Applied Radiation and Isotopes 133 (2018) 22–30

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

Calculation of the detection limits for radionuclides identified in gamma-ray spectra based on post-processing peak analysis results

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HIGHLIGHTS

- Detection limits for radionuclides detected in gamma-ray spectra are calculated.
- As inputs for the calculations the results of peak-analyzing programs are used.
- The method is applicable to gamma-ray emitters exhibiting peaked background.
- Detection limits for overlapping peaks can be calculated as well.
- For multi-gamma-ray emitters detection limits from more peaks can be calculated.

ARTICLE INFO

Keywords:
Gamma-ray spectrometry
Peak analysis
Background
Detection limit
ROI method
Least-squares method
Uncertainty
Correlation coefficient

ABSTRACT

A new method for calculating the detection limits of gamma-ray spectrometry measurements is presented. The method is applicable for gamma-ray emitters, irrespective of the influences of the peaked background, the origin of the background and the overlap with other peaks. It offers the opportunity for multi-gamma-ray emitters to calculate the common detection limit, corresponding to more peaks. The detection limit is calculated by approximating the dependence of the uncertainty in the indication on its value with a second-order polynomial. In this approach the relation between the input quantities and the detection limit are described by an explicit expression and can be easy investigated. The detection limit is calculated from the data usually provided by the reports of peak-analyzing programs: the peak areas and their uncertainties. As a result, the need to use individual channel contents for calculating the detection limit is bypassed.

1. Introduction

The detection limit represents the smallest signal that can be detected with a predefined probability under specified measurement conditions (ISO, 2010). It is used to check whether the specific measurement conditions comply with the guideline values defining the quality of the measurement results. The detection limit can be calculated from the data obtained from a null measurement, i.e., a measurement where the measurand is not present or does not contribute to the result (ISO, 2007) or from measurements of test samples where the measurand is present (Gilmore, 2008). Therefore, it represents an assessed quantity value, since from the circumstances in an existing measurement we conclude about the circumstances in another measurement performed in identical counting conditions, albeit with a different value of the measurand. Since this measurement was actually not preformed the detection limit represents an a-priori quantity value. The detection limit is calculated from the equation

$$y^{\#} = y^{*} + k_{1-\beta} \cdot u(y^{\#}) \quad (1)$$

where y^{*} denotes the decision threshold (ISO, 2010), $k_{1-\beta}$ is the quantile of the standardized normal distribