Nuclear Inst. and Methods in Physics Research, A 877 (2018) 138–142

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

# Multi-detector system approach for unattended uranium enrichment monitoring at gas centrifuge enrichment plants

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ARTICLE INFO

*Keywords:*
Uranium enrichment monitor
UF6 source
Gas centrifuge enrichment plant

ABSTRACT

Improving the quality of safeguards measurements at Gas Centrifuge Enrichment Plants while reducing the inspection effort is an important objective given the number of existing and new plants that need to be safeguarded. A useful tool in many safeguards approaches is the on-line monitoring of enrichment in process pipes. One requirement of such a monitor is a simple, reliable and precise passive measurement of the 186-keV line from $^{235}U$. The other information required is the amount of gas in the pipe, which can be obtained by a transmission or pressure measurement. We describe our research to develop such a passive measurement system. Unfortunately, a complication arises in the interpretation of the gamma measurements, from the contribution of uranium deposits on the wall of the pipe to the 186-keV peak. A multi-detector approach to address this complication is presented where two measurements, one with signal primarily from gas and one with signal primarily from deposits, are performed simultaneously with different detectors and geometries. This allows a correction to be made to the 186-keV peak for the contribution from the deposit. We present the design of the multi-detector system and the results of the experimental calibration of the proof-of-principle prototype built at LANL.

Published by Elsevier B.V.

## 1. Introduction

Improving the quality of safeguards measurements at Gas Centrifuge Enrichment Plants (GCEPs) while reducing the inspection effort is an important objective given the number of existing and new plants that need to be safeguarded. A useful tool in many safeguards approaches is the on-line monitoring of enrichment in process pipes. One requirement of such a monitor is a simple, reliable and precise passive measurement of the 186-keV line from $^{235}U$. The other information required is the amount of gas in the pipe, which can be obtained by a transmission or pressure measurement. We describe our research to develop such a passive measurement system [1–7]

Unfortunately, a complication arises in the interpretation of the gamma measurements, from the contribution of uranium deposits on the wall of the pipe to the 186-keV peak [8] A multi-detector approach to overcome this complication is presented where two measurements, one with signal primarily from gas and one with signal primarily from deposits, are performed simultaneously with different detectors and geometries. This allows a correction to be made to the 186-keV peak for the contribution from the deposits. Our approach is based on NaI scintillation detectors: high length-to-diameter ratio longitudinal detectors for deposit measurements and larger diameter detectors for gas measurements. We have chosen to use scintillation detectors because they require low maintenance and are less expensive than HPGe semiconductor detectors. We present the design of the multi-detector system and the results of the experimental calibration of the proof-of-principle prototype built at LANL. In order to characterize the multi-detector system, in our laboratory we built a $UF_6$ source using a 50-cm long by 10.8-cm (4.25-inch) diameter pipe plated with thin deposits of $UO_2F_2$ that was 69% isotopically enriched in $^{235}U$. The plated source was characterized, in composition and thickness of uranium deposits, by means of Rutherford Backscattering Spectrometry (RBS) using witness coupons prepared under identical conditions. The source was used to evaluate the performance of the multi-detector system by changing the $UF_6$ gas pressure in steps from 0 to 65 Torr. The experimental results obtained are presented and discussed.

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https://doi.org/10.1016/j.nima.2017.09.033
Received 4 August 2017; Received in revised form 13 September 2017; Accepted 14 September 2017
Available online 4 October 2017
0168-9002/Published by Elsevier B.V.

## 2. Fundamentals of deposit corrections method

### 2.1. Two-geometry approach

In this section we derive and discuss the so called two-geometry approach [7–9] for uranium deposit corrections, and how the method is extended for the design of our multi-detector system.

The count rates measured in the 186-keV gamma peak are a function of the $UF_6$ enrichment in the pipe, of internal radius $R$, and can be expressed as following:

$$U_{186} = \tilde{\rho}_{UF6} \cdot p_{UF6} \cdot \pi R^2 \cdot f \cdot E_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^G_{186} + D_U \cdot f' \cdot E'_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^D_{186} \quad (1)$$

where:

$U_{186}$ is the count rate in the 186-keV gamma peak [c/s];

$\tilde{\rho}_{UF6}$ is the $UF_6$ density per unit of pressure [g/cm$^3$/Torr];

$p_{UF6}$ is the pressure of the $UF_6$ gas [Torr];

$\pi R^2$, is the area of internal cross section of the pipe, $R$ is the internal radius [cm];

$f$, $f'$ are the mass fraction of uranium per $UF_6$ unit mass [g of U/g of $UF_6$], and per $UO_2F_2$ unit mass of deposit [g of U/g of $UO_2F_2$], respectively.

$E_{w-U235}$, $E'_{w-U235}$ are the enrichments expressed as the weight fraction in the $UF_6$ gas and in the uranium deposits, respectively [g of $^{235}$U/g of U];

$r_{U-235}$ is the $^{235}$U specific activity ($\gamma$ per unit mass per second) of the 186-keV gamma ray [g$^{-1}$·s$^{-1}$], see [10]

$D_U$ is the mass of the uranium deposited on the wall of the pipe per unit of length [g/cm];

$L_{eff}$ is the effective pipe length [cm]; this length is less than the actual length of the pipe and is related to the efficiency of the detector;

$\varepsilon^G_{186}$ is the full peak efficiency of the detector at the 186-keV gamma ray line for the $UF_6$ gas. This value is also a function of the length of the pipe;

$\varepsilon^D_{186}$ is the full peak efficiency of the detector at the 186-keV gamma ray line for the uranium deposits on the wall. This value is also a function of the length of the pipe.

In Eq. (1) we can identify the contributions to the $U_{186}$ signal due to $UF_6$ gas (G) and due to the uranium deposits (D):

$$U_{186} = \underbrace{\tilde{\rho}_{UF6} \cdot p_{UF6} \cdot \pi R^2 \cdot f \cdot E_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^G_{186}}_{G} + \underbrace{D_U \cdot f' \cdot E'_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^D_{186}}_{D}$$

And finally the equation reads:

$$U_{186} = G + D \quad (2)$$

An approach to identifying the two components $G$ and $D$ is the so-called two-geometry approach. In this method, two different collimator shapes (for example a wide-$w$, and a narrow-$n$) are introduced causing different detection efficiencies. With this approach Eq. (2) becomes:

$$\begin{cases} U^w_{186} = G + D \\ U^n_{186} = \alpha G + \beta D \end{cases} \quad (3)$$

where $\alpha$ and $\beta$ are the relative efficiencies for detecting the 186-keV line in the two geometries for the gas and deposits respectively. The two key assumptions of the two-geometry approach are: the deposit is uniformly deposited around the wall of the pipe, and the $UF_6$ gas density in the pipe is uniform: The values of $\alpha$ and $\beta$ are constant for a given pipe geometry (pipe diameter and wall thickness) and material, and detector systems set-up and can be determined in a laboratory for a given class of header pipes [8,11]. The deposit thickness on the inner surface of the pipe is of the order of sub-μm (see section "$UF_6$ source plated with deposit" in this paper) and the self-shielding effect of the 186-keV is in general negligible. [8]. For $\alpha \neq \beta$, the equation system in (3) results in solutions that can be solved for $G$ and $D$, and the contribution from the gas and deposit are then given by:

$$G = \frac{U^n_{186} - \beta \cdot U^w_{186}}{\alpha - \beta} \quad (4)$$

$$D = \frac{\alpha \cdot U^w_{186} - U^n_{186}}{\alpha - \beta} \quad (5)$$

By determining the amount of gas in the pipe, which can be obtained by an X-ray transmission or knowing the pressure, the $UF_6$ gas enrichment is obtained [1].

This approach can be generalized by introducing two different geometry measurements. The generalization can also be achieved by using two different detector systems. One configuration must be very sensitive to $UF_6$ pressure in comparison with the signal coming from the deposits (gas-sensing configuration). The second configuration should have very low sensitivity to $UF_6$ gas pressure while still keeping a good efficiency for the deposit signal (deposit-sensing configuration). The possibility that the two configurations can be used simultaneously is an advantage of using this method.

In the generalized method the Eqs. (3) can be written as following:

$$\begin{cases} U^1{}_{186} = \overbrace{\tilde{\rho}_{UF6} \cdot \pi R^2 \cdot f \cdot E_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^G_1 \cdot p_{UF6}}^{G} \\ \quad + \overbrace{D_U \cdot f' \cdot E'_{w-U235} \cdot r_{U-235} \cdot L_{eff} \cdot \varepsilon^D_1}^{D} \\ U^2{}_{186} = \underbrace{\tilde{\rho}_{UF6} \cdot \pi R^2 \cdot f \cdot E_{w-U235} \cdot r_{U-235} \cdot L'_{eff} \cdot \varepsilon^G_2 \cdot p_{UF6}}_{\alpha G} \\ \quad + \underbrace{D_U \cdot f' \cdot E'_{w-U235} \cdot r_{U-235} \cdot L'_{eff} \cdot \varepsilon^D_2}_{\beta D} \end{cases} \quad (6)$$

Eqs. (6) show the linear behavior of the count rate as function of the $UF_6$ pressure for both the configurations; slopes are proportional to uranium enrichment in the pipe. The factors $L_{eff} \cdot \varepsilon^G_1$ and $L'_{eff} \cdot \varepsilon^G_2$ weight the slopes in the two configurations, so they weight the different sensitivities to the $UF_6$ pressure in the two configurations. In the same way the factors $L_{eff} \cdot \varepsilon^D_1$ and $L'_{eff} \cdot \varepsilon^D_2$ weight the count rate in the deposits. Here it is worth mentioning that in the generalized two-geometry approach $L_{eff}$ and $L'_{eff}$ could be different, because the two configurations (gas-sensing configuration and deposit-sensing configuration) have very different detector–collimator–geometry arrangements.

Comparing Eqs. (3) and (6) the values of $\alpha$ and $\beta$ are obtained and given by the following equations:

$$\alpha = \frac{L'_{eff} \cdot \varepsilon^G_2}{L_{eff} \cdot \varepsilon^G_1} = l\frac{\varepsilon^G_2}{\varepsilon^G_1}, \quad \beta = \frac{L'_{eff} \cdot \varepsilon^D_2}{L_{eff} \cdot \varepsilon^D_1} = l\frac{\varepsilon^D_2}{\varepsilon^D_1} \quad (7)$$

where $l$ is the ratio of the effective lengths.

Eq. (7) show that the values of $\alpha$ and $\beta$ are independent of the enrichment of the gas and deposits, so they can be obtained by calibration in a laboratory environment with a $UF_6$ pipe internal surface plated. This conclusion is still valid when the uranium deposits and $UF_6$ gas have different enrichments from each other. $\alpha$ coincides with the value extracted from the ratio of the slopes of the two Eqs. (6) and $\beta$ is obtained from the ratio of the count rate at zero $UF_6$ pressure. In the two-geometry generalized approach, it is worth mentioning that we are limited in the use of an HPGe detector, but scintillation detectors are satisfactory alternatives.

## 3. Multi-detector system

### 3.1. Development and design

Following the generalized two geometry approach, we designed a multi-detector system where high length-to-diameter longitudinal

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