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New technology for transmission measurements in process pipes

C.E. Moss *, A. Favalli, J.M. Goda, K.D. Ianakiev, M. Lombardi, C.W. McCluskey, M.T. Paffett, M.T. Swinhoe

Los Alamos National Laboratory, Los Alamos, NM 87545, USA

HIGHLIGHTS

- Replaced radioactive transmission source with X-ray tube.
- Used X-ray filter to generate an X-ray peak for transmission.
- Achieved excellent long term stability.
- Provided UF_6 enrichment monitor as example.

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ABSTRACT

Transmission measurements of radiation through process pipes provide a non-intrusive method of determining the amount of product present in the pipes. The product could be a liquid, a slurry, or a gas, which is the most challenging because of the low density. Traditionally, these techniques have used a radioactive source that has to be replaced periodically. We have developed a transmission technique based on an X-ray tube instead of a decaying source. A notch filter is used to provide a narrow transmission line, and a thin silicon transmission detector is used to monitor the X-ray tube output. The transmitted X-rays are measured with a high-throughput gamma spectrometer that consists of a NaI(Tl) detector and an MCA with precise dead time correction. This spectrometer provides stable transmission measurements with an accuracy of a fraction of a percent. The shielding and collimator are made of machinable tungsten for thermal mechanical stability, as well low-cost, low-weight tungsten powder in polymer castings. We describe two methods of measuring the pipe wall thickness without evacuating the pipe. Our particular application was for enrichment monitors for UF_6 in process pipes. Enrichment monitors that are independent of the plant data require two measurements: a transmission measurement to determine the total amount of uranium in the pipe and a measurement of the 186-keV gamma-ray line to determine the amount of ^{235}U present. The ratio of these values gives the enrichment. Previous designs used a decaying radioactive source such as ^{57}Co (122 keV, $T_{1/2}=272$ days) or ^{109}Cd (22 keV, $T_{1/2}=1.2$ years). A major effort was required to access and periodically replace these sources in operating plants. In this report, we describe the use of an X-ray tube, which eliminated the source problem, and other innovations. Then we present data from an enrichment monitor that incorporates these innovations.

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1. Introduction

Gamma-ray and X-ray transmission measurements, as well as the related techniques of Compton scattering, are used in a wide variety of industrial and medical applications. These applications include measurements of the composition of multi-phase flow in piping from oil wells, thickness gauges for production of flat-rolled steel and aluminum, level measurements in vessels, bone density measurements, inspection of the explosive charge in artillery shells, monitoring of the ash in coal on conveyor belts, monitoring the amount of tobacco in cigarettes, and soil density gauges. Tjugum et al., (2002), Johansen and Jackson (2004), Hoeppner (2009), and Kelly and Boyes (2009) give more details about some of these applications. Most of these instruments employ a radioactive source, which requires conformance to safety regulations and must be periodically inspected for leaks, or replaced, if sources with short half-lives, such as ^{57}Co (272 days) or ^{109}Cd (463 days), are used.

In this report we discuss measuring material in pipes. The material could be a liquid, a slurry, or a gas, which is the most challenging because of the low density. A previous report described two methods that have been applied for assaying UF_6 in pipes (Moss et al., 1999). The first was based on the transmission of gamma rays while the second was based on X-ray

* Corresponding author.
E-mail address: cmoss@lanl.gov (C.E. Moss).

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fluorescence. The present report first describes several innovations that might improve many transmission applications. It then presents results from an improved UF_6 enrichment monitor that incorporates these innovations.

2. Original transmission design

Our original transmission design was part of a UF_6 enrichment monitor. A cross section is shown in Fig. 1. The pipe was steel, 100 mm ID and 108 mm OD, containing UF_6 at approximately 5.33 kPa. The transmission source was approximately 100 μCi of ^{57}Co, which has a 122-keV line that can penetrate the steel pipe, and the detector was a 76.2-mm diameter × 12.7-mm thick NaI(Tl) scintillator. The tungsten collimators on the source and the detector were designed to maximize the size of the beam incident on the detector without causing edge effects in the detector. A NaI(Tl) detector was used instead of a cheaper ionization chamber or a Geiger–Mueller (GM) counter because it provided greater sensitivity and allowed a region of interest (ROI) to be set.

The well-known transmission source formula is the following:

$$I = BI_{inc}e^{-\mu_w x_w}e^{-\mu_m x_m} \quad (1)$$

where I=flux at detector, B=buildup factor, I_{inc}=flux incident on pipe, μ_w=wall linear attenuation coefficient, x_w=wall thickness, μ_m=linear attenuation coefficient of material in pipe covered by beam, x_m=thickness of material in pipe.

