

## **True coincidence summing corrections for an extended energy range HPGe detector**

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### **Abstract**

True coincidence summing (TCS) effect for natural radioactive families of U-238 and Th-232 represents a problem when an environmental sample with a close source-detector geometry measurement is performed. By using a certified multi-nuclide standard source to calibrate an energy extended range (XtRa) HPGe detector, it is possible to obtain an intensity spectrum slightly affected by the TCS effect with energies from 46 to 1836 keV. In this work, the equations and some other considerations required to calculate the TCS correction factor for isotopes of natural radioactive chains are described. It is projected a validation of the calibration, performed with the IAEA-CU-2006-03 samples (soil and water).

Gamma ray spectrometry is a technique widely used for quantitative and qualitative characterization of samples with radioactive content, and the most common tool for this purpose are Ge detectors. In this process, if two or more  $\gamma$ -rays (or a  $\gamma$ -ray and an X-ray) are emitted in cascade from an excited nucleus, there is a certain probability that more than one of these will be detected simultaneously within the resolving time of the detector. This is true coincidence summing effect (TCS) [1].

The detector treats those photons as a single event and the effect takes two forms: "summing in" and "summing out". "Summing out" occurs when a  $\gamma$ -ray that should have been recorded in the full-energy peak (FEP), is recorded elsewhere,

due to the simultaneous detection and summing with another photon. "Summing in" is the reverse, when an event is added to the FEP by the simultaneous detection of two lower energy photons. All this results in an inaccurate value of the count rate and hence erroneous results for the efficiency values [1-3]. There are a few points to be taken into account in  $\gamma$ -ray spectrometry about TCS [1]:

- The effect is inversely proportional to the distance between source and detector.
- It gets worse with large detectors and is worst of all when using a well detector.
- It may be worse if a detector with a thin window is used, because the X-rays (which contribute to the summing) will not be absorbed.
- TCS can be expected when nuclides with a complex decay scheme

The objective of this study is to present the TCS correction factors equations and some considerations required when using standard calibration sources for an extended range HPGe detector, as well as the measurement of environmental sources containing the decay series of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ .

## **Method**

### **Efficiency Calibration**

Not all radiation reaching the detector produces a pulse. For  $\gamma$  spectrometry applications, it is very important to relate each peak area in the spectrum (generated by the study sample) to the amount of radioactivity it represents (a corresponding activity value in  $\mu\text{Ci}$ ,  $\text{kBq}$ , etc), by performing an efficiency calibration. Therefore, it is

required to know the absolute full energy peak efficiency. The equation for FEP efficiency EFEP is given by are measured.

$$\varepsilon_{FEP} = \frac{A_{net}}{A_{known} \cdot t \cdot I_{\gamma}} \cdot \prod_{i=1}^n k_i \quad (1)$$

Where  $A_{net}$  is the net area under the FEP;  $A_{known}$  represents the known activity of the source;  $t$  is the measuring time;  $I_{\gamma}$  is the probability of emission of the particular  $\gamma$ -ray being measured; and  $k_i$  represents several correction factors (depending on the particular case).

In order to perform the efficiency calibration, since the amount of decays per unit of time that the system can detect is closely related with the measurement geometry (source configuration as shown in Fig. 1, and the distance between the source and the detector), then an efficiency calibration for every type of measurement is required [1, 4]. Nevertheless, it is not possible to have standard sources for the infinite arrangements of geometry, density, composition, etc. to calibrate the equipment. A valid calibration curve is only measured under close geometry conditions, by using TCS-free radionuclides (list shown in Table 1, see Fig. 2 and Fig. 3) [1].

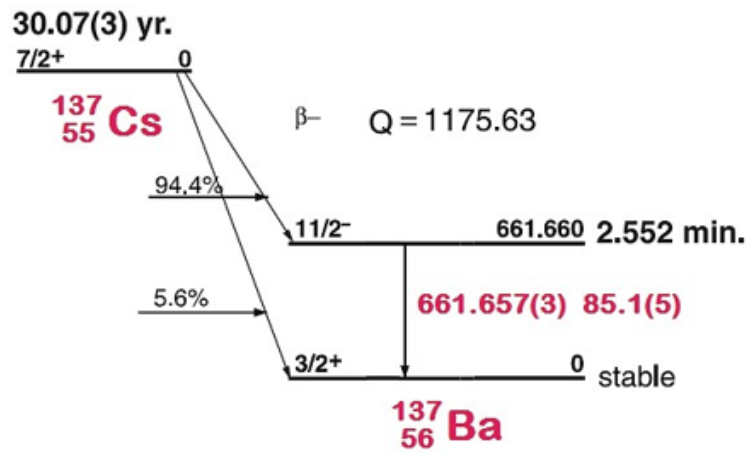


(a)

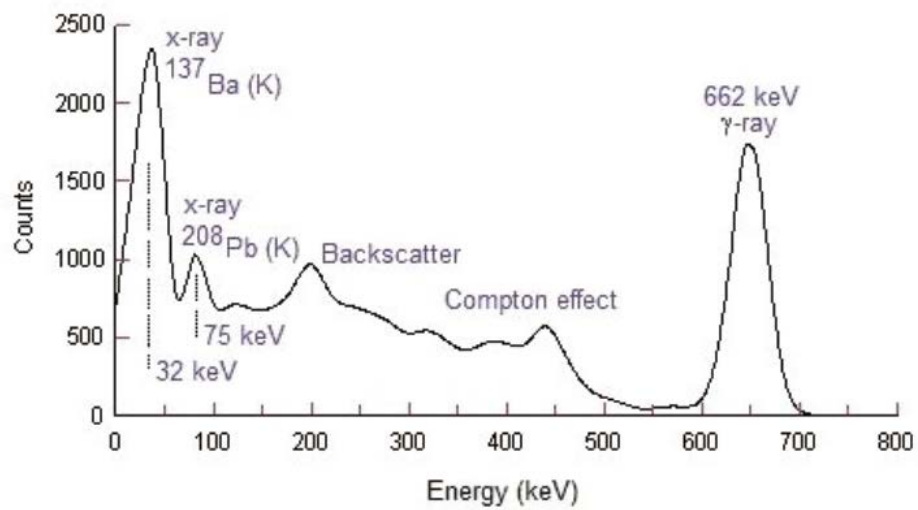


(b)

**FIGURE 1.** Different geometries for several sources. (a) Geometries for Petri dishes with different volumes. (b) Standard source.



**FIGURE 2.** Decay scheme for TCS-free  $^{137}\text{Cs}$  [5]



**FIGURE 3.**  $^{137}\text{Cs}$  spectrum [6].

TABLE 1. Radionuclides suitable for close geometry efficiency calibrations [1].

Radionuclide	$\gamma$ -ray energy (keV)	Nuclide type <sup>a</sup>	Standard available <sup>b</sup>
<sup>7</sup> Be	477.60	S	Y
<sup>40</sup> K	1460.82	S	(Y)
<sup>42</sup> K	1524.67	M	Y
<sup>51</sup> Cr	320.08	SX	Y
<sup>54</sup> Mn	834.84	SX	Y
<sup>57</sup> Co	122.06, 136.47	MX	Y
<sup>64</sup> Cu	1345.77	SX	Y
<sup>65</sup> Zn	1115.54	S(X)	Y
<sup>95</sup> Zr	724.19, 756.73	M	-
<sup>95</sup> Nb	765.80	S	Y
<sup>103</sup> Ru	497.08	S	-
<sup>109</sup> Cd ( <sup>109m</sup> Ag)	88.03	S	Y
<sup>113</sup> Sn ( <sup>113m</sup> In)	391.70	M	Y
<sup>131</sup> I	364.49, 636.99	M	Y
<sup>137</sup> Cs	661.66	S	Y
<sup>139</sup> Ce	165.86	SX	Y
<sup>141</sup> Ce	145.44	S	Y
<sup>144</sup> Ce	133.52	M	Y
<sup>198</sup> Au	411.80	M	Y
<sup>203</sup> Hg	279.20	S	Y
<sup>210</sup> Pb	46.54	S	-
<sup>241</sup> Am	59.54	M	Y

<sup>a</sup> S indicates a nuclide emitting a single  $\gamma$ -ray.

