Atomic Energy Agency (IAEA) inspection. Measurement results provided by the CHEM were used by the IAEA in a verification experiment to provide confidence that the US successfully blended excess highly enriched uranium (HEU) down to low enriched uranium (LEU). The CHEM measured the uranium enrichment in two cascade header pipes, a 20.32-cm HEU pipe and a 7.62-cm product LEU pipe. The CHEM determines the amount of ^{235}U from the 185.7-keV gamma-ray photopeak and the amount of total uranium by x-ray fluorescence (XRF) of the 98.4-keV x-ray from uranium with a ^{57}Co XRF source. The ratio yields the enrichment. The CHEM consists of a collimator assembly, an electromechanically cooled germanium detector, and a rack-mounted personal computer running commercial and custom software. The CHEM was installed in December 1997 and was used by the IAEA inspectors for announced and unannounced inspections on the HEU and LEU header pipes through October 1998. The equipment was sealed with tamper-indicating enclosures when the inspectors were not present.

II. Introduction

In September 1993, President Clinton issued Presidential Decision Directive 13, which offered excess fissile material no longer needed for national security for International Atomic Energy Agency (IAEA) inspections. Between December 12, 1997, and October 8, 1998, Los Alamos and IAEA personnel conducted a verification experiment at the Portsmouth Gaseous Diffusion Plant (PGDP) at Piketon, OH. The Cascade Header Enrichment Monitor (CHEM) was installed by the Los Alamos National Laboratory as part of this experiment. The CHEM is a nondestructive assay (NDA) instrument that measures the uranium enrichment of gaseous UF_6 in cascade header pipes. At the PGDP, the CHEM was installed to verify the enrichment of highly enriched uranium (HEU) in a 7.62-cm (3-in.) section of header pipe and of blended low-enriched uranium (LEU) product in a 20.32-cm (8-in.) section of header pipe. The IAEA inspectors required a method to verify that the uranium enrichment of the UF_6 gas in cascade header pipes is consistent with the declaration of the facility: the LEU blended product enrichment is less than 20% and the HEU feed material is greater than 20%. During this period the IAEA personnel made several short notice random inspections (SNRIs) of the PGDP blending operation.

A brief description of the operation of the CHEM, the particular modifications required for the PGDP, and the CHEM verification data are given. The operating principles and development of the CHEM have been described in previous papers [1-4]. More details on the operation of the CHEM as implemented at the PGDP can be found in the CHEM operations manual [5].

III. Theory

The enrichment of the UF_6 gas is the ratio of the total amount of ^{235}U in the gas to the amount of total uranium in the gas. The CHEM uses a high-purity germanium (HPGe) detector to obtain a gamma-ray spectrum from the gaseous UF_6 in the header pipe. The spectrum contains photopeaks whose areas are proportional to these two quantities. In this application, because of the low pressure of the gas (the HEU line is approximately 10 Torr), the transmission method [1] is not appropriate.

The total amount of ^{235}U in the gas is proportional to the ^{235}U 185.7-keV gamma-ray count rate from the gas. The amount of total uranium in the gas is proportional to the uranium K_{α_1} 98.4-keV x-ray count rate from the gas. The K_{α_1} x-ray emission is induced by x-ray fluorescence (XRF) of the gaseous UF_6 using a ^{57}Co source. This source was chosen because the 122-keV gamma ray from ^{57}Co is just above the 115.591-keV K-absorption edge for uranium electrons. A measurement of these two quantities on a header pipe containing UF_6 gas of a known enrichment enables calculation of the proportionality constant. The measurement procedure results in an enrichment measurement that is independent of the UF_6 gas pressure in the header pipe. However, the detectability limit is approximately 1 Torr in a 30-min counting time.

A complication to this procedure is the formation of uranium deposits on the pipe walls. As will be discussed below, the detector and source collimation and their relative angle eliminate the contribution from fluorescencing the deposit. However, the pipe deposit within the detector collimation solid angle contributes to the 185.7-keV gamma-ray count rate. One way to account for the deposit is to measure the count rate while no gas is present in the pipe, and then to subtract it from the total 185.7-keV gamma-ray count rate. While this is not always possible in practice, it was possible for this work. Another method, the two-geometry technique, has been developed to measure the enrichment of UF_6 gas in a header pipe in the presence of deposits, without the need for a separate deposit measurement [2-4]. If the deposits had increased significantly during the blending operation, then the deposits would have been remeasured or the two-geometry technique would have been implemented.

The enrichment of the UF_6 process gas in the header pipe is calculated using the expression

$$E = K\frac{R_{186(Total)} - R_{186(BG)}}{R_{XRF}}, \tag{1}$$

where

E = ^{235}U enrichment (in weight %) of the process gas,
$R_{186(Total)}$ = ^{235}U 185.7-keV total count rate (c/s) (gas + deposit + room background),
$R_{186(BG)}$ = ^{235}U 185.7-keV background count rate (c/s) (deposit + room background),
R_{XRF} = uranium 98.4-keV K_{α_1} x-ray count rate (c/s/mCi) induced by ^{57}Co XRF source, and
K = calibration constant.

The numerator is a measure of the total amount of ^{235}U in the UF_6 gas, and the denominator is a measure of the amount of total uranium in the gas.

The uncertainty in the enrichment is calculated using the expression

$$\Delta E = \sqrt{E^2\left[\left(\frac{\Delta R_{XRF}}{R_{XRF}}\right)^2 + \left(\frac{\Delta K}{K}\right)^2\right] + \left(\frac{K}{R_{XRF}}\right)^2\left[\left(\Delta R_{186(Total)}\right)^2 + \left(\Delta R_{186(BG)}\right)^2\right]}, \tag{2}$$

where

ΔE = uncertainty in the enrichment E,

$\Delta R_{186(Total)}$ = uncertainty in the ^{235}U count rate $R_{186(Total)}$,

$\Delta R_{186(BG)}$ = uncertainty in the background ^{235}U count rate $R_{186(BG)}$,

ΔR_{XRF} = uncertainty in the XRF count rate R_{XRF}, and

ΔK = uncertainty in the calibration constant K.

IV. Hardware Description

The CHEM consists of an HPGe detector, a ^{57}Co XRF source, a collimator assembly to collimate and hold the detector and source rigidly to the exterior of the monel header pipe, a multichannel analyzer (MCA), a computer, and data acquisition software. These system components are described below.

Detector

The CHEM uses an HPGe detector from Princeton Gamma Tech (PGT) (see Figure 1) to detect the 98.4-keV x-ray and the 185.7-keV gamma ray. The detector is electromechanically cooled by a Cryotiger compressor from APD Cryogenics, Inc. Due to the microphonics present on the 7.62- and 20.32-cm cascade header pipes, an anti-vibration mount from PGT was incorporated. The detector has an active area of 2000 mm^2 and a thickness of 10 mm. The energy resolution measured by PGT in December 1997 was 691 eV full-width at half maximum (FWHM) at 122 keV. When not in use, the detector and compressor were placed on a specially designed storage cart (see Figure 2).

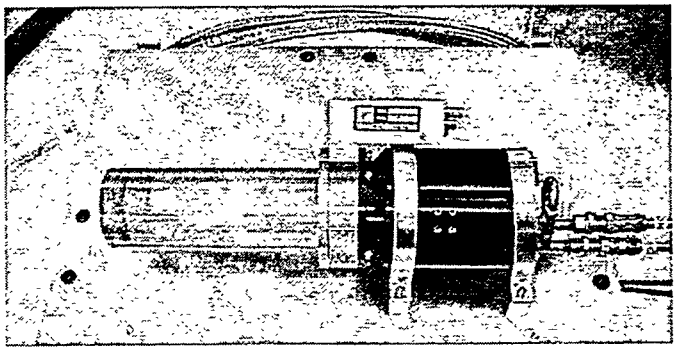


Figure 1. The HPGe detector.