The buildup factor B is approximately equal to 1 if the collimators are narrow and the region of interest (ROI) used to analyze the transmission peak in the spectrum is small enough to reject scattered radiation. The linear attenuation coefficient μ is equal to the mass attenuation coefficient μ/ρ times the density ρ. The mass attenuation μ/ρ is often what is tabulated because it is independent of the density, which is especially important with gases, and it depends only on the atomic cross section.

Such a traditional design has several advantages. Sources are available that provide monoenergetic or well-separated lines. Commonly used sources, such as ^{241}Am, ^{109}Cd, ^{137}Cs, ^{60}Co, and ^{228}Th, provide energies up to 2.6 MeV. Sources have stable outputs, except for the decay for which a correction is easily made. Sources are small in size and do not require a power supply, unlike X-ray tubes. However, sources also have some disadvantages. They must be replaced or inspected periodically for leaks. Access to them may be difficult and expensive in some facilities or remote locations. They require a license and a trained source handler.

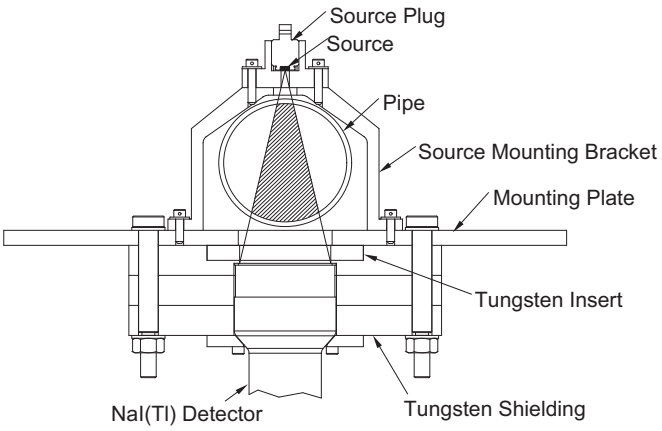

Fig. 1. Cross section of the original transmission design for the enrichment monitor.

3. New transmission design

3.1. X-ray tube and filter

X-ray tubes are used in place of radioisotope sources for measurements in process pipes as well as for many other applications. In the simplest design, the bremsstrahlung beam with superimposed characteristic anode X-rays from the X-ray tube passes through the pipe and is detected by a GM counter. In a more complex design, the bremsstrahlung beam strikes a target to produce the K_α and K_β characteristic X rays of the target material. These X-rays are then transmitted through the pipe to a detector. A recent example of this type of system is the one developed by Tjugum and Mihalca (2009) for multi-phase flow composition measurements.

We have chosen an X-ray tube with a transmission filter to replace the decaying isotopic source. The existence of a K edge in the filter material can be used to create a peak in the transmitted spectrum at the K-edge energy. Fig. 2 (top) shows the photon mass attenuation coefficient of bismuth, which has the highest energy K edge, 90 keV, of the non-radioactive elements. Fig. 2 (bottom) shows the calculated transmitted spectra after a 2-mm Bi filter and after passing through 5 mm of steel pipe wall. Goda et al. (2009) and Lombardi et al. (2012) provide additional details of similar calculations. The X-ray tube anode was tungsten and the tube was operated at 120 kV. The width of the peak can be reduced by increasing the filter thickness but with a corresponding loss of intensity and a small shift of the peak centroid to higher energy. Increasing the output from the x-ray tube, provided the maximum output is not exceeded, can compensate for the loss of intensity with increasing filter thickness.

