APPLICATION OF THE DIFFERENTIAL PEAK ABSORPTION METHOD AS AN AUXILIARY TOOL IN ENRICHMENT MEASUREMENTS OF UF_6 CYLINDERS FOR SAFEGUARDS PURPOSES

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Abstract

The ABACC (Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials) uses the system MMCG (Mini Multi-Channel analyzer and hyper-pure Germanium detector) with the software MGAU in enrichment meter mode for the determination of enrichment of UF_6 cylinders during safeguards inspections. One of the important conditions that must be met for this measurement to be acceptable is the infinite- thickness of UF_6 in the position where the detector is located during measurement. This is called the Infinite-Thickness Condition (ITC). If this condition is not met, the results obtained by the inspector in the field will be incorrect. With the goal of helping the inspector in this procedure, ABACC is developing a software tool to analyze the spectra achieved, and to determine if the ITC is met. The method under development is based on the Differential Peak Absorption (DPA) technique, in which the relation of count rates measured for different gamma rays photopeaks coming from the same isotope depends on the thickness of the materials present between the isotope and the detector. Applying this technique, the relation between count rates at the photopeaks of ^{235}U in a spectrum obtained from an UF_6 cylinder can be compared with the expected value for the ITC. This article presents the theoretical study of the proposed method, and the laboratory tests performed in order to determine the relation between count rates for the photopeaks of ^{235}U in the ITC. The results produced with calibrated laboratory samples and the experiences reached in the field are presented, showing the efficiency of the proposed method as a tool to improve the quality of measurements in safeguards inspections.

Introduction

The DPA technique (Differential Peak Absorption) is based on the measurement of the count ratio of different γ energies, emitted by the same radionuclide. The difference between the measured value and the value at infinite thickness shows how much the signal has been attenuated while crossing through the sample. When the sample is homogeneous it is possible to determine the γ photopeaks attenuation and the sample thickness, if the auto-shielding is strong enough, and the radionuclide of interest emits γ rays of suitable intensity and energy difference. The greater the difference in energies, the more effective is the proposed technique.

When the enrichment meter method is used to measure enrichment, the result is valid only if the sample thickness is infinite in all directions as seen by the detector. This condition can be determined by means of the DPA technique, measuring the relative intensity of the γ rays coming from ^{235}U, because this nuclide emits γ radiation of different energies with acceptable intensities.

If this simple test can be performed by the inspector in the field after acquiring the spectrum, the quality of enrichment measurement can be improved, by assuring that proper measurement conditions are met in the place where the spectrum was taken.

Theory

Self absorption of gamma radiation

The intensity of photons transmitted through a sample of thickness x_s can be approximated by the following expression:

$$I_s = I_0\left[\frac{1-e^{(-m_s r_s x_s)}}{m_s r_s x_s}\right] \quad (1)$$

where:

I_s = Intensity of photons transmitted with no interaction in the sample

I_0 = Intensity of photons originated in the sample, that would reach the detector in absence of the self absorption

m_s = Sample mass attenuation coefficient (cm^2/g)

r_s = Sample apparent density (g/cm^3)

x_s = Sample thickness (cm)

In the case of a sample emitting photons of energies g_1 and g_2, contained in a recipient of thickness x_w , the intensity of photons of energies g_1 and g_2 transmitted with no interaction with the sample (*s*) and the container wall (*w*) is given by the following expressions, deduced from the expression (1):

$$I_{g1} = k_{g1}\left[\frac{1-e^{(-m_{sg1} r_s x_s)}}{m_{sg1} r_s x_s}\right] \quad (2)$$

where, $k_{g1} = I_{0g1} e^{(-m_{wg1} r_w x_w)}$

$$I_{g2} = k_{g2}\left[\frac{1-e^{(-m_{sg2} r_s x_s)}}{m_{sg2} r_s x_s}\right] \quad (3)$$

where, $k_{g2} = I_{0g2} e^{(-m_{wg2} r_w x_w)}$

In expressions (2) and (3), I_{0g} represents the intensity of photons of energies g that would be observed by the detector in absence of the container wall; so, the exponential factor in the expression for k_g represents the attenuation in the container wall.

The ratio of intensity of photons of energies g_1 and g_2 transmitted with no interaction with the sample and container wall can be obtained by dividing equations (2) and (3) member by member:

$$\frac{I_{g1}}{I_{g2}} = \frac{k_{g1} m_{sg2}}{k_{g2} m_{sg1}}\left[\frac{1-e^{(-m_{sg1} r_s x_s)}}{1-e^{(-m_{sg2} r_s x_s)}}\right] \quad (4)$$

We are interested in the ratio between the intensities of radiation detected for different energies, to the energy of 186 keV, in the asymptotic cases. Let's call that ratio as R. Taking the limit of equation (4) as x approaches 0, for g_2 = 186 keV, we get:

a) Disregarding the absorption in the container wall:

$$R_{g,0} = \lim_{x_s \to 0}\frac{I_g}{I_{186}} = \lim_{x_s \to 0}\frac{I_{0,g}\, m_{s,186}}{I_{0,186}\, m_{sg}}\left[\frac{1-e^{(-m_{sg} r_s x_s)}}{1-e^{(-m_{s,186} r_s x_s)}}\right] = \frac{I_{0,g}\, m_{s,186}}{I_{0,186}\, m_{sg}}\left[\frac{m_{sg}\, r_s}{m_{s,186}\, r_s}\right] = \frac{I_{0,g}}{I_{0,186}}$$

b) Taking into account the absorption in the container wall:

$$R_{gw,0} = \lim_{x_s \to 0} \frac{I_g}{I_{186}} = \lim_{x_s \to 0} \frac{k_g m_{s,186}}{k_{186} m_{sg}} \left[\frac{1 - e^{(-m_{sg} r_s x_s)}}{1 - e^{(-m_{s,186} r_s x_s)}} \right] = \frac{k_g m_{s,186}}{k_{186} m_{sg}} \left[\frac{m_{sg} r_s}{m_{s,186} r_s)} \right] = \frac{k_g}{k_{186}}$$

Taking the limit of equation (4) as x approaches ∞, for g_2 = 186 keV, we get:

a) Disregarding the absorption in the container wall:

$$R_{g,\infty} = \lim_{x_s \to \infty} \frac{I_g}{I_{186}} = \lim_{x \to 0} \frac{I_{0,g} m_{s,186}}{I_{0,186} m_{sg}} \left[\frac{1 - e^{(-m_{sg} r_s x_s)}}{1 - e^{(-m_{s,186} r_s x_s)}} \right] = \frac{I_{0,g} m_{s,186}}{I_{0,186} m_{sg}}$$

b) Taking into account the absorption in the container wall:

$$R_{gw,\infty} = \lim_{x_s \to \infty} \frac{I_g}{I_{186}} = \lim_{x_s \to \infty} \frac{k_g m_{s,186}}{k_{186} m_{sg}} \left[\frac{1 - e^{(-m_{sg} r_s x_s)}}{1 - e^{(-m_{s,186} r_s x_s)}} \right] = \frac{k_g m_{s,186}}{k_{186} m_{sg}}$$

In the table 1 are shown the relevant parameters for U_3O_8, and Al + Ni as material of the container wall.

Mass attenuation coefficient (cm^2/g)				
Eg (keV)	**I_{0g}**	m_g **U_3O_8**	m_g **Ni**	**k_g (2mm Al + 6 mm Ni)**
143.8	*0.10962*	*2.431*	*0.2328*	*0.0308*
185.7	*0.5724*	*1.346*	*0.1714*	*0.2208*
205.3	*0.050112*	*1.051*	*0.1750*	*0.0190*

Table 1: Some relevant mass attenuation coefficients

With the values of table 1, and the expressions obtained above, we can build the table 2, showing the theoretical values of peak ratios with respect to E*g* = 186 keV:

