Advanced Enrichment Monitoring Technology Based on Transmission Measurements with an X-ray Source and NaI(Tl) Spectrometer

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Abstract—In this paper we report our progress toward the development of an advanced enrichment monitoring technology for safeguarding gas centrifuge enrichment plants. We compare the UF₆ gas pipe attenuation and sensitivity to X-ray tube HV variations for two transmission energies: 22 keV and 25.5 keV. The first experimental enrichment results taken with a static UF₆ gaseous source and X-ray tube based transmission source over a wide gas pressure range are presented.

Index Terms—GCEP, enrichment monitoring, X-ray tube, uranium hexafluoride

I. INTRODUCTION

The classical approach to enrichment monitoring in process pipes of Gas Centrifuge Enrichment Plants (GCEPs) is based on two simultaneous measurements by a NaI(Tl) gamma spectrometer: a) the intensity of the 186-keV gamma ray characteristic of ²³⁵U decay and b) the intensity of a low energy transmission source used to correct for variations in the UF₆ gas density [1]. A Continuous Enrichment Monitor (CEMO) has been implemented for qualitative monitoring on a product header pipe [2]. However, routine replacement of

the decaying ¹⁰⁹Cd isotope source presents a major problem for the future implementation of this technology in GCEPs. We have proposed a new concept for an Advanced Enrichment

Monitor (AEM) in which the radioactive transmission source is replaced with an X-ray tube generator [3]. We have reported UF₆ gas transmission measurements with an X-ray tube and a ruthenium notch filter generating a transmission peak at 22.1 keV as an equivalent replacement of the ¹⁰⁹Cd isotope source [4]. The measured results confirm the attenuation in UF₆ gas and aluminum pipe similar to those reported in [2]. One practical factor that has to be addressed is the sensitivity of the results to high voltage instability. We have measured significant sensitivity (about 6-8 times) of the transmission peak intensity to variations of the anode high voltage (HV) power supply. The 22-keV transmission energy gives very good sensitivity to the UF₆ gas pressure (attenuation ~0.8% /torr attenuation for 100-mm gas thickness), but it is not optimal for the attenuation in the aluminum pipe. The attenuation in the pipe itself is important because it is a calibration parameter that enters into the expression for the enrichment. The effect of an error in the determination of the pipe thickness results in an even larger error in the attenuation in the pipe. We will present a transmission source with an energy chosen to reduce the attenuation in the aluminum pipe. We have used silver (Ag) as a notch filter that provides a transmission peak energy at 25.5 keV (about 20% higher than 22 keV). The attenuation in the UF₆ gas and aluminum pipe, and the tracking ratio between a flux monitor and NaI(Tl) detector, will be discussed. Initial enrichment measurements with a UF₆ static gas source and an enrichment monitor based on an X-ray tube will be presented and discussed.

II. DESIGN CONCEPT OF THE ENRICHMENT MONITOR.

Our enrichment monitoring setup as described in [3] consists of a UF₆ static gas source [5] and an enrichment monitoring head installed on the pipe (see Fig. 1). The enrichment monitoring head consists of an X-ray tube, a notch transmission filter and a NaI(Tl) detector in a tungsten shield and collimator. A flux monitoring detector (not shown) is

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placed between the notch filter and the UF₆ pipe. The additional modules necessary for the enrichment monitor are an X-ray tube power supply, Ortec Digidart MCA, and data acquisition module (DAQ). The DAQ is used to monitor and control the operation of the X-ray tube (high voltage and beam current settings) as well as to collect temperature data and the signal from the flux monitoring detector. For safety and noise immunity reasons, all electronics installed on the pipe is powered by DC voltage. Two separate DC voltages have been used: 24 V for the X-ray tube and 12 V for the MCA and DAQ modules. This ensures that the ²³⁵U passive measurements continue in case of a malfunction in the X-ray system. Both the MCA and DAQ interfaces are connected to a standalone PC via optically decoupled USB interfaces. Acquisition software [6], based on the existing BDMS [7], is used to collect, store, and display the information from the NaI(Tl) spectrometer, X-ray generator, and flux monitor.

Fig. 1. Block diagram of the enrichment monitoring prototype.

III. COMPARISON BETWEEN RU AND AG TARGETS

A. Attenuation in Pulse Height Spectrum

The attenuation of the X-ray tube bremsstrahlung spectrum through the transmission target, Al pipe, and UF₆ gas was calculated using a one-dimensional model based on mass attenuation coefficients [4, 8]. Because of significant attenuation in the Al pipe at 22 keV, we chose a 0.19 mm thick Ru target that provides about 1500 cps intensity of the transmission peak with the X-ray tube operated at 25 kV and 200 μ A current. The calculated spectra are presented in Fig. 2. This shows the initial spectrum from the X-ray tube, the spectrum after the Ru foil, and the effect of the Al pipe wall and the UF₆ gas.