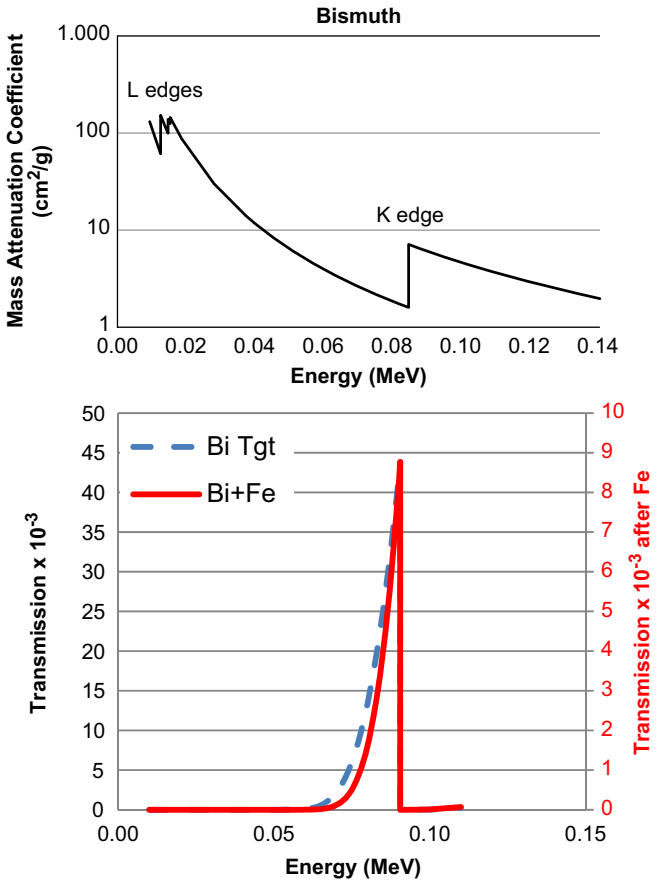

Fig. 2. Top: bismuth photon mass attenuation coefficient. Bottom: calculated photon spectrum incident on the pipe (dashed line) and after passage through the pipe (solid line).

Table 1
Properties of some metallic transmission filters.

Element	K-edge energy (keV)	Density (g/cm^3)
Ni	**8.3**	**8.9**
Mo	**20.0**	**10.2**
Ag	**25.5**	**10.5**
Sn	**29.2**	**7.3**
Gd	**50.2**	**7.9**
Ta	**67.4**	**16.7**
Au	**80.7**	**19.3**
Bi	**90.5**	**9.8**
U	**115.6**	**18.9**

An X-ray tube with filter has several advantages over radioisotope sources. Many energies from 0.18 keV (B) to 90 keV (Bi) are possible; Table 1 lists the properties of some readily available filter materials (European Synchrotron Radiation Facility website, 2006). The X-ray tube must generate energies higher than the K edge, but such tubes are available from several vendors. After the filter, the beam is essentially monoenergetic, and the intensity can be easily changed by changing the x-ray tube filament current. If a tube is operated well below its rating, it will last many years and does not need to be periodically replaced or inspected. The most important advantage is that when power is turned off, there is no radiation, and consequently, there are fewer regulations compared to those for a radioisotope source. The main disadvantage is that an X-ray tube requires a power supply. If high accuracy of the measurement is required, additional electronics may be required for stability. These additional requirements make the system more complex than one based on a radioactive source and hence potentially less reliable. The X-ray technique cannot be used above 90 keV (Bi), unless one is willing to use the radioactive elements Th, U, Np, or Pu (122 keV) as filters. If the application requires a higher energy for sufficient penetration, then one may be forced to use an isotopic source such as ^{137}Cs (662), ^{60}Co (1173, 1333), or ^{208}Tl (2614). In addition, an X-ray tube is larger in size than a radioisotope source and therefore requires more space and a larger shield.

In a recent enrichment monitor design for an aluminum pipe we have been able to use the silver K-edge peak at 25.5 keV instead of the 122-keV peak that we used for the steel pipe. We chose an X-ray tube with a tungsten K edge at 69.5 keV and operated it with a HV of 33 kV. The choice of transmission energies is a complex tradeoff between the attenuation in the pipe and UF_6 gas, operating voltage of the X-ray tube, and the NaI(Tl) detector response (Ianakiev et al., 2009b). For example, at low energy the attenuation by the gas is higher but a higher energy beam can more easily penetrate the pipe walls.

3.2. *Transmission source stability*

The type of X-ray tube employed is critical for long-term stability and reliability. The life expectancy of an X-ray tube is driven mainly by filament erosion and vacuum stability. Therefore, we have chosen an X-ray tube with a tungsten anode capable of operating at nominal beam currents many times higher than needed for this particular application. The reduction of the operating current, and therefore the filament and anode operating temperatures, leads to dramatic reduction of all aging effects: increasing filament life expectancy while decreasing anode erosion, and vacuum deterioration. For example, reduction of the operating current by a factor of two leads to an increase of the filament life expectancy by more than an order of magnitude.

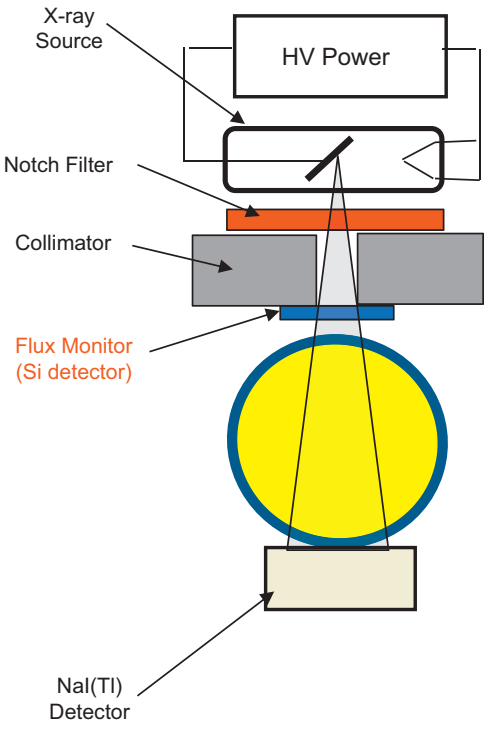

Fig. 3. Schematic of the transmission system showing the filter and flux monitor.