M indicates a nuclide for which the  $\gamma$ -ray mentioned is the major one and has little coincidence summing.

X indicates that summing with the accompanying X-rays (or other low energy  $\gamma$ -rays may be a problem on thin-window or n-type detectors).

<sup>b</sup> Standards for these nuclides are available from radionuclide standard suppliers.

## True Coincidence Summing Correction Factors

The efficiency calibrations obtained with nuclides which do not suffer from TCS (Table 1) are used to determine the mathematical corrections for study samples containing isotopes with complex decay schemes. Besides, there are many multinuclide standard sources with a wide range of energies, including high energies, which contains some radionuclides with TCS problem (e.g. <sup>60</sup>Co) with a special interest when a detector is calibrated for environmental analysis. For this, the efficiency corrections must be applied.

Since there are two possible cases that generates deviations on the efficiency value (*summing in* and *summing out*), the correction factors for each energy must be greater than 1 (if summing out, to compensate the loss of counts) or smaller than 1 (if summing in), and can be obtained from equation (2):

$$k_{TCS} = \frac{n_o}{n'_o} \quad (2)$$

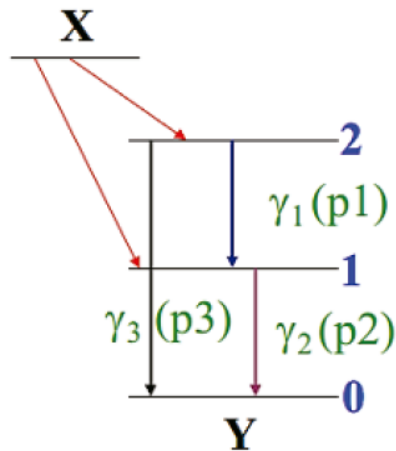
Where  $n_o$  is the count rate in the FEP, and  $n'_o$  is the net peak area of the  $\gamma$ -ray of interest, considering the loss of counts of that peak by summing with the  $i^{\text{th}}$  gamma ray.  $n_o$  is given by equation (3) whereas  $n'_o$  has three possibilities (according to Fig. 4): it may represent the net peak area for  $\gamma_1$  in equation (4), which is a summing out event; the net peak area for  $\gamma_2$  in equation (5) (representing another summing out event) or the net peak area for  $\gamma_3$  in equation (6), which is, in fact, a summing in event [1].

$$n_o = Ap_o \varepsilon_o \quad (3)$$

$$n'_o = Ap_1 \varepsilon_1 - Ap_1 \varepsilon_1 \varepsilon_{t2} \quad (4)$$

$$n'_o = Ap_2 \varepsilon_2 - Ap_1 \varepsilon_2 \varepsilon_{t1} \quad (5)$$

$$n'_o = Ap_3 \varepsilon_3 + Ap_3 \varepsilon_1 \varepsilon_2 \quad (6)$$



**FIGURE 4.** Typical decay scheme of a nuclide X decaying to Y [6].

Where the subscript  $o$  indicates the gamma ray of interest (1, 2 or 3);  $A$  is the number of atoms decaying;  $p_i$  represents the probability of simultaneous emission of  $i^{\text{th}}$  gamma and the gamma ray of interest;  $E_{ti}$  is the total efficiency of  $i^{\text{th}}$  gamma ray; and  $E_i$  represents the probability of  $\gamma_i$  of being detected and appearing in the FEP [2]. Total efficiencies of equations (4), (5) and (6) can be replaced by TCS-free efficiencies obtained from nuclides of Table 1.

For example,  $^{60}\text{Co}$  decays by  $\beta$  to  $^{60}\text{Ni}$  and emits two gamma rays in cascade (Fig. 5), 1173 ( $\gamma_1$ ) and 1332 ( $\gamma_2$ ) keV. These two gamma rays may reach the detector at the same time and can sum up to give counts at 2505 keV in the spectrum (Fig. 6). The loss in count rate for both peaks ( $\gamma_1$  and  $\gamma_2$ ) is a summing out effect.

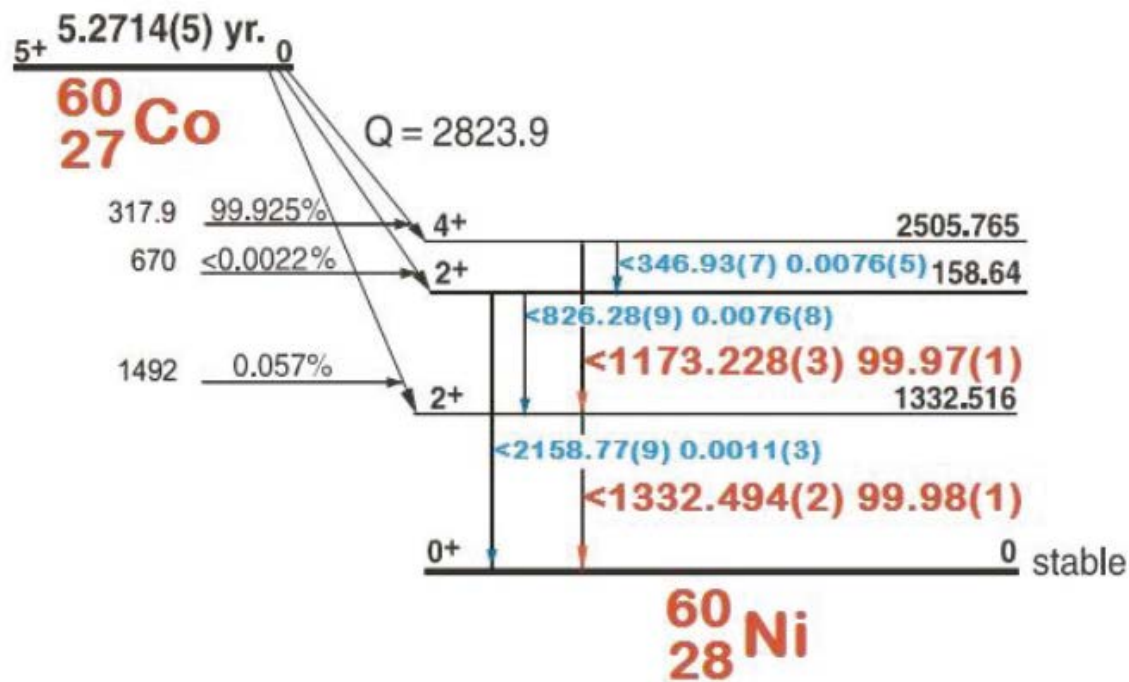


FIGURE 5.  $^{60}\text{Co}$  decay scheme [5].

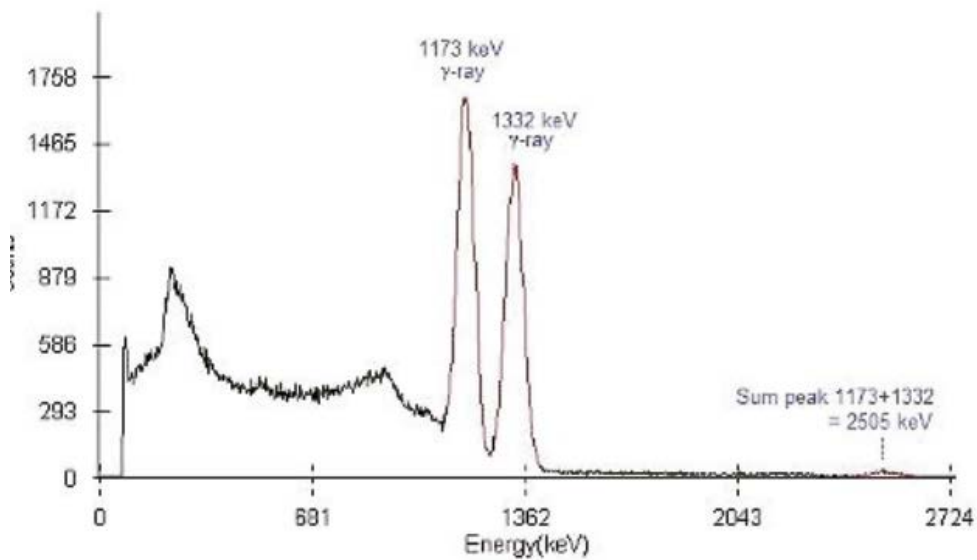


FIGURE 6.  $^{60}\text{Co}$  spectrum.



According to equations (2) to (6), the TCS correction factors for the two energy peaks of  $^{60}\text{Co}$  are the following:

$$k_{1173} = \frac{1}{1 - \varepsilon_{t1332}} \quad (7)$$

$$k_{1332} = \frac{1}{1 - \frac{p_{1173}}{p_{1332}} \varepsilon_{t1173}} \quad (8)$$

### Considerations for Natural Decay Series ( $^{235}\text{U}$ , $^{238}\text{U}$ and $^{232}\text{Th}$ )

The NORM (Naturally Occurring Radioactive Materials)  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series contain many radionuclides, but not all of them are measurable by gamma spectrometry, being either because their  $\gamma$  emissions are too weak or because they are emitted at very low energies. Of all the nuclides that can be measured, several of them have complicated decay schemes and TCS problems [1, 7].

For the  $^{235}\text{U}$  series there are potential true summing possibilities, although TCS effects on the  $\gamma$ -rays normally used to measure  $^{235}\text{U}$  are likely to be small [1]. However, it is suggested to choose the 143.76 and 163.33 keV peaks emitted by  $^{235}\text{U}$ , and both peaks from  $^{227}\text{Th}$ , of 235.96 and 256.23 keV, which are TCS-free [1, 7-10].

Regarding the  $^{238}\text{U}$  decay series, the emission of 63.28 keV of  $^{234}\text{Th}$  does not require any kind of correction. The 1001.03 keV peak of  $^{234\text{m}}\text{Pa}$  has a slight possibility of summing out with an energy of 43.5 keV, and a potential summing in due to the sum of the peaks of 258.26 and 742.81 keV, but the emission probability of those coincident energies is very low. From  $^{214}\text{Pb}$ , there are two suitable  $\gamma$ -rays for measuring, 351.93 and 295.22 keV, respectively, although the latter has an insignificant interference from  $^{212}\text{Bi}$ . Also, the 1764.49 keV peak of  $^{214}\text{Bi}$  is a coincidence-free  $\gamma$ -ray [1, 7-10].

Finally,  $^{232}\text{Th}$  series is the only one that has TCS effects with all their nuclides and the correction factors for the most probable  $\gamma$  emissions of  $^{228}\text{Ac}$  and  $^{208}\text{Tl}$  are required. For  $^{228}\text{Ac}$ , the  $\gamma$ -rays of 911.20 and 968.97 keV are the most significant and strongest lines [1, 6-8]. According to

equations (2) to (6) and the decay scheme of  $^{228}\text{Ac}$  (Fig. 7), the coincidence correction factors for each of those photons are the following:

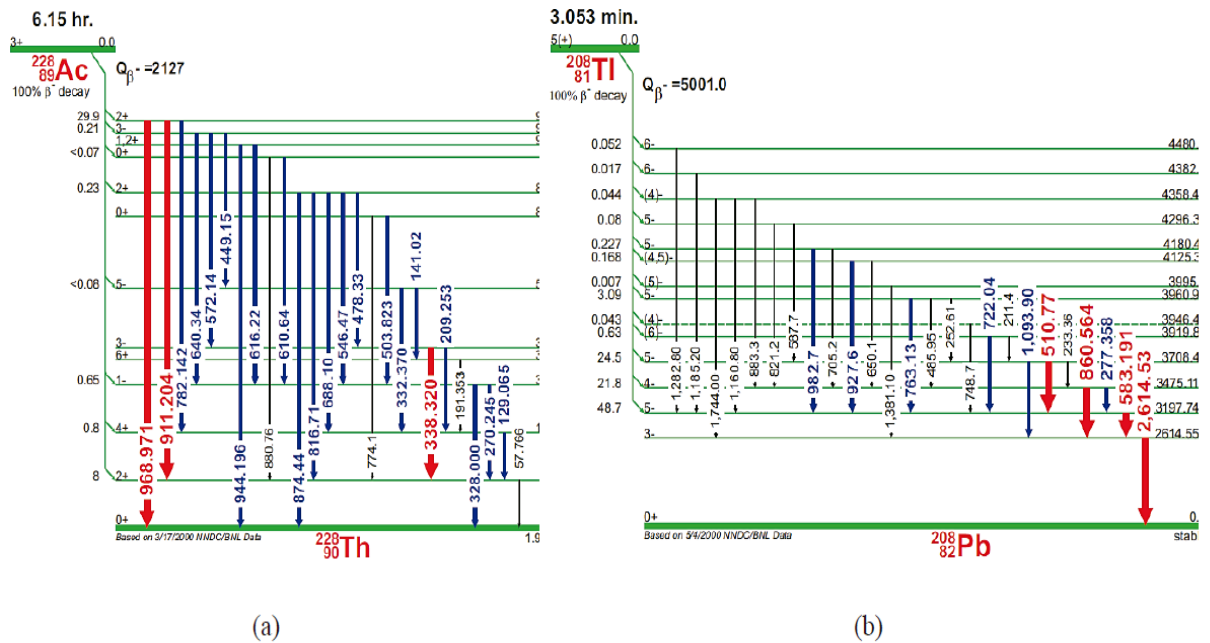
$$k_{911} = \frac{1}{1 - \epsilon_{t57.76}} \quad (9)$$

$$k_{968} = \frac{1}{1 + \frac{p_{911}\epsilon_{911}\epsilon_{57}p_{57}}{p_{968}\epsilon_{968}}} \quad (10)$$

The correction factors for both of the 583.19 and 860.56 keV peaks of  $^{208}\text{Tl}$  are given by equations (11) and (12), respectively.

$$k_{583} = \frac{1}{1 - (\epsilon_{t57.76} + \frac{p_{510}}{p_{583}}\epsilon_{t510})} \quad (11)$$

$$k_{860} = \frac{1}{1 - \epsilon_{t2614}} \quad (12)$$



**FIGURE 7.** (a) Decay scheme of  $^{228}\text{Ac}$ . (b) Decay scheme of  $^{208}\text{Tl}$ . Red arrows represent the most relevant lines that can be used to determine the activity, after being corrected for TCS [11, 12].

## Results and Discussion

### True Coincidence Summing

When measuring calibration sources and samples which suffer from TCS, the obtained results for the efficiency and, therefore, the activities are especially affected with closer source-to-detector geometries (see Fig. 8) and the solid angle subtended at the detector by the source [1, 13]. At the same time, working under those conditions may cause problems in nuclide identification (i.e. a pure sum peak is erroneously attributed to a nuclide not present in the sample or a nuclide is not recognized due to an incorrect match of the count rate) [14]. Nonetheless, there is always some degree of summing at any source-to-detector distance, but after a certain distance (depending upon the detector size) TCS effect will be negligible [1].



**FIGURE 8.**  $^{60}\text{Co}$  standard source being measured on the window of a CANBERRA™ XtRa HPGe detector.

Regarding TCS correction factors equations obtained for study  $\gamma$ -rays, the

total efficiency are needed for each peak (as already mentioned), but as Fig. 6 shows (taking  $^{60}\text{Co}$  as an example), there is no possibility to “separate” one peak at a time to calculate its efficiency, and then repeat the process with the other peak. For this reason, the use of Monte Carlo methods results very helpful. This method of calculation considers the “journey” that many thousands of gamma rays emitted from different parts of the source, interacting with different parts of the detector, might suffer. The program construct an efficiency curve but sometimes may just calculate correction factors to be applied to results from  $\gamma$ -rays measurements [1].

Just in a few cases, for very common matrix samples, there is a possibility to obtain efficiency values directly from the measurement by using standard sources sold by the International Atomic Energy Agency (IAEA), i.e. RGU-1, RGTh-1 and RGK-1.

### **Natural Decay Series**

Despite the fact that  $\gamma$ -ray spectrometry of NORM decay series is difficult, there are several ways to obtain accurate results. Both uranium decay chains can be considered as TCS-free, and even when is recommended use only a few lines of some nuclides to perform measurements, if the proper corrections are applied (e.g. for spectral interferences), then is possible to use any energy peak.

According to the intensity of the peak, it may be better to accept a lower-intensity peak free of summing rather than a higher-intensity alternative that would have to be corrected for TCS. However, if there is no option to the summing, like when working with the  $^{232}\text{Th}$  series, it is very important to calculate the summing correction factors for the required energies.

## Conclusions

True Coincidence Summing is a very important effect and must be taken into account when calibrating and performing measurements of sources containing nuclides with complex decay schemes in non-ideal source-to-detector geometrical conditions. By moving the source a relatively small distance away from the detector will achieve a large decrease in TCS with only a small increase in MDA (minimum detectable activity) [1]. Nevertheless, in case the summing effects are inevitable, there will be necessary to obtain correction factors for each energy involved, determining if the case is summing in or summing out from the decay scheme of the corresponding nuclide.

For both the  $^{235}\text{U}$  and  $^{238}\text{U}$  decay series, there is no need to calculate TCS correction factors for some nuclides, which are suitable for being measured, whereas the  $^{232}\text{Th}$  series requires, with no exception, the calculation of TCS correction factors for all their energy lines.

## ACKNOWLEDGMENTS

The support given by Centro de Investigación en Materiales Avanzados (CIMAV) and Consejo Nacional de Ciencia y Tecnología (CONACYT) for the financial support.

## References

- [1] G. Gilmore, *Practical Gamma-ray Spectrometry*. Chapters 7-16. Wiley, Chichester , (2008).
- [2] C. Agarwal *et al.*, J. Radioanal. Nucl. Chem. **289**, 773–780 (2011).
- [3] R.C., McFarland, Radioact. Radiochem. **1**, 12-14 (1990).
- [4] Genie2000 Tutorials Manual, Canberra Industries, Inc. 6 [www.laraweb.free.fr](http://www.laraweb.free.fr) (2004).
- [5] R.L. Heath, “Scintillation Spectrometry Gamma-Ray Spectrum Catalogue” Phillips Petroleum Company (1964).
- [6] Emission Spectrum of <sup>137</sup>Cs,  
<http://www.physics.fsu.edu/users/ng/courses/phy2054c/Labs/Expt10/Expt-10.htm>,  
February 20<sup>th</sup> (2015).
- [7] Chapter 6: True Coincidence Summing Corrections,  
[http://shodhganga.inflibnet.ac.in/bitstream/10603/4710/15/15\\_chapter%206.pdf](http://shodhganga.inflibnet.ac.in/bitstream/10603/4710/15/15_chapter%206.pdf),  
February 3<sup>rd</sup> (2015).
- [8] M. García-Talavera *et al.*, Appl. Radiat. Isot. **54**, 769-776 (2001).
- [9] M. García-Talavera *et al.*, Appl. Radiat. Isot. **59**, 165-173 (2003).
- [10] E. Şahiner *et al.*, Radiat. Phys. Chem. **96**, 165-173 (2014).
- [11] H. Yücel *et al.*, Appl. Radiat. Isot. **69**, 890-897 (2011).
- [12] <sup>228</sup>Ac Decay Scheme,  
[http://www.radiochemistry.org/periodictable/gamma\\_spectra/pdf/th232.pdf](http://www.radiochemistry.org/periodictable/gamma_spectra/pdf/th232.pdf).,  
February 27<sup>th</sup> (2015).
- [13] <sup>208</sup>Tl Decay Scheme,  
[http://www.radiochemistry.org/periodictable/gamma\\_spectra/pdf/tl208.pdf](http://www.radiochemistry.org/periodictable/gamma_spectra/pdf/tl208.pdf), February 27<sup>th</sup>  
(2015).
- [14] H.V. Klapdor-Kleingrothaus *et al.*, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, **511**, 335-340 (2003).
- [15] O. Sima, “True Coincidence Summing Corrections – Theory”, International Atomic Energy Agency Technical Visit on Coincidence summing and geometry correction in gamma spectrometry IAEA Laboratories, Seibersdorf, Austria (2010).