E*g* [keV]	**R**	**No Containment**		**Attenuator: 6 mm Ni + 2 mm Al**	
143.8	R_o	$I_{0,143} / I_{0,186}$	*0.192*		
	R_∞	$(I_{0,143}.\mu_{186})/(I_{0,186}.\mu_{143})$	*0.106*		
	R_o			k_{143} / k_{186}	*0.139*
	R_∞			$(k_{143}.\mu_{186})/(k_{186}.\mu_{143})$	*0.077*
205.3	R_o	$I_{0,205} / I_{0,186}$	*0.087*		
	R_∞	$(I_{0,205}.\mu_{186})/(I_{0,186}.\mu_{205})$	*0.112*		
	R_o			k_{205} / k_{186}	*0.086*
	R_∞			$(k_{205}.\mu_{186})/(k_{186}.\mu_{205})$	*0.110*

Table 2: Asymptotic peak ratios

For the computation of the values in table 2, the following parameters were also used:

$k_{143} = I_{0,143} e^{(-m_{Ni,143} r_{Ni} x_{Ni})} e^{(-m_{Al,143} r_{Al} x_{Al})}$ $\quad$ $k_{186} = I_{0,186} e^{(-m_{Ni,186} r_{Ni} x_{Ni})} e^{(-m_{Al,186} r_{Al} x_{Al})}$

$I_{0,143}$, $I_{0,186}$, $I_{0,205}$ = Probability of emission of γ_{143} , γ_{186} and γ_{205}

m_{143}, m_{186}, m_{205} = Mass attenuation coefficient of U_3O_8, for energies γ_{143}, γ_{186} and γ_{205}

$m_{Ni, 143}$, $m_{Ni, 186}$, $m_{Ni, 205}$ = Mass attenuation coefficient of Ni, for energies γ_{143}, γ_{186} and γ_{205}

$m_{Al, 143}$, $m_{Al, 186}$, $m_{Al, 205}$ = Mass attenuation coefficient of Al, for energies γ_{143}, γ_{186} and γ_{205}

r = Sample density (U_3O_8) = 2.52 g/cm^3 $\quad$ r_{Ni} = Ni density = 8.6 g/cm^3

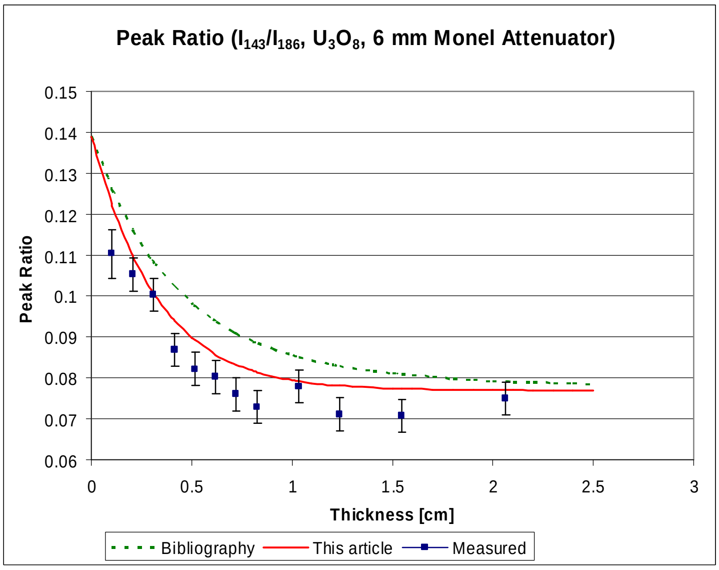
r_{Al} = Al density = 2.7 g/cm^3 $\quad$ x = Sample thickness (cm)