Fig. 4. Detector shield made of tungsten powder in a special polymer.

To further improve the long-term stability for high accuracy measurements we investigated monitoring the beam intensity with a thin silicon photodiode placed between the X-ray tube and the pipe as shown in Fig. 3. We operate the photodiode in the photovoltaic mode (without bias), and measure the current with a picoammeter. The measured current varies slightly with temperature, but we record the temperature and correct the current. Possible radiation damage to the photodiode was investigated by irradiating it at 1000 times the normal operating level, but no change in the performance was observed (Lombardi et al., 2011). The output of this monitor could be fed back to the filament current to keep the peak intensity constant or used in the analysis to correct the transmission peak intensity. Stability data for the enrichment monitor are presented in Section 5.

3.3. Shielding

In the original enrichment monitor design we used machinable tungsten metal for shielding. In the new design we use machinable tungsten only where thermal mechanical stability and accurate dimensions are needed, for example, in collimators, and special castings of tungsten powder in a polymer (supplied by Tungsten Heavy Powder, Inc.) elsewhere. The latter material has a density of 11 g/cm^3, which is comparable to the density of lead, and it is non toxic. It is much less expensive than tungsten metal, which has to be sintered, and can be cast into any shape, but it is somewhat fragile and subject to chipping. Fig. 4 shows an example of the powder tungsten shielding.

3.4. Detector and electronics

We are using a NaI(Tl) detector because it provides good efficiency and moderate resolution and is relatively inexpensive. It has been well-known for many years that the output of a NaI(Tl) detector varies with temperature, which is a problem for outdoor applications. The gain varies with temperature and is routinely stabilized with a circuit that maintains a peak in the spectrum at a fixed channel position. It was believed that the output intensity was nonlinear until it was reported (Ianakiev et al. 2009a) that the output is linear but the relative contributions of the fast and low scintillation components change. We developed and now use a simple circuit to compensate for this variation with temperature.

Pileup can also affect high-accuracy measurements because it is not linear with count rate. We are using a multichannel analyzer with digital signal processing. We worked with the vendor to optimize the various parameters of the fast and slow channels to minimize the pileup effect. We correct the transmitted count rate for the remaining pileup (Knoll, 2010).

4. Calibration

4.1. Empty pipe

The simplest calibration technique requires measurement of the transmitted intensity when the pipe is empty. In this case, the measured intensity is given by

$$I = I_{inc} e^{-\mu_w x_w} e^{-\mu_m x_m} = I_0 e^{-\mu_m x_m} \tag{2}$$

where I_0 is the measured empty pipe intensity. If the material normally in the pipe is a liquid or slurry, then air in the empty pipe may be acceptable. However, if the material normally in the pipe is a gas, then vacuum is preferred for the empty measurement. When material is present in the pipe,

$$\rho_m x_m = \frac{1}{(\mu_m \rho_m)} \ln\left(\frac{I_0}{I}\right) \tag{3}$$

The quantity μ_m/ρ_m is the mass attenuation coefficient. Either ρ_m or x_m can be determined if the other is known. Determining x_m is required if the pipe is horizontal and only partially full.

4.2. Two-energy method

If an empty pipe measurement is not possible in an operating facility, then other approaches may be possible. The two-wall thickness, x_m, is usually the unknown that must be determined for the calibration. In the x-ray region, ≤ 90 keV, because of the existence of X-ray edges, it may be possible to find two energies where the attenuation coefficients of the material in the pipe are equal but the coefficients of the pipe wall material are different. If

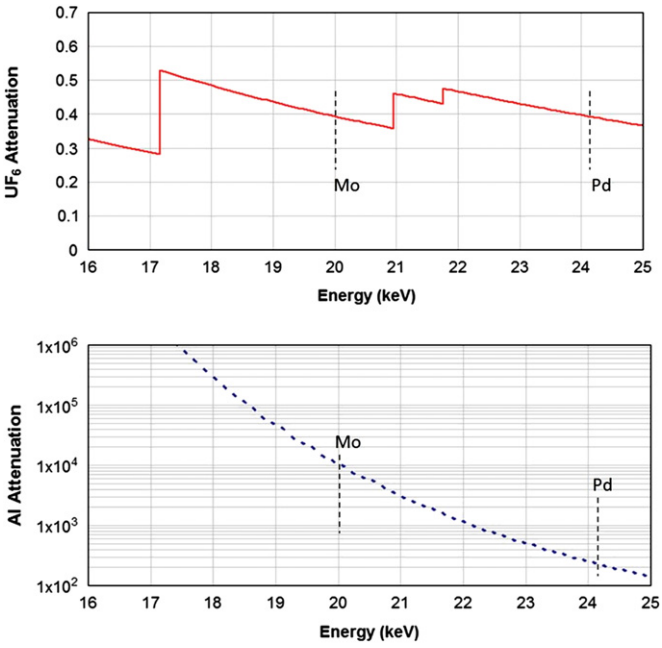

Fig. 5. Attenuation as a function of energy in UF_6 gas (top) and an Al pipe with 100 mm ID and 110 mm OD (bottom). Note the log scale on the Al attenuation. The energies of the molybdenum and palladium X-ray K edges are indicated.

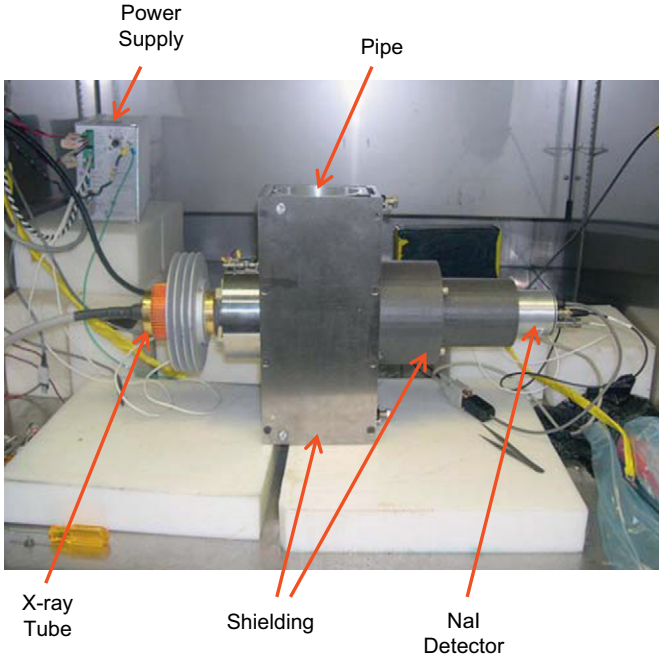

Fig. 6. A laboratory version of an enrichment monitor that includes all of the techniques discussed in this report. In this setup, the pipe does not contain gas.

subscripts 1 and 2 denote the two energies, then

$$I_1 = I_{inc1} e^{-\mu_{w1} x_w} e^{-\mu_{m1} x_m} \tag{4}$$

$$I_2 = I_{inc2} e^{-\mu_{w2} x_w} e^{-\mu_{m2} x_m} \tag{5}$$

If $\mu_{m1} = \mu_{m2}$, then Eqs. (4) and (5) can be solved for x_w, assuming I_{inc1}, I_{inc2}, μ_{w1}, μ_{w2}, and ρ_w are known.

$$x_w = \frac{1}{(\mu_{w2} - \mu_{w1})} \ln\left(\frac{I_1 I_{inc2}}{I_2 I_{inc1}}\right) \tag{6}$$

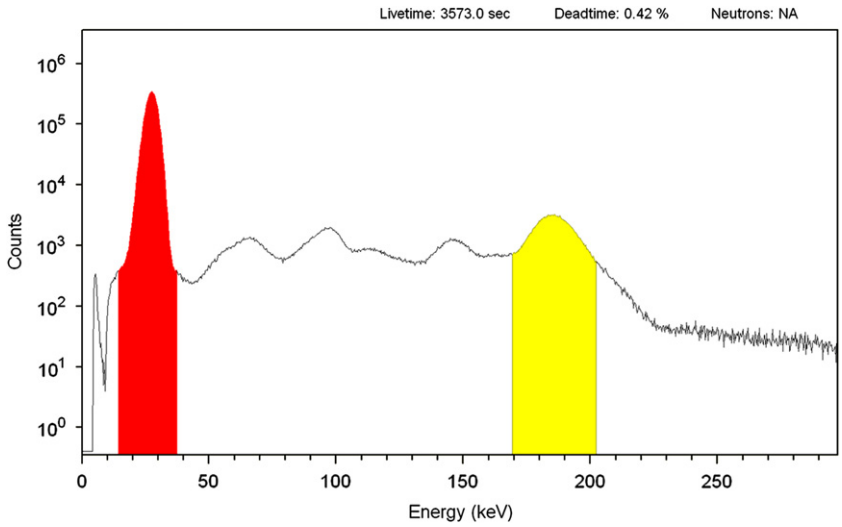

Fig. 7. Spectrum measured with the new enrichment monitor design when the pipe contains 6.67 kPa of UF_6. The 25-keV transmission peak is produced by the X-ray tube with a silver filter. The 186-keV peak is from ^{235}U.