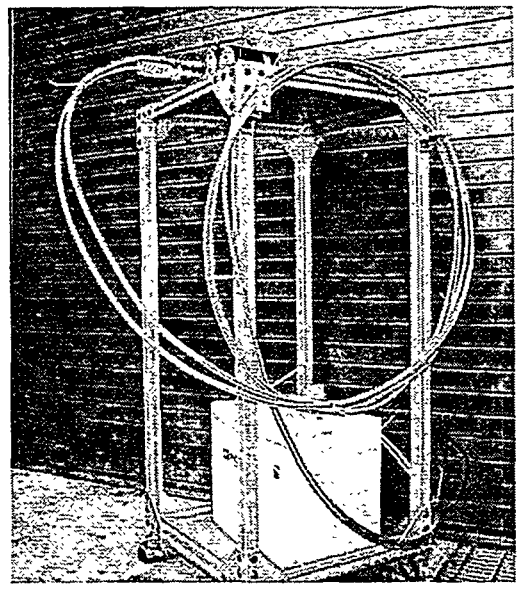


Figure 2. The CHEM storage cart with the detector on top and compressor on the bottom.

At the PGDP, the detector was also exposed to a high-temperature environment. Temperatures of the cascade header pipes, to which the CHEM was attached, in the diffusion cell can exceed 85 °C. The high temperature caused outgasing of the aluminum end cap on the detector, degrading the vacuum in the detector cryostat and the energy resolution of the detector. The detector was returned to the manufacturer once for repair and once for repair and modification in efforts to improve the energy resolution in the high-temperature environment. The modification added insulation and changed the end cap to stainless steel, which outgases less than aluminum. In addition, an external vac-ion pump was connected to the detector to further help maintain the vacuum in the cryostat.

The first repair of the detector in March 1998 was not sufficient for it to maintain good energy resolution while attached to the header pipe. It was sent for a second repair and the modification in May 1998. The detector was returned to the PGDP in July 1998, and was available for use on July 11, 1998. As a result of the modification, there are two calibration constants for the CHEM, one for December 1998–May 1998 and one for July 11, 1998–October 8, 1998. The calibration discussed here uses a subset of the measurements whose selection is based on their statistical quality and on characteristics of the plant enrichment data taken during those measurements. This selection is described below (Section VI).

However, after the detector repair and modification, the detector still did not perform to the resolution desired even though the cryostat vacuum was adequate, and was returned to the manufacturer for further work. These problems affected the number of usable measurements taken by the CHEM during the experiment and also the availability of the instrument during some of the IAEA inspections.

Collimator Assembly

Two collimator assemblies were built, one for the 7.62-cm HEU pipe and one for the 20.32-cm LEU pipe. The collimator assembly for the larger header pipe is shown schematically in Figure 3. The collimator assembly for the smaller pipe is similar. This diagram shows the intersecting solid angles of the HPGe detector and the ^{57}Co XRF source that excludes the detection of the fluoresced x-rays from the deposit.

The detector is inserted into a cylindrical tungsten collimator, and views the center of the pipe through the rectangular aperture of the collimator. The dimensions of the detector collimator for both assemblies are 1.27 cm (W) x 7.30 cm (D) x 3.81 cm (H). The rectangular collimator aperture is oriented so the long axis of the collimator is parallel to the long axis of the header pipe. It can also be rotated 90° for the two-geometry technique. A thin Teflon sleeve is used between the tungsten collimator and the detector to ease insertion and removal of the detector (see Figure 4).

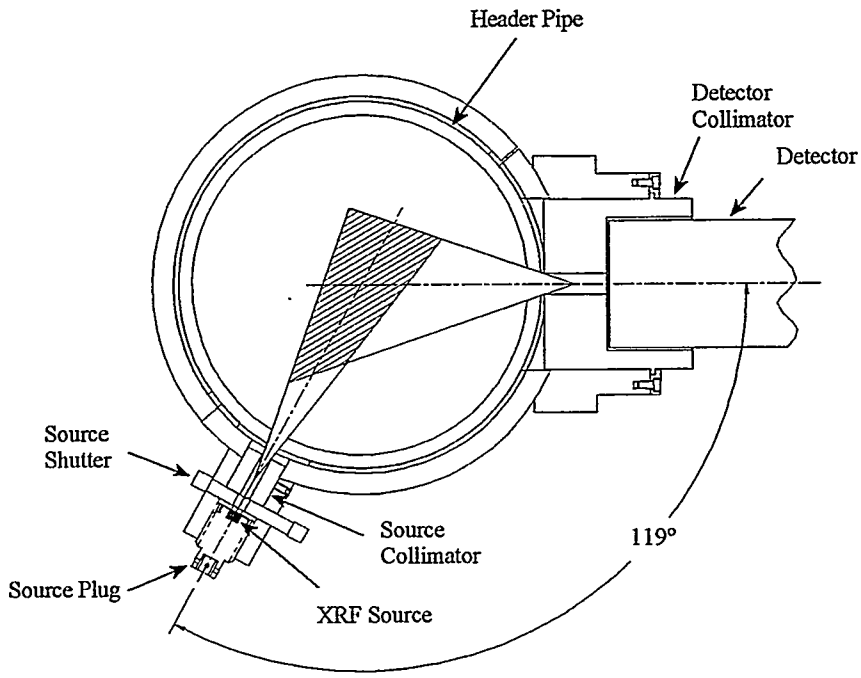


Figure 3. CHEM collimator assembly for the 20.32-cm pipe.

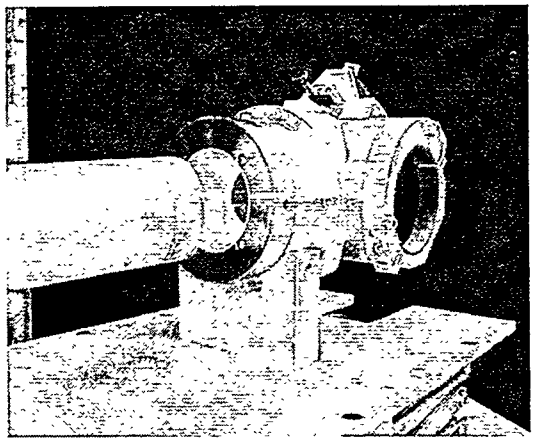


Figure 4. The 7.62-cm collimator assembly, Teflon insert, and detector.

The source collimator is also made of tungsten, and directs the source perpendicular to the central pipe axis. The rectangular aperture of the source collimator for both assemblies is 0.635 cm (W) x 5.03 cm (H). It is fan-shaped, with the aperture 3.81 cm from the source.

It is necessary to create a fluorescencing volume inside the header pipe which excludes the pipe walls, and therefore any deposits. In order to maximize this volume, the detector and source axes are angled with respect to each other, and each of the collimator dimensions are optimized. The height, width, and depth of the detector collimators are strongly influenced by the detector and pipe diameters. Figure 3 shows this interrogated volume as a cross-hatched region. The angle between the detector and source axes was chosen to be 119°, which is optimized for the maximum signal-to-noise ratio considering the Compton scattering effect. For example, with a relative detector-source angle of 119°, the collimators and pipe walls produce a Compton scattering peak at about 110 keV. Changing the angle to 90° would move this peak to 100 keV, thereby interfering with the XRF peak at 98.4 keV.

The detector and source collimator housings are attached to a rigid aluminum structure which surrounds half of the monel header pipe. Semicircular clamps placed around the other half of the pipe secure the collimator assembly to the pipe. A lead sheet is placed opposite the detector and behind the header pipe to shield the detector from nearby pipes. A tungsten shutter is used to block the XRF source when the CHEM is not in use.

Figures 5, 6, and 7 show additional details of the CHEM collimator assembly. Figure 5 shows the collimator assembly on the 7.62-cm header pipe. The detector collimator slit is shown in the parallel position, i.e., the long axis of the collimator slit is parallel to the long axis of the header pipe. This is the position of the detector collimator for all the measurements at the PGDP for the IAEA verification experiment.

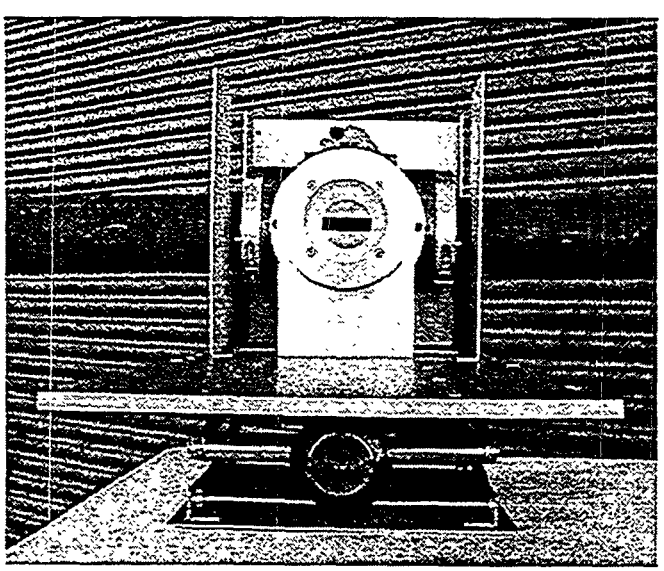


Figure 5. The 7.62-cm collimator assembly in parallel orientation.

The operation of the XRF shutter is shown in Figure 6. The two thumb screws are used to drive the tungsten shutter bar from the open position (the "O" is visible) to the closed position (the "X" is visible). The shutter is in the "O" position for all the measurements. At the conclusion of the measurement, the shutter is moved to the "X" position and left in the "X" position until the next measurement. The complete tamper indicating enclosure (TIE) for the 20.32-cm header pipe is shown in Figure 7. The TIE remains installed at all times on both collimator assemblies between inspection visits by the IAEA.

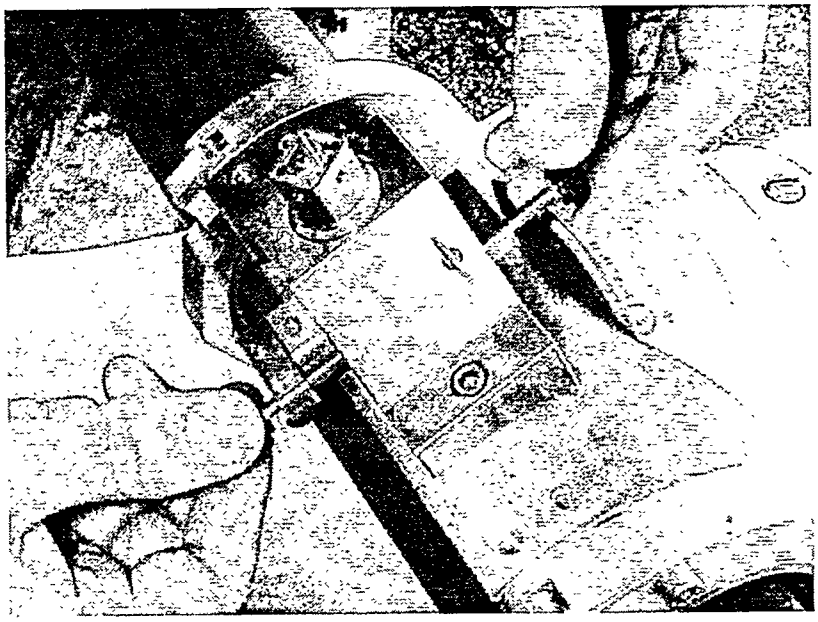


Figure 6. The XRF source collimator, showing thumb screws used to open and close the shutter.

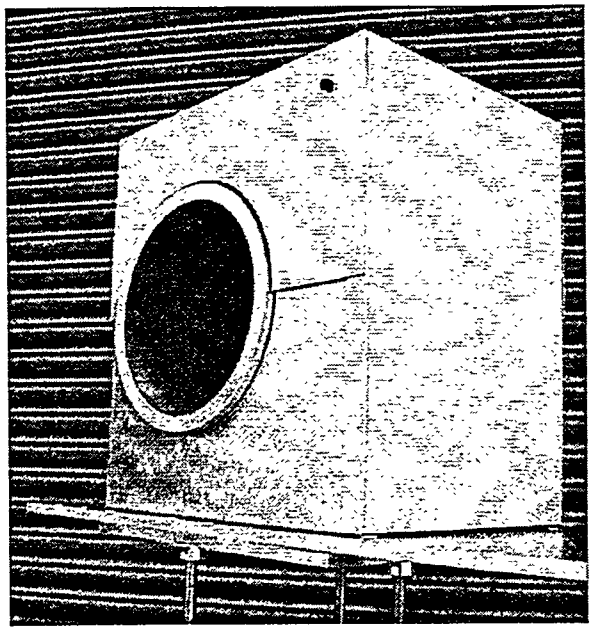


Figure 7. The complete TIE for the 20.32-cm header pipe.

Source

Two ^{57}Co sources ($t_{1/2}$ = 271.8 days) were used, one for the HEU header pipe, and one for the LEU header pipe. The sources were nominally 40 mCi on the manufacture date and are sealed with a 0.25-mm beryllium window. The source is held in the end of a tungsten source plug by a small aluminum cap (see Figure 8). The threaded source plug screws into the source collimator. For the time period of this verification experiment, no source change was required. A TIE and tamper indicating devices are installed on the source and collimator when they are not in use. Figure 8 shows part of the TIE installed on the 7.62-cm collimator assembly.

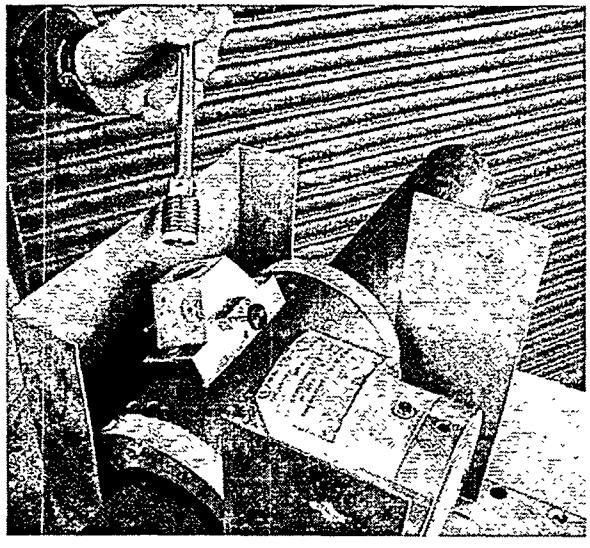


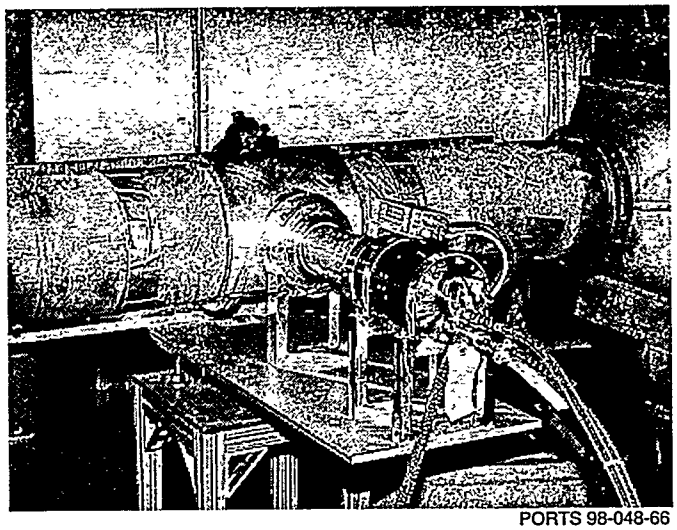
Figure 8. The ^{57}Co source holder and collimator, shown encased by the TIE.

Multichannel Analyzer

The MCA is a Canberra InSpector. It is set to use a 4-μs shaping time constant and 2,048 channels.

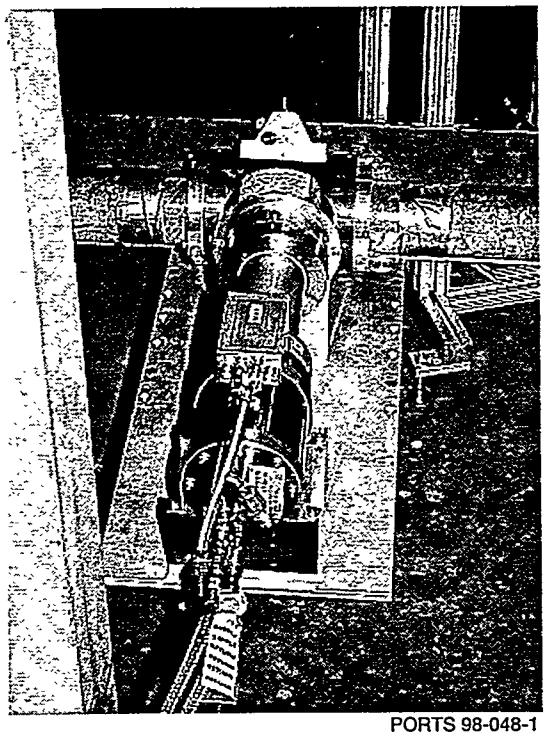
Installation

Figure 9 shows a photograph of the 20.32-cm header pipe collimator assembly and the detector installed on the LEU cascade header pipe at the PGDP. Figure 10 shows a photograph of the 7.62-cm header pipe collimator assembly and the detector installed on the HEU cascade header pipe at the PGDP.



PORTS 98-048-66

Figure 9. The detector and 20.32-cm collimator installed on the LEU header pipe.



PORTS 98-048-1

Figure 10. The detector and 7.62-cm collimator installed on the HEU header pipe.

V. Computer and Software Description

The CHEM uses a rack-mounted, Pentium-based computer for control of the Canberra InSpector, data acquisition software, and data analysis. A custom software user interface has been written for energy calibration and for enrichment measurements and calculations on the two header pipes. The program is called **XRF**, which stands for "x-ray fluorescence." This interface accesses Canberra's Genie-PC spectroscopy software to control data acquisition and to extract peak locations and areas

from the spectra. The program then performs the enrichment calculations as discussed in Section III. The CHEM software operation was tested for operation in the year 2000, and no problems were found. Virus protection software was installed on the computer at the request of the PGDP.

This section gives a brief overview of the software operation, including the energy calibration which must be performed on the CHEM prior to enrichment measurements. For more detailed information, see the CHEM operations manual [5].

Software Startup

Once the computer has booted up, the **GeniePC** program group icon is visible on the screen. Double-clicking this icon will make the **GENIE-PC – Icon View** window active (see Figure 11). The three icons of interest in the group are the **XRF Enrichment Monitor**, **Virtual Data Manager (VDM)**, and **Spectroscopy Assistant** icons. Normally, only the **XRF Enrichment Monitor** icon is needed.

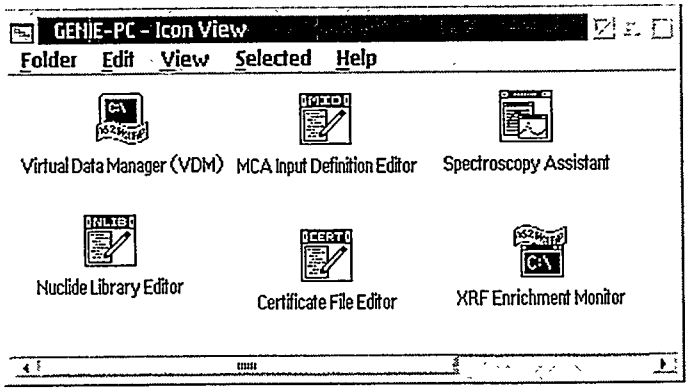


Figure 11. "GENIE-PC – Icon View" window.

To start the **XRF** program, the user must double-click the **XRF Enrichment Monitor** icon. This will open the **XRF** Main Menu (see Figure 12). The energy calibration and measurement for each of the two diameter header pipes are initiated from this menu. The energy calibration has been improved and simplified since the publication of the CHEM operations manual [5]. The changes are described briefly below, along with a brief description of the enrichment measurement procedure.

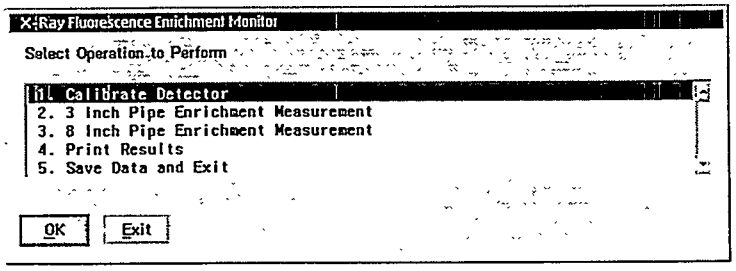


Figure 12. "XRF" Software main menu.

CHEM Energy Calibration

The first item in the **XRF** main menu is **Calibrate Detector**. Selecting this item will present a message box asking the user to input an acquisition time. The default value is 10 min. After the time

is entered and accepted, a spectrum window opens to display the gamma-ray spectrum from the InSpector. Data acquisition then starts automatically, and the accumulating incoming data are visible.

At the end of the entered acquisition time, the data acquisition will stop automatically. The spectrum will disappear briefly, then reappear with a message box just above the spectrum window. The message box asks the user to place the cursor on the 59.3-keV photopeak. (This is the tungsten K_{α_1} x-ray from the tungsten collimator, fluoresced by the ^{57}Co XRF source.) The user then places the cursor on this peak and selects **OK** in the message box. The user then repeats this for the second energy calibration photopeak at 185.7 keV. Figure 13 shows a CHEM energy spectrum as displayed by the Genie-PC software with the two energy calibration photopeaks labeled. A message box then indicates whether the energy calibration was successful, and a second box asks if the new calibration is satisfactory. If the calibration is satisfactory, the user selects **Yes** and is returned to the main menu for the enrichment measurements. If necessary, the energy calibration may be repeated.

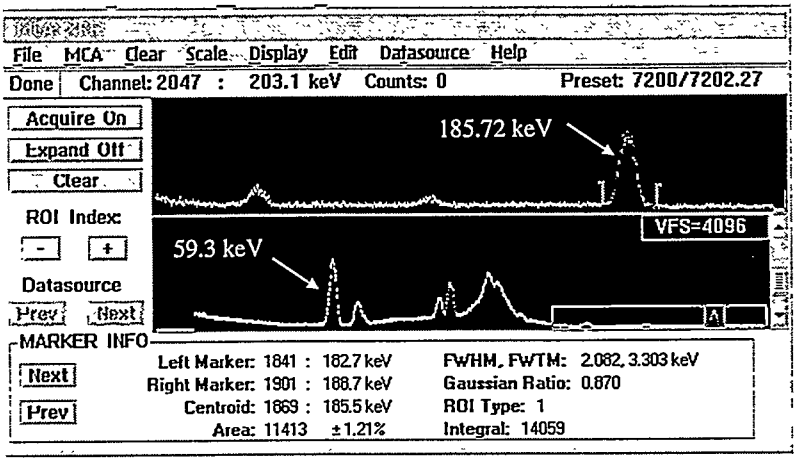


Figure 13. Genie-PC energy spectrum showing CHEM calibration photopeaks.