x_{Ni} = Ni wall thickness = 0.6 cm $\quad$ x_{Al} = Al wall thickness = 0.2 cm

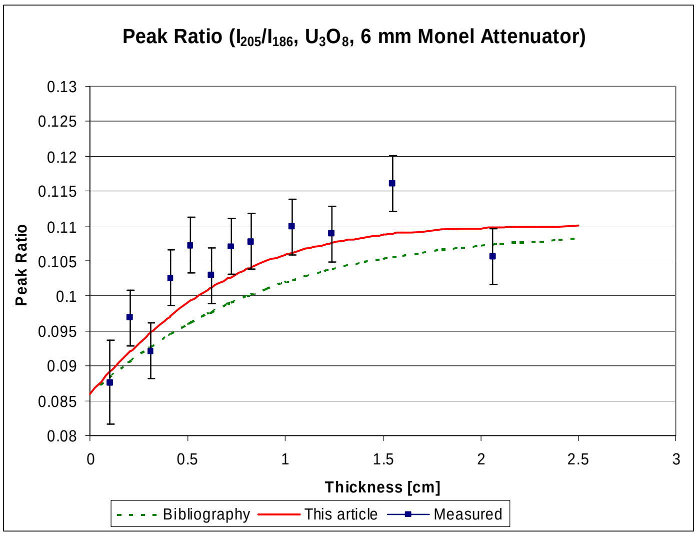
The results obtained here are similar to those produced by the expression given in the bibliography [1], that uses the following expression for the transmitted intensity of unscattered and unabsorbed gamma rays escaping from a sphere whose attenuation is characterized by $X = m_s r_s x_s$:

$$R_0 = I_0 \frac{3}{2X}\left[1 - \frac{2}{X^2} + e^{(-X)}\left(\frac{2}{X} + \frac{2}{X^2}\right)\right]$$

Figure 1 shows the comparison between both functions that describe peak ratio as a function of thickness, the one given by the bibliography and the expression proposed here. Peaks of 143 and 205 keV are analyzed. There is a good equivalence on the expressions results, especially in the limit of infinite thickness. The values obtained in the measurements described below are also shown.



a) 143 keV



b) 205 keV

Figure 1: Comparison of photopeak ratio between bibliography, this article and measurements: a) for the photopeak of 143 keV. b) for the photopeak of 205 keV.

Experimental Tests

Three different experiments were performed, in order to confirm the validity of the proposed theoretical analysis.

1) Several samples of U_3O_8, each one with different thickness, contained in a 70 mm diameter CBNM recipient were analyzed. Some spectra were obtained with no attenuator and others with a 6 mm Monel attenuator. The gamma spectrum was acquired for each sample, using an HPGe detector, in order to confirm the behavior of the photopeak ratios for the energies of interest. Also the enrichment was computed to confirm its variation as a function of the sample thickness. The procedure can be summarized as follows:

    a. Aliquots of 10 g of U_3O_8 powder were added to the 70 mm diameter recipient, and uniformly distributed. The computed thickness was 0.103 cm/10 g.

    b. The spectrum was registered for each added aliquot. Counting times of 1000 s, 2000 s, 5000 s and 10000 s were used. The measurement was done by using a HPGe detector, with 6 mm thickness Monel attenuator.

    c. The net areas under the main Eγ peaks were determined (143, 186 and 205 keV) using the code WinSPEC, and the enrichment was computed by using MGAU-EM. Figure 2 shows the results of the enrichment measurement, confirming the importance of the infinite thickness condition to get reliable results.

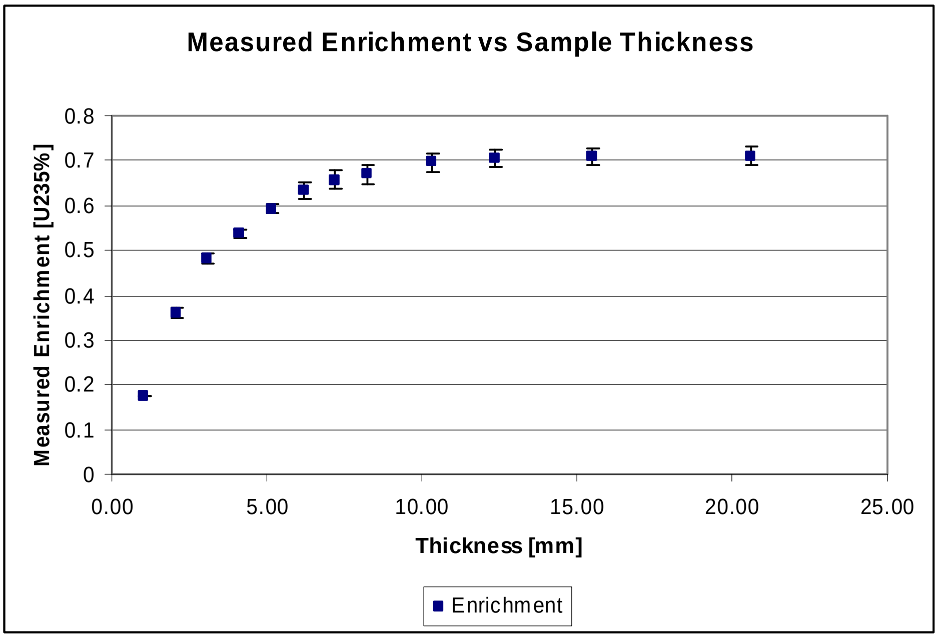


Figure 2: Measured enrichment of Natural U, for different sample thickness.