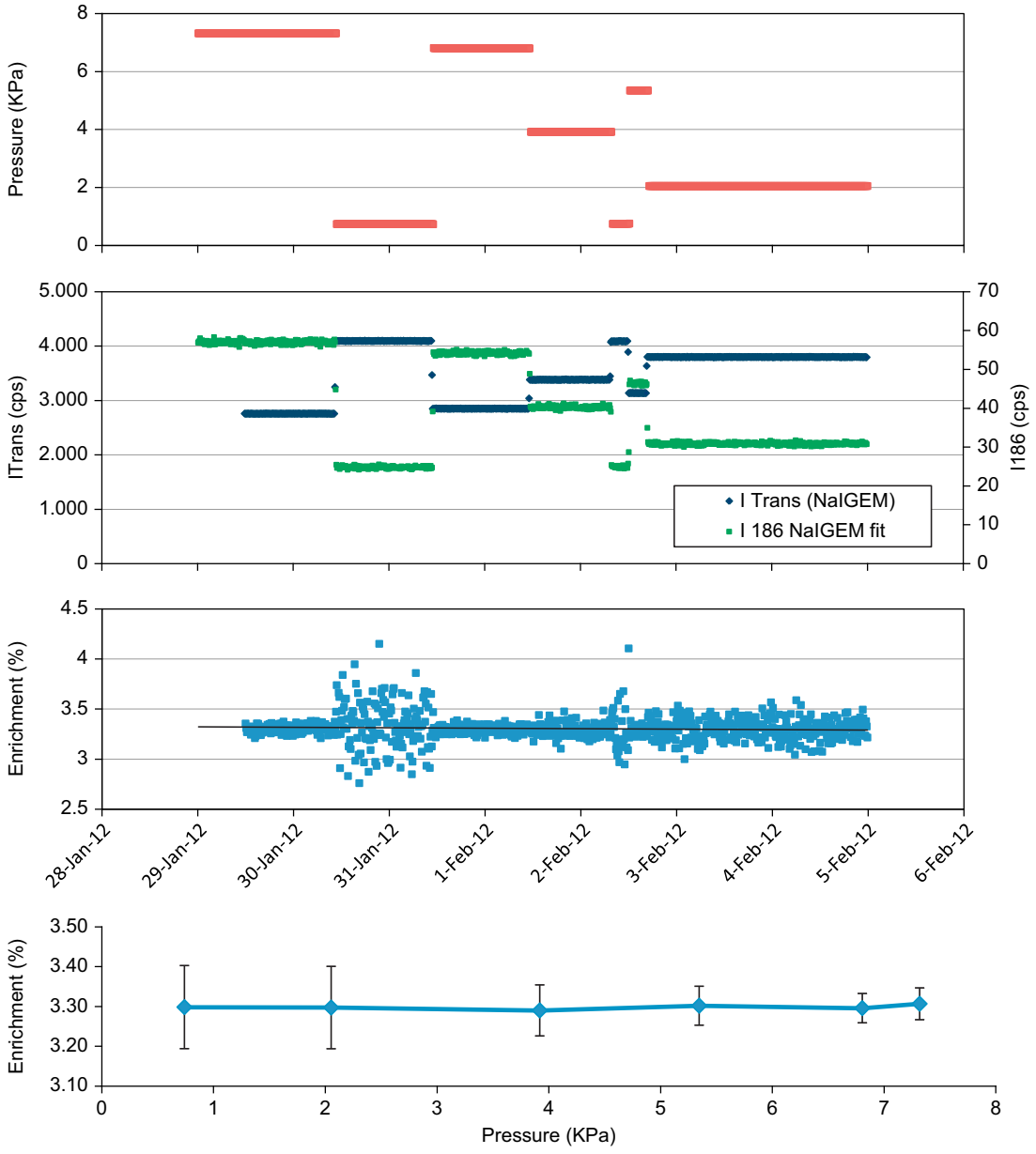

Fig. 8. Performance of the enrichment monitor when the pressure is changed. The top chart shows the pressure vs. date. The second chart shows the 122-keV transmission peak and the 186-keV peak counting rates vs. date. The third chart shows the ^{235}U enrichment vs. date. The fourth chart shows the ^{235}U enrichment vs. pressure.