The attenuation factor of the ROI below 22.1 keV is 66 in the Ru target, $\sim$ 2800 in the Al pipe, and 1.8 in the UF₆ gas. The different slopes of the spectra after the Ru and Al indicate a significant difference in the shapes of the transmission peak before and after the pipe and UF₆ gas. These different shapes lead to differences in the relative response between the flux monitor and the NaI(Tl) detector. An increase in the thickness of the Ru target would reduce the difference in the spectrum shapes but at the price of a significant loss of intensity. We therefore chose a different strategy to generate the transmission line at a different energy by using a different

material for the filter (see Table I).

Fig. 2. Calculated relative pulse height spectra from the X-ray source, after the 0.19-mm Ru target, 10-mm Al pipe wall, and 10-cm UF₆ gas at 50 torr.

Table I
Some Feasible Energies for Transmission Peaks

Target	Ru	Pd	Ag	Cd	In	Sn
E (keV)	22.1	24.4	25.5	26.7	27.9	29.2

The attenuation using a 0.5-mm Ag filter with a 33-kV anode voltage and 250- μ A anode current is shown in Fig. 3. The attenuation of the region of interest below 25.5 keV is $\sim$ 1300 in the Ag target, $\sim$ 200 in the Al pipe, and 1.6 in the UF₆ gas.

Fig. 3. Calculated relative pulse height spectra from the X-ray source, after the 0.5-mm Ag target, 10-mm Al pipe wall, and 10-cm UF₆ gas at 50 torr.

The reduced sensitivity of the transmission peak attenuation due to gas pressure for Ag is shown in Fig. 4.

Fig. 4. Attenuation of 22.1-keV and 25.5-keV transmission peaks from 0.19-mm Ru and 0.5-mm Ag targets calculated at various UF_6 gas pressures. The sensitivity with Ag is $\sim 15\%$ less at 50 torr.

B. Shape of Transmission Spectra for Ru and Ag Targets

The transmission spectrum after the notch filter and the Al pipe is not a monoenergetic line but a pulse height distribution whose shape depends on the bremsstrahlung energy and the absorption in the notch filter. The normalized shapes of the spectra after attenuation in the target and Al pipe are shown for comparison in Figs. 5 and 6.

Fig. 5. Relative pulse height spectra after attenuation in the 0.19-mm Ru target and Al pipe, and attenuation in the Al pipe. The attenuation is ~ 1000 at 22.1 keV but increases to ~ 2800 for the total absorption edge peak due to the greater attenuation at lower energies.

The difference in attenuation in the target and the Al pipe in Figs. 2 and 3 affects the difference in the transmission peak pulse height distribution before and after attenuation in the Al pipe. Because of the narrower shape, the pulse height spectra from the Ag target are much easier to track for stabilization purposes.

Fig. 6. Relative pulse height spectra after attenuation in the 0.5-mm Ag target and Al pipe, and attenuation in the Al pipe. The attenuation is ~ 100 at 25.5 keV but increases to ~ 200 for the total absorption edge peak due to the greater attenuation at lower energies.

IV. TRANSMISSION PEAK STABILITY

A. Sensitivity to Variation of HV Power Supply

The combination of the high attenuation of the aluminum pipe and its energy dependence leads to a significant dependence of the transmitted intensity as a function of HV. We calculated the sensitivity to HV voltage as a ratio between the relative changes in the transmission peak. The experimentally determined sensitivity is compared with the calculated value in Fig. 7.

Fig. 7. Experimental and calculated sensitivity of the transmission peak to changes in the X-ray tube high voltage. The beam current is kept constant 200 μA .

The transmission peak intensity for the Ag target changes by about four times the change in high voltage. In the case of the ruthenium target the change is about twice as great. Propagation of this uncertainty to the measured enrichment for 50-torr UF_6 gas as calculated in [9] increases the sensitivity of the enrichment value to the high voltage to about a factor of

10. This implies that a high voltage supply with variations of a fraction of a percent (from temperature drift, aging, etc.) would cause a relative enrichment error of the order of several percent. To address this issue we have developed three approaches: improvement of the stability of commercial HV power modules for X-Ray tubes, use of a flux monitor to correct for the total instability of the transmission source, and use of a self-calibration methods that correct the instability of both the X-ray tube and the NaI(Tl) spectrometer. While this is an object of a separate study that will be reported later [10], we provide some initial results about tracking the intensity of the transmission peak with an integral flux monitor.