2) Several standard samples of different enrichments were measured, with the same attenuator. The spectra were obtained using WinSPEC, the areas under the main photopeaks were calculated, and the enrichment and peak ratios were also evaluated. These measurements were done to verify that the photopeak ratio is independent of the sample enrichment.
3) Finally, several spectra obtained during safeguards inspection to measure the enrichment of UF_6 cylinders (Depleted, Natural and Low-Enriched U), were analyzed. The ratio between

photopeak areas in the energies of 205 and 186 keV were calculated (let's call this value *H*), and compared against the expected theoretical infinite-thickness value (called *T*). The goal was to verify if these "real" measurements were performed under the required condition of infinite thickness.

It can be observed in figure 3 that some points are far from the expected value of $H/T = 1$, showing that the infinite-thickness condition was not met for that particular measurement.

Results

The results of the measurements of photopeaks ratio for different sample thickness, as described in preceding paragraphs, are summarized in figure 1. Those data show the applicability of the DPA technique for the estimation of the thickness of a homogeneous sample of Uranium materials (UO_2, UF_6, U_3O_8, etc.) from the spectra obtained with a high resolution gamma detector, using times acceptable to be used during safeguards inspections. The main gamma rays emitted by ^{235}U, even when near in energy, can be used to apply this technique.

By observing the values in the table 2, it can be seen that the ratio R_{143} and R_{205} approaches 0.195 and 0.087 respectively, as the thickness approaches 0 (zero). On the other hand, this ratio decreases to the 54% of that value as the thickness approaches infinite, for R_{143}, and increases by 28% for R_{205}. Even when the ratios between photopeaks are affected by the thickness of the container, the percent change of the ratio from zero thickness to infinite is not significantly affected. This important difference in the peak ratio with the thickness of the sample makes possible the application of the DPA technique for ^{235}U.