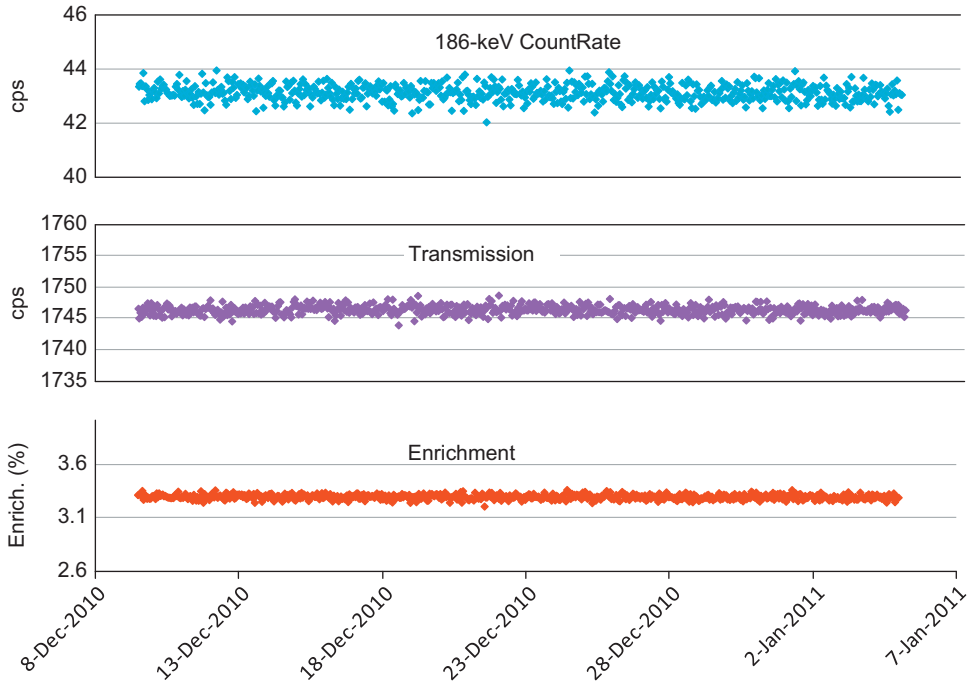

Fig. 9. UF_6 enrichment monitor data acquired with the new enrichment monitor design. The transmission is the 25-keV peak count rate. The enrichment is calculated with Eq. (8).

We applied this method as shown in Fig. 5 to an aluminum pipe containing UF_6 gas at 50 Torr (Lombardi et al., 2012). The attenuation in UF_6 at the Mo K edge (20.0 keV) is the same as the attenuation at the Pd K edge (24.4 keV), but the attenuation in the aluminum varies by a factor of approximately 60. We were able to determine the aluminum pipe wall thickness to $\pm 1\%$.

4.3. Different materials in pipe

If an empty pipe measurement is not possible but the material in the pipe can be varied and the properties are known, then a calibration may also be possible. The goal in this method is to determine the empty pipe intensity I_0. If a and b denote the different materials in the pipe, then

$$I_0 = \frac{I_b - I_a}{e^{-\mu_{mb}x_m} - e^{-\mu_{ma}x_m}} \tag{7}$$

5. UF_6 enrichment monitor

In a transmission-based enrichment monitor, the transmitted beam gives the total amount of uranium present. The 186-keV radiation from the decay of ^{235}U gives the amount of ^{235}U present. The enrichment is calculated with the following formula:

$$E = K\frac{(R-B)}{\ln(I/I_0)} \tag{8}$$

where $E = {}^{235}U$ enrichment (in weight percentage) of the gas, K = calibration constant, $R = {}^{235}U$ 186-keV total count rate (gas + deposit + room background), $B = {}^{235}U$ 186-keV background count rate (deposit + room background), I = transmission count rate attenuated by the gas and pipe, and I_0 = transmission count rate attenuated by the empty pipe.

In the original design the pipe was steel and the transmission source was ^{57}Co(122 keV). The ratio I/I_o was approximately 0.97 because the attenuation in the gas only decreased the transmission by 0.03 compared to the attenuation in the steel pipe. Thus, the enrichment was very sensitive to small changes in I and I_0.

We implemented the ideas presented in this report in the design of a new enrichment monitor (Ianakiev et al., 2009c). The pipe is aluminum and the transmitting source is an X-ray tube with a silver filter. Fig. 6 shows a laboratory version of the new design, and Fig. 7 shows the spectrum with the ROIs indicated.