Enrichment Measurement

After the CHEM has been energy-calibrated, the user is returned to the main menu shown in Figure 12. From this menu the user may perform an enrichment measurement on either the 7.62-cm or 20.32-cm header pipe, print the results obtained so far, or store all the results and exit. Selecting one of the header pipe enrichment measurement options presents a series of dialog boxes reminding the user to check the orientation of the detector collimator, open the XRF source shutter, and enter the data acquisition time, if the default value is to be changed. Once these are confirmed, the CHEM begins taking data automatically.

At the end of the acquisition period, the data acquisition stops. The CHEM then analyzes the spectrum, calculates the enrichment, and presents the results in a window on the screen. Selecting **OK** in this window presents a dialog box for a second geometry measurement. The user should select **No** and return to the main menu, since the two-geometry technique is not necessary (see Section III). From the main menu, the user may perform an enrichment measurement on the second pipe, print the results obtained so far, or store all the results and exit. Measurements may be

repeated as necessary and the results may be printed at any time. However, only the most recent two spectra files for a particular pipe measurement are stored on the computer hard disk.

File Structure and Data Retrieval

The data are stored in directories according to the date and time at which the **XRF** enrichment monitor software program was started. For example, if the **XRF** software was started on March 5, 1998, at 14:35:00 hrs, the data for that run would be stored in the directory "C:\XRF\DATA\19980305\143500."

The results of all calculations for that run are stored in the file named REPORT.TXT in that directory. This file is created or updated when the results are printed and when the **XRF** program is exited. The spectrum for the 7.62-cm (3-in.) pipe measurement is stored in the file PIPE_3_1.CNF. The spectrum for the 20.32-cm (8-in.) pipe measurement is stored in the file PIPE_8_1.CNF. The "1" in the file name indicates that the parallel collimator geometry was used for the measurement. This is the first and only collimator geometry used at the PGDP. The program has the capability to make a second measurement, the perpendicular collimator geometry. If this selection were made, a "2" would appear in the file name. If two or more runs are taken, only the two most recent spectra are saved. The most recent is stored in the file with a .CNF extension, and the second most recent is stored in the file with a .BAK extension.

VI. Data Analysis

Several measurements using the CHEM were made during the IAEA inspection period. The enrichments declared by the facility during these measurements for both the 7.62-cm and 20.32-cm header pipes allowed a calibration of the CHEM. An initial calibration analysis of the CHEM data was done in December 1997. A second calibration was done in October 1998 using all available data. Results of the October calibration are presented here.

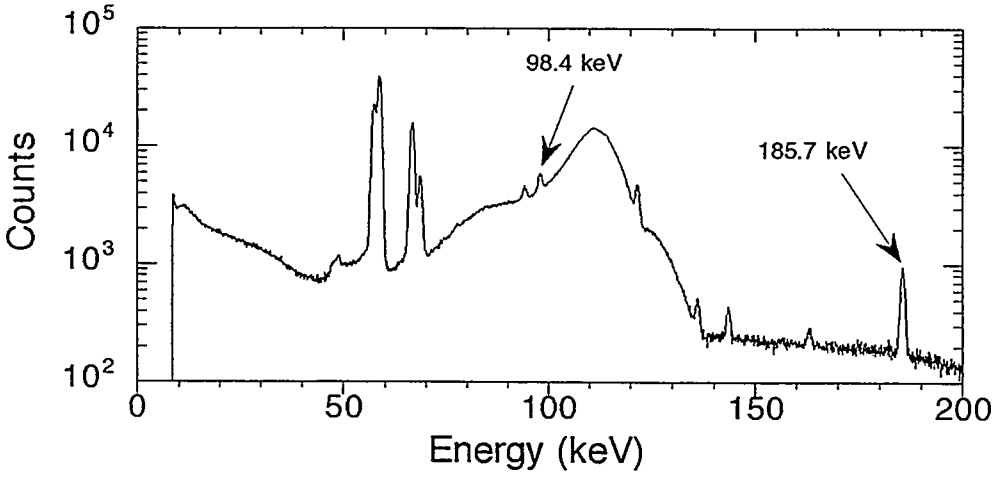


Figure 14. Energy spectrum from 7.62-cm HEU header pipe.

Figure 14 shows a typical HPGe spectrum taken on the HEU header pipe. The LEU spectra are similar. The resolution of this spectrum is 1.0 keV FWHM at 122 keV and 1.2 keV FWHM at

186 keV. The suggested minimum counting times for the HEU and LEU header pipes are 120 min and 30 min, respectively. A deposit measurement was made in an adjacent off-line cell, and also on the on-line cell during purges with dry nitrogen gas. The deposits were very small, but were included in the analysis.

The declared facility assays for the 20.32-cm LEU line were obtained from gas samples taken from diffusion cells at or near the measurement cell 25-7-3. The enrichments for the 7.62-cm HEU line were calculated from the declared enrichments for the cylinders on-stream (valved in) to the plant cascade as documented on the PGDP "X-326 HEU Side Feed Log" sheets. The calculation performed a weighted average of the enrichments of the cylinders valved in during the measurement based on the mass feed rates. The mass feed rate for each cylinder, R_{f_c}, was calculated by

$$R_{f_c} = M_c / t_c \ , \tag{3}$$

where M_c is the mass of UF_6 fed and t_c is the total feed time as indicated on the log sheet.

If one or more cylinders were valved in or out during a measurement, a corrected feed rate was calculated. For this, a fraction of the cylinder feed rate was used equal to the fraction of the time the cylinder was valved in during the measurement. Once the feed rates, and if necessary, corrected feed rates, were calculated for all valved-in cylinders, the facility enrichment, E_f, was calculated as

$$E_f = \sum_c R_{f_c} E_c / \sum_c R_{f_c} \ , \tag{4}$$

where E_c is the enrichment of the UF_6 in feed cylinder c.

As a result of the detector energy-resolution problem and modification described above, the CHEM has two calibrations, one for December 1997–May 1998 and one for July 11, 1998–October 8, 1998.

In order for the HEU data to be used as a calibration point, four conditions had to be met. Only the first two of these were required for the LEU data.

1. Sufficient statistics. Many of the measurements were not long enough to obtain sufficient statistics. These measurements may have been taken at normal gas pressures but were very short (less than 45 min for the 7.62-cm pipe or less than 10 min for the 20.32-cm pipe), or measurements that would normally be long enough but which were taken when there was virtually no UF_6 in the pipe. Typical counting statistics for the data used for the enrichment calibration were less than 5% for the 185.7-keV gamma-ray photopeak and less than 10% for the 98.4-keV x-ray photopeak.

2. Good energy resolution—less than 2.1 keV FWHM at 186 keV. During the measurement period, the CHEM HPGe detector occasionally experienced energy-resolution problems which compromised the quality of the measurements, mostly on the 7.62-cm HEU line.

3. Stable plant assay during entire measurement time. Cylinders have different feed rates depending on their size, mass, and time on-stream. To reduce the effect of different feed rates of different enrichment cylinders, measurements selected for calibration corresponded to times when all feed cylinders were of nearly the same assay or when the feed rate of a low-assay cylinder was very small compared to the feed rates of the other cylinders. This condition applies only to the 7.62-cm pipe measurements.

4. Stable feed during entire measurement time. Some of the 7.62-cm pipe measurements were performed when no cylinders were on-stream, or when cylinders were on-stream for only part

of the measurement. Therefore, the enrichment is known only to be somewhere between the LEU value in the 20.32-cm pipe and the enrichment provided by the cylinders most recently on-stream. Measurements selected for calibration had to be taken when cylinders were on-stream for the entire measurement. This condition applies only to the 7.62-cm pipe measurements.

For the 20.32-cm header pipe, a total of 24 measurements satisfied the first condition (sufficient statistics) and the results of these are given in Table 1. Of those 24, 12 measurements prior to the detector modification were also consistent with the second condition above and were used for the present calibration. After the modification to the detector, five measurements had sufficient statistics and were also consistent with the second condition, so the measurements were used for the calibration after the modification.

For the 7.62-cm pipe, a total of 27 measurements had sufficient statistics, and the results of these are given in Table 2. Of those 27, four measurements prior to the detector modification were left for the calibration after applying the remaining three conditions. After the repair, five measurements were two hours or more, but one had poor resolution, two had too low gas density, and one was taken on a day when no feed cylinders were on-stream and no assay sample was taken. This leaves only one measurement, on July 11, 1998, at 12:50 p.m., for calibration of the 7.62-cm data after the detector modification.

The new calibration constant K ΔK was obtained by plotting Eq. 1 as

$$\frac{R_{186(Total)} - R_{186(BG)}}{R_{XRF}} vs. E_f . \tag{5}$$

The slope of the data in this plot is 1/K, and a linear least squares fit to the data provides a value and uncertainty for K. This analysis results in four values of K, one for each pipe before and after the detector repair, as follows:

20.32-cm pipe December 1997–May 1998	K = 0.56 +/- 0.02
20.32-cm pipe July 11, 1998–October 1998	K = 0.53 +/- 0.02
7.62-cm pipe December 1997–May 1998	= .0 +/- 0.4
7.62-cm pipe July 11, 1998–October 1998	= .2 +/- 0.7

As can be seen above, the two K values for the 20.32-cm pipe agree, so the data cannot distinguish a change in the calibration from before to after the detector modification. The two K values for the 7.62-cm pipe nearly agree. However, since there is only one point after the detector modification which can be used for that calibration, it is not clear whether the 7.62-cm pipe calibration has changed. In either case, future installations of the CHEM will require a new calibration measurement.

VII. Results

Table 1 shows the data and calculated results for the 20.32-cm pipe measurements. The columns are: Date and Time in YYMMDD HHMMSS (year, month, day; hour, minute, second on a 24-hour clock), uranium 98.4-keV XRF count rate and uncertainty, ^{235}U 185.7-keV count rate and uncertainty, plant assay from a sample, the CHEM enrichment and uncertainty obtained from the calibration constant K, and a comment about the measurement. The comment states if the measurement was used for calibration and if not, why.

Table 2 shows the data and calculated results for the 7.62-cm pipe measurements using the same column headings, except the plant assay is obtained from cylinder feed data as discussed above in Section VI. The two additional columns are an alternate enrichment calculation discussed below at the end of this section.

Figure 15 shows the CHEM enrichment calculation for all 24 of the 20.32-cm LEU header pipe measurements along with the plant sample assays vs. the acquisition date. The average result is 7.8% +/- 0.7%, which is 17 standard deviations from 20% enrichment. The worst case point (a poor-resolution measurement) is 12.2% +/- 1.3%, which is 6 standard deviations from 20%. More than 1/3 of the enrichments from the plant sample assays lie outside the CHEM error bars, and possible explanations for this are discussed below (Section VIII).

Figure 16 shows the CHEM enrichment results for the 7.62-cm HEU header pipe measurements along with the enrichments calculated from the plant cylinder feed data vs. the acquisition date. Nineteen of the 27 measurements are shown. The eight measurements not shown are those noted in Table 2 as being taken when the detector energy resolution was poor, resulting in erroneously large CHEM enrichment values. As with the 20.32-cm pipe data, more than 1/3 of the enrichments from the plant sample assays lie outside the CHEM error bars, and possible explanations for this are discussed below (Section VIII).

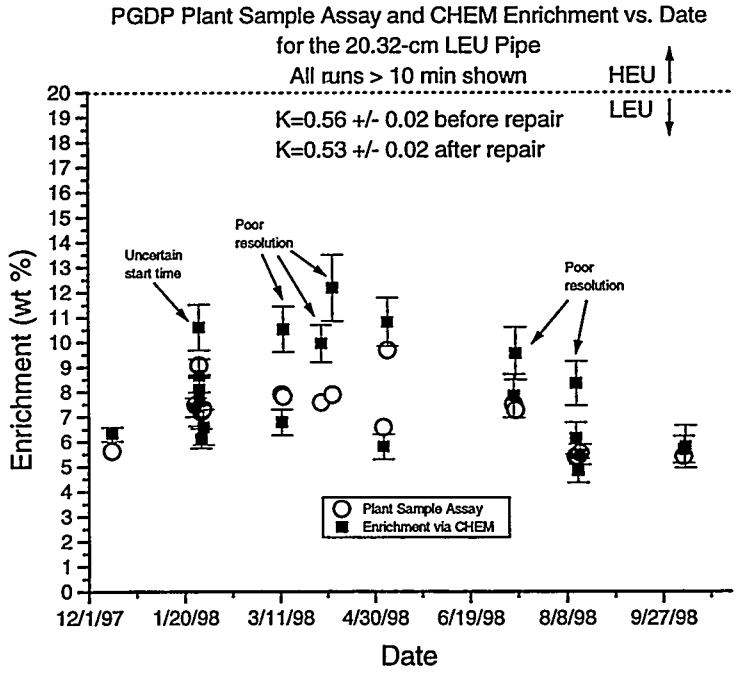


Figure 15. The 20.32-cm pipe plant assay and CHEM results.

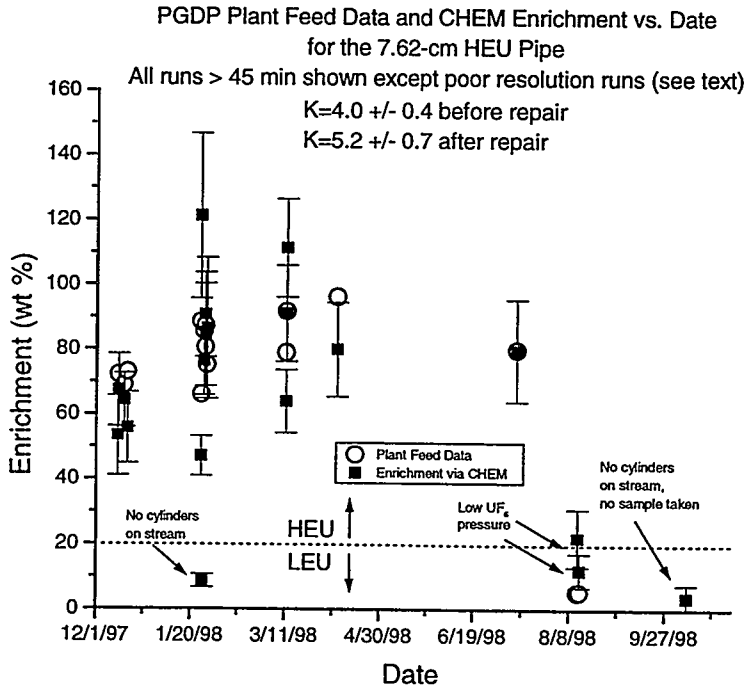


Figure 16. The 7.62-cm pipe plant assay and CHEM results.

Table 1. CHEM data for the 20.32-cm pipe

Date and Time	98.4-keV Rate (c/s)	98.4-keV Rate Uncertainty (c/s)	185.7-keV Rate (c/s)	185.7-keV Rate Uncertainty (c/s)	Plant Assay (wt. %)	CHEM enrichment* (wt. %)	CHEM enrichment uncertainty (wt. %)	Comment
971213 001100	0.074	0.001	0.87	0.01	5.605	6.30	0.28	Used for calibration
980125 155055	0.118	0.003	1.58	0.03	7.460	7.37	0.38	Used for calibration
980127 110549	0.097	0.006	1.88	0.07	9.061	10.6	0.91	File not written correctly to disk
980127 113020	0.119	0.010	1.65	0.14	9.061	7.59	0.98	Used for calibration
980127 113733	0.114	0.006	1.79	0.07	9.061	8.65	0.67	Used for calibration
980127 120018	0.118	0.005	1.73	0.06	9.061	8.02	0.57	Used for calibration
980127 123018	0.118	0.005	1.75	0.07	9.061	8.10	0.59	Used for calibration
980127 130018	0.117	0.005	1.72	0.06	9.061	8.08	0.57	Used for calibration
980128 113509	0.108	0.004	1.21	0.04	7.181	6.13	0.40	Used for calibration
980129 144815	0.126	0.009	1.52	0.11	7.286	6.57	0.71	Used for calibration
980311 173308	0.138	0.006	1.70	0.08	7.898	6.77	0.52	Used for calibration
980312 144650	0.101	0.006	1.93	0.08	7.795	10.5	0.91	Poor resolution
980401 104327	0.089	0.005	1.61	0.06	7.562	9.94	0.75	Poor resolution
980407 135200	0.081	0.007	1.80	0.09	7.864	12.2	1.3	Poor resolution
980504 110838	0.117	0.006	1.24	0.07	6.559	5.78	0.51	Used for calibration
980506 144030	0.099	0.007	1.94	0.09	9.671	10.8	0.97	Used for calibration
980711 150455	0.070	0.006	1.07	0.06	7.480	7.83	0.88	Used for calibration
980712 124609	0.074	0.007	1.37	0.06	7.247	9.55	1.1	Poor resolution
980812 111851	0.092	0.007	1.09	0.06	5.364	6.13	0.64	Used for calibration
980812 153742	0.066	0.006	1.08	0.05	5.364	8.33	0.89	Poor resolution
980813 132442	0.067	0.004	0.65	0.04	5.394	4.83	0.49	Used for calibration
980814 130150	0.104	0.005	1.11	0.04	5.524	5.47	0.41	Used for calibration
981007 164500	0.105	0.007	1.15	0.06	5.381	5.67	0.55	Used for calibration
981008 133200	0.060	0.007	0.69	0.05	--	5.79	0.85	No sample taken

*K = 0.56 +/- 0.02 December 1997–May 1998; K = 0.53 +/- 0.02 July 11, 1998–October 1998 (see discussion following Eq. 5).

Table 2. CHEM data for the 7.62-cm pipe

Date and Time	98.4-keV Rate (c/s)	98.4-keV Rate Uncertainty (c/s)	185.7-keV Rate (c/s)	185.7-keV Rate Uncertainty (c/s)	Plant Assay (wt. %)	CHEM enrichment* (wt. %)	CHEM enrichment uncertainty (wt. %)	E_{186}** (wt. %)	E_{186} uncertainty (wt. %)	Comment
971212 114600	0.039	0.006	0.58	0.06	--	53.4	12.4	43.0	5.92	Discontinuous feed
971212 172500	0.062	0.006	1.11	0.05	72.12	67.4	11.1	87.6	8.65	Large low-assay feed
971215 191600	0.076	0.002	1.28	0.02	68.89	64.4	8.30	102	8.73	Large low-assay feed
971217 104000	0.073	0.009	1.08	0.09	72.95	55.8	11.0	85.3	10.4	Used for calibration
980125 122700	0.038	0.006	1.20	0.05	88.46	121	25.5	95.4	8.95	Large low-assay feed
980125 185700	0.068	0.003	0.86	0.02	66.10	47.1	6.17	66.4	5.80	Large low-assay feed
980126 163000	0.062	0.003	0.19	0.02	--	8.71	2.04	11.2	2.34	Discontinuous feed
980127 010000	0.058	0.003	1.16	0.02	85.49	76.3	10.4	91.4	7.89	Large low-assay feed
980127 201600	0.075	0.010	1.64	0.08	87.22	84.4	15.9	131	12.7	Used for calibration
980127 220000	0.038	0.003	0.93	0.02	80.51	90.7	13.1	72.5	6.34	Used for calibration
980128 142611	0.043	0.009	1.02	0.07	75.16	86.5	21.7	79.8	9.11	Large low-assay feed
980128 200000	0.016	0.005	0.91	0.04	86.38	213	69.7	71	6.76	Poor resolution
980129 000100	0.014	0.005	0.81	0.04	86.38	212	77.1	62.4	6.16	Poor resolution
980129 040000	0.005	0.005	0.90	0.04	86.38	611	554	69.7	6.65	Poor resolution
980129 080000	0.013	0.007	0.80	0.05	89.04	238	131	62.1	6.86	Poor resolution
980311 115700	0.079	0.006	1.32	0.04	78.88	63.9	9.71	105	9.50	Large low-assay feed
980311 151700	0.057	0.005	1.35	0.07	91.66	91.1	14.8	108	10.8	Used for calibration
980311 185900	0.045	0.002	1.31	0.03	91.66	111	15.2	104	9.09	Large low-assay feed
980312 080000	0.022	0.006	1.20	0.05	92.22	189	56.1	94.8	8.98	Poor resolution
980401 130000	0.018	0.006	1.45	0.07	83.08	163	61.6	116	11.3	Poor resolution
980407 112400	0.048	0.005	1.02	0.07	96.20	80.0	14.5	80.2	8.71	Large low-assay feed
980506 122100	0.011	0.006	1.51	0.06	94.57	223	119	121	11.3	Poor resolution
980711 125000	0.053	0.006	0.88	0.05	79.86	79.8	15.8	80.2	6.90	Used for calibration
980712 103700	0.035	0.006	1.21	0.05	79.86	172	40.1	112	8.21	Poor resolution
980812 115700	0.027	0.005	0.17	0.03	5.364	22.1	8.85	11.1	3.62	Very little gas
980813 101100	0.040	0.005	0.15	0.03	5.394	12.1	5.20	9.15	3.55	Very little gas
981008 141035	0.052	0.006	0.10	0.04		3.73	4.00	3.68	3.88	No sample taken

*K = 4.0 +/- 0.4 December 1997–May 1998; K = 5.2 +/- 0.7 July 11, 1998–October 1998 (see discussion following Eq. 5).

**Q = 84.0 +/- 9.0 December 1997–May 1998; Q = 97.6 +/- 6.1 July 11, 1998–October 1998 (see discussion following Eq. 6).

When the detector had poor energy resolution, the count rate for the 98.4-keV XRF photopeak was inaccurate (too low) because of other nearby photopeaks and Compton scattering interfering with the background, and therefore net counts determination. This is especially true for the 7.62-cm pipe because of the tighter geometry resulting in a larger Compton scattering background. As a result, the enrichment values provided by the CHEM for the 7.62-cm HEU line were often larger than 100%. However, the 186-keV photopeak from ^{235}U is isolated, and an accurate measurement of the area can be obtained even if the detector energy resolution is poor (~2-3 keV). Therefore, the 186-keV photopeak area can be used alone to monitor the UF_6 enrichment if a few assumptions are made: (1) the enrichment does not vary greatly, (2) the pressure of the UF_6 gas does not vary greatly, and (3) there is no increase in the deposit on the pipe near the CHEM. These should be reasonable assumptions at the PGDP. To investigate this, a new quantity, Q, can be defined in analogy to Eq. 1 as

$$E = Q\left(R_{186(Total)} - R_{186(BG)}\right). \tag{6}$$

When this equation is plotted as $R_{186(Total)} - R_{186(BG)}$ vs. E_f, the slope of the data is 1/Q. The data from the 7.62-cm pipe measurements chosen for calculation of K before July 1998 were plotted, and a linear least squares fit to the data provided a value and uncertainty for Q. The results of this analysis gives Q = 84.0 +/- 9.0. This value was then multiplied by the 186-keV photopeak gas count rates of all 7.62-cm pipe measurements to produce a second enrichment value, E_{186}, as follows:

$$E_{186} = Q\left(R_{186(Total)} - R_{186(BG)}\right). \tag{7}$$

The Q value used after the detector repair was obtained from the measurement on July 11, 1998, at 12:50 p.m., which was also used to determine the K value. This value is Q = 97.6 +/- 6.1. Under the above assumptions, this second enrichment calculation should provide better enrichment values for the low-resolution CHEM measurements on the 7.62-cm pipe.

The last two numerical columns in Table 2 show E_{186} from Eq. 7 and the uncertainty in E_{186}. These enrichment results are plotted in Figure 17, along with the enrichments obtained from the plant cylinder feed data vs. acquisition date. This figure shows all 27 measurements listed in Table 2, even the poor-resolution measurements. There is reasonable agreement between the plant data and the calculation. This agreement indicates that these values can be used as alternate enrichment values for the erroneously large CHEM values which resulted from measurements with poor energy resolution.

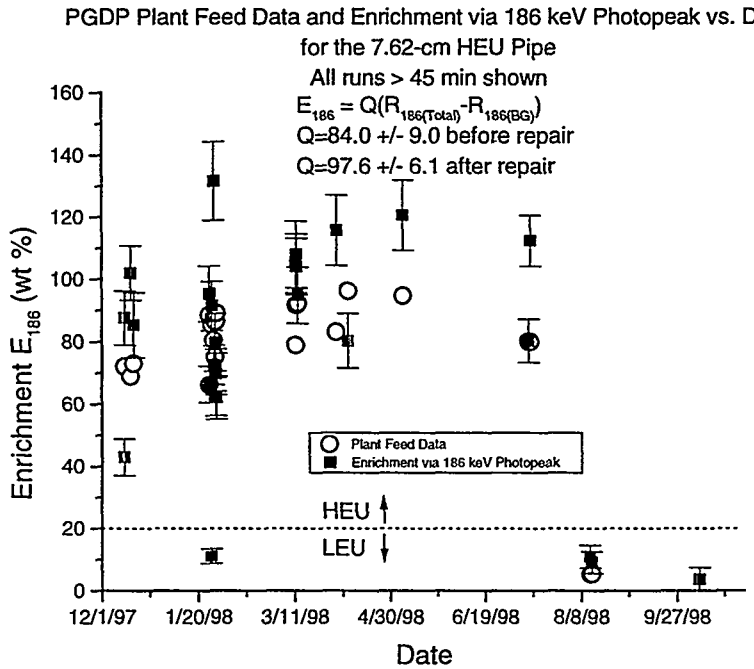


Figure 17. The 7.62-cm pipe plant assay and enrichment using only the 186-keV photopeak count rate.

VIII. Discussion

There are several factors which cause uncertainty in the facility enrichment even when cylinder or sample data are well known. They are difficult to quantify, and add to the dispersion of the CHEM data compared to the facility data. The CHEM calibration is therefore also susceptible to these uncertainties.

For the 20.32-cm pipe the uncertainties are as follows:

1. How similar is the assay of the cell where the measurements were taken to that of nearby cells where assay samples were taken?
2. Did the assay change between the time the assay sample was taken and the CHEM measurement was made?

For the 7.62-cm pipe the uncertainties are as follows:

1. What is the time between when a cylinder is valved in or out and when the effect reaches the measurement cell?
2. How should negative values of cylinder weights be handled? Cylinder weights as reported on the "X-326 HEU Side Feed Log" sheets were occasionally slightly negative, which indicates a weighing error. This suggests errors in positive weights of comparable size. However, this mass error is generally small and should have a negligible effect. Also, if several measurements are averaged, these uncertainties should tend to cancel, provided positive and negative weighing errors are equally likely.

3. What is the effect of cylinders with different feed rates? As mentioned above, cylinders have different feed rates depending on their size, mass, and time on-stream. This would affect the weighted average calculated in Eq. 4. However, over a measurement period of several hours, the average feed rate given by Eq. 3 is approximately equal to the actual rate at each point in the period. Therefore, this is expected to contribute a negligible effect.

These uncertainties for the 7.62- and 20.32-cm pipes might explain why more than 1/3 of the facility enrichment values are outside the error bars of the CHEM measured enrichment values. The calibration of both the 7.62- and 20.32-cm pipes would be much more accurate if samples could be taken during the CHEM calibration measurement on each pipe, and in the same cell being monitored by the CHEM.

IX. Conclusion

A system for rapid on-line NDA measurements of gaseous UF_6 enrichment has been installed and operated on HEU and LEU cascade monel header pipes at the PGDP. These activities were in support of the IAEA monitoring of the blend-down of 3.5 metric tons of United States excess HEU to LEU. The Los Alamos team worked with the IAEA during their verification experiment at the PGDP. The IAEA also conducted SNRI between January 1998 and July 1998. During several of these inspections, the detector was unavailable because of modifications being made to improve energy resolution. At other times, the HEU measurement was either consistent with cylinder feed data or significantly high, depending on the detector energy resolution being adequate or poor, respectively. The LEU measurements were consistently comparable to the plant declarations. The problem of energy resolution is more pronounced on the HEU line because of the tighter geometry, resulting in a larger Compton scattering background.

The conditions at the PGDP presented extremes in vibration and temperature for the CHEM. The vibration extremes were handled well by PGT's anti-vibration detector mount. However, the temperature extremes caused the energy-resolution problems. Modifications to the detector were made by the manufacturer to enable the detector to achieve good energy resolution at these temperature extremes. However, after the modifications, the detector still did not produce a spectrum of adequate quality, even though the cryostat vacuum was adequate. Additional work on the detector is planned.

All of the data have been analyzed and calibration constants obtained for both the 7.62-cm (3-in.) HEU and 20.32-cm (8-in.) LEU-product header pipes. All analyses and enrichment calibrations were based on the facility enrichment declaration. It was not possible to obtain a facility independent enrichment calibration. There were measurements when the resolution of the HPGe detector did not allow reliable enrichment values to be obtained for the HEU header pipe using the preferred analysis technique. For these data an alternate analysis, suggested by Mr. Tamas Biro of the IAEA, did provide reliable enrichment values.

LEU-product plant assays varied from 5.4% to 9.7%. The corresponding measurements varied from 4.8% +/- 0.5% to 10.8% +/- 1.0%. The average result for the LEU-product data is 7.8% +/- 0.7%, which is 17 standard deviations from 20% enrichment. The worst case for the LEU-product data is 12.2% +/- 1.3%, which is 6 standard deviations from 20% enrichment. This latter result was for data with poor resolution. There is basically

no probability that either of these measurements could be consistent with 20% enriched material. These results definitely confirmed that all LEU product was well below 20% enrichment.

Similarly, for the HEU feed, all measurements verified that the input was HEU. Calculated HEU plant assays varied from 66.1% to 96.2%. The corresponding measurements varied from 47.1% +/- 6.2% to 80.0% +/- 14.5%. Using the alternate analysis technique mentioned above, the measurements varied from 66.4% +/- 5.8% to 80.2% +/- 8.7%. When data were taken while the detector had poor resolution, the measurements using the standard analysis technique resulted in assays that were very high. With the alternate analysis technique, the largest result was 132% +/- 13%, less than 3 standard deviations greater than 100%. For those periods when the detector resolution was not adequate for the standard analysis technique, the alternate data analysis technique provides meaningful results.

This information, coupled with other containment/surveillance information, should allow the IAEA to verify that the HEU was indeed downblended.

X. Acknowledgements

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