B. Flux Monitor Tracking

We have used a thin Si photodiode [11] placed in the collimator after the transmission target to monitor the intensity of the transmission peak before the attenuation in the aluminum pipe and UF_6 gas. The initial data have been taken with a Keithley picoammeter Model 6487 with a sensitivity of 10 fA. The relative changes of the flux monitor current and the intensity of the transmission peak of the NaI(Tl) spectrometer versus high voltage are plotted in Fig. 8.

Fig. 8. Measured sensitivity of flux monitor current and intensity of transmission peak to HV normalized at 30 kV.

The ratio between the flux monitor current and transmission peak intensity changes by only 2% for a 10% relative change of HV. This virtually eliminates the effect of any expected HV fluctuations on the enrichment value. Further investigations on aging and temperature dependence of the flux monitor are in progress.

V. ENRICHMENT MEASUREMENTS

A. Experimental Setup

The first prototype of a UF_6 source and Advanced Enrichment Monitor developed under the Flow and Enrichment Monitor (FEMO) project has been used to take initial enrichment measurements at different pressures. A photograph of the UF_6 source and enrichment measurement head installed on the pipe is shown in Fig 9. The measurement head with the X-ray tube and NaI(Tl) detector are installed on the pipe; the rest of the equipment is on the bench.

Fig. 9. Prototype overview of the static UF_6 source and enrichment monitoring equipment.

B. Description of Experiment

The pressure in the pipe was increased stepwise from 3.3 torr to 70 torr, 10 torr per step, over several days. The pressure was changed twice per day allowing four to eight 2-hour spectra to be acquired for each pressure point. The data at 50 and 60 torr were used to calibrate the instrument based on the known value (in our case 3.3% ^{235}U). That calibration was used for calculation of the calibration at other gas pressures.

C. Experimental Data for Transmission and Isotope Peak Intensities versus UF_6 Gas Pressure

The measured intensity of the 186-keV line and the attenuation of the 25-keV transmission line in UF_6 gas are presented in Figs. 10 and 11.

D. Enrichment Data

The data in Figs. 12 and 13 show the measured enrichment and its relative deviation from the nominal enrichment of 3.3%.

Fig. 10. Intensity of the 186-keV line from 3.3% enriched UF_6 gas measured at different pressures.

The good linearity and negative offset of the 186-keV peak intensity data indicate that there is practically no buildup of wall deposits even though the pipe has been filled with gas at different pressures since November 2008. The response of the 186-keV peak is about 0.52 cps/torr for 100-mm diameter

header pipe. In real applications in GCEPs this implies a few cps for the low pressure header pipe and a few tens of cps for the high pressure header pipe.

Fig. 11. UF₆ gas attenuation of 22-keV and 25.5-keV transmission peaks from 0.19-mm Ru and 0.5-mm Ag targets measured at different UF₆ gas pressures.

The measured sensitivity to UF₆ gas pressure for a 100-mm header pipe is about 0.78%/torr for a Ru (22.1 keV) target and 0.66%/torr for a Ag (25.5 keV) target. One reason for the systematically lower sensitivity from that reported in [2] could be the wider angle of transmission flux, which shortens the effective path length through the UF₆ gas.

Fig. 12. Measured enrichment at different UF₆ gas pressures. A 3.3% enrichment value has been assigned at 50 torr.

Fig. 13. Relative deviation of measured enrichment values at different UF₆ gas pressures.

VI. DISCUSSIONS AND CONCLUSIONS

This paper has presented the first results of an Advanced Enrichment Monitor that is intended for use in Gas Centrifuge Enrichment Plants. The instrument measures the ²³⁵U content of the gas by measuring the intensity of the 186-keV peak and measures the gas density by the transmission of a low energy X-ray. A single NaI(Tl) detector is used for both measurements. The transmission peak is created by using a notch filter of Ag or Ru. The Ag filter is expected to lead to a lower overall uncertainty because of the order of magnitude less attenuation in the Al pipe with only a ~15% less attenuation in the UF₆ gas. The measurements show good linearity with gas pressure. A 2-hour measurement gives results that differ from the known value by less than 1% relative (at most pressures). Experiments have been carried out to study the effect of the stability of the HV source. Three methods are being pursued to minimize this effect: 1) improve the stability of the X-ray tube HV, 2) monitor the X-ray tube output, and 3) correct for both the tube and NaI(Tl) instability with a self-calibration method. The demonstrated performance of the method would be useful for both safeguards and operator process measurements.

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