Fig. 8 is a demonstration of the capability of the system to track changes. The pressure of the UF_6 in the pipe was varied over a period of approximately one week, and the counting rate of the ^{57}Co 122-keV line and the ^{235}U 186-keV line were measured. The areas of the peaks were determined with the peak fitting code NaIGEM (Gunnink et al., 1997). The third chart shows the enrichment calculated with the Eq. (8). The bottom chart shows that the uncertainty in the enrichment is larger when the pressure is low, as expected.

Fig. 9 shows the enrichment monitor data over a period of one month. Based on these results, the rates of change of the 186-keV count rate, the transmission count rate, and the enrichment are -0.0034 ± 0.0015 counts/s/day, -0.0021 ± 0.0036 counts/s/day and $-0.00027 \pm 0.00012\%$/ day, respectively. These results do not include correction with the flux monitor data because it was deemed not necessary for our application. We believe that the enrichment monitor will show similar stability over much longer periods.

6. Conclusions

We have developed several new techniques to monitor material in pipes using transmission measurements. These include X-ray tubes with filters to produce essentially monoenergetic transmission beams, a flux monitor for X-ray beams, improved electronics for NaI(Tl) detectors, less expensive shielding, and calibration methods when an empty pipe measurement is not possible. We are applying these innovations to improve enrichment monitors for UF_6 gas in pipes. These innovations can be used for other industrial applications.

Acknowledgments

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References

European Synchrotron Radiation Facility website, 2006. 〈http://www.esrf.eu/UsersAndScience/Experiments/StructMaterials/ID11/ID11UserGuide/ID11Edges〉.

Goda, J.M., Ianakiev, K.D., Moss, C.E., 2009. Development of a model of an X-ray tube transmission source. In: Proceedings of the Conference Record of the IEEE Nuclear Science Symposium.

Gunnink, R., Arlt, R., Bernt, R., 1997. New Ge and NaI analysis methods for measuring ^{235}U enrichments. In: Proceedings of the 19th Annual ESARDA Symposium on European Safeguards Research and Development Association.

Hoeppner, C.H., 2009. Radiation densitometers. 4th ed. In: Liptak, B.G (Ed.), Instrument Engineers' Handbook—Process Measurement and Analysis, vol. 1. CRC Press, London.

Ianakiev, K.D., Alexandrov, B.S., Littlewood, P.B., Browne, M.C., 2009a. Temperature behavior of NaI(Tl) scintillation detectors. Nucl. Instrum Methods A 607, 432–438.

Ianakiev, K.D., et al., 2009b. Advanced technology for enrichment monitoring for gas centrifuge enrichment plants. J. Radioanal. Nucl. Chem. 282, 657–661.

Ianakiev, K.D., et al., 2009c. Advanced enrichment monitoring technology based on transmission measurements with an x-ray source and NaI(Tl) spectrometer. In: Proceedings of the First International Conference on Advancements in Nuclear Instrumentation Measurement Methods and their Applications (ANIMMA).

Johansen, G.A., Jackson, P., 2004. Radioisotope Gauges for Industrial Process Measurements. John Wiley & Sons, Ltd., Chichester, ISBN: 9780470021088.

Kelly, D.A., Boyes, W., 2009. Measurements employing nuclear techniques. In: Boyes, W. (Ed.), Instrumentation Reference Book, 4th ed. Butterworth-Heinemann, Burlington, pp. 549–565.

Knoll, G.F., 2010. Radiation Detection and Measurement, Fourth ed. John Wiley & Sons, Hoboken.

Lombardi, M.L., Favalli, A., Goda, J.M., Ianakiev, K.D., Moss, C.E., 2011. Flux monitor diode radiation hardness testing. Nucl. Instrum. Methods A 652, 112–115.

Lombardi, M.L., Favalli, A., Goda, J.M., Ianakiev, K.D., McArthur, D.W., Moss, C.E., 2012. Experimental determination of the thickness of aluminum cascade pipes in the presence of UF_6 gas during enrichment measurements. Nucl. Instrum. Methods A 672, 69–74.

Moss, C.E., Close, D.A., Johnson, W.S., Kerr, P.L., Romero, C.D., 1999. In: Proceedings of 4th Topical Meeting on Industrial Radiation and Radioisotope Measurement Applications (IRRMA-4), Raleigh, NC.

Tjugum, S., Frieling, J., Johansen, G.A., 2002. A compact low energy multibeam gamma-ray densitometer for pipe-flow measurements. Nucl. Instrum Methods B 197, 301–309.

Tjugum, S., Mihalca, R., 2009. X-ray based densitometer for multiphase flow measurement. In: Proceedings of the 27th International North Sea Flow Measurement Workshop, Tonsberg, Norway.