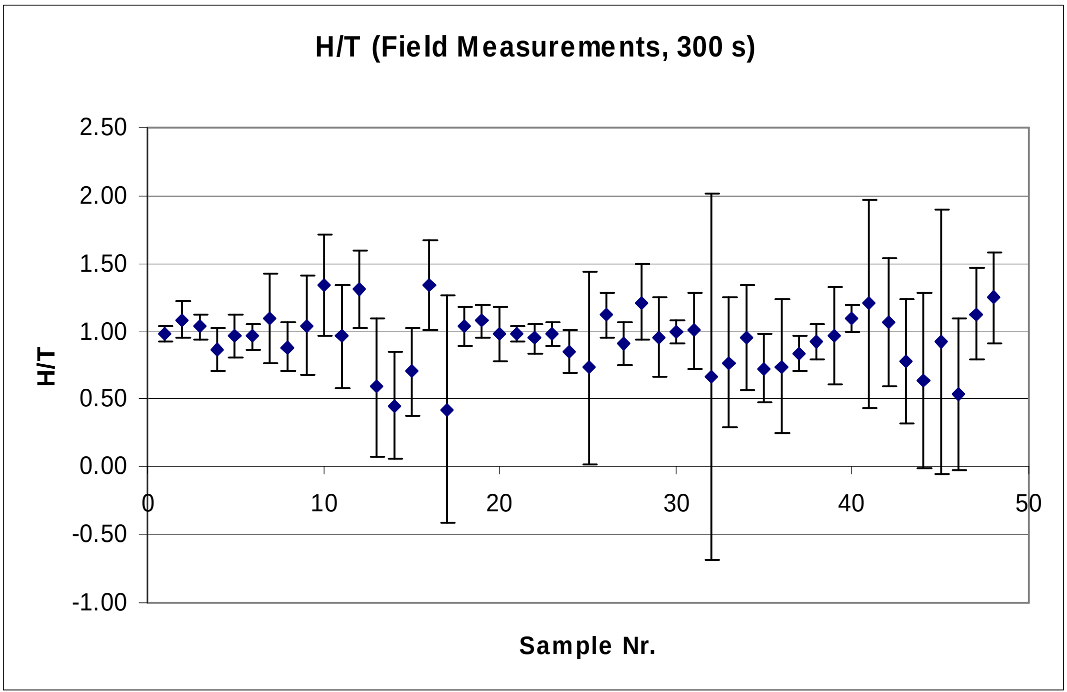


Figure 3: Infinite-thickness deviation of field measurements.

Table 3 shows the relation $R/R_{¥}$ varies with the sample thickness approaching a value of 1 when the thickness approaches infinite.

	$R/R_{¥}$ when R à $R_{¥}$	s $R/R_{¥}$
I_{143}/I_{186}	*0.98*	*0.04*
I_{205}/I_{186}	*0.99*	*0.016*

Table 3: Asymptotic peak ratios

The statistical error in the measurement of the net area under the γ photo-peak is related to the probability of emission, the measurement time, and the attenuation of γ photopeaks in the sample matrix and the container wall.

	sA% (mean value, 1-20 mm thickness)		
Measurement time [s]	**143.7 keV**	**185.7 keV**	**205.8 keV**
1000	*13.4*	*1.4*	*10.6*
2000	*9.6*	*1.0*	*7.5*
5000	*5.7*	*0.6*	*4.5*
10000	*4.3*	*0.4*	*3.3*

Table 4: Asymptotic peak ratios

Table 4 shows the relative error of the main ^{235}U photopeaks ($\sigma A_{143}\%$, $\sigma A_{186}\%$ y $\sigma A_{205}\%$) for measurement times from 1000 to 10000 s. These values were used in the computation of the standard deviation of the ratio R_{γ}, for different measurement times and sample thicknesses ($sR_{(143/186)}$ and $sR_{(205/186)}$).

It was observed that, even with measurement times as low as 1000 s, the statistical error in the relations $R_{(143/186)}$ and $R_{(205/186)}$ is low enough to accurately estimate the thickness of the sample.

On the other hand, the photopeak ratios for samples with infinite thickness do not vary with the enrichment, being constrained in the statistical error $sR_{¥}$. The obtained values are shown in table 5.

Peak	$R_{¥}$	$sR_{¥}$	$sR_{¥}$ %
143/186	*0.074*	*0.004*	*5*
205/186	*0.113*	*0.003*	*3*

Table 5: Asymptotic peak ratios

Proposed Analysis Procedure

In view of the obtained results, the DPA analysis proposed in this work to verify the infinite-thickness condition of different uranium materials with homogeneous matrix (oxides, fluorides, etc.) can be divided in the following steps:

a) Spectrum acquisition, with high resolution gamma detector (HPGe);
b) Analysis of the spectrum, with a software designed to compute the net area under the photopeaks of interest, with the corresponding uncertainty;
c) Computation of the relative peak ratios $R_{(143/186)}$ and $R_{(205/186)}$, with the corresponding error;
d) Comparison of the obtained value, with the expected value for the infinite thickness condition.

It should be taken into account that very short counting times with cylinders containing depleted or natural uranium can produce a very poor spectrum, with small photopeaks in the region of interest, and the results of the calculation proposed here can be inadequate. In that case, a larger counting time is advisable to get a more reliable conclusion.

Conclusion

The obtained results confirm the applicability of DPA technique to verify if the infinite thickness condition is met in the place where the spectrum was acquired for the measurement of enrichment of a UF_6 cylinder, thus improving the quality of measurements done in the field during safeguards inspections.

The next step is the automation of this analysis by the development of a computer program capable to determine if the conditions for enrichment computation are good enough, and advice the inspector if those conditions are not met.

Bibliography

[1] Passive Nondestructive Assay of Nuclear Materials. NUREG/CR-5550 LA-UR-90-732, Pgs. 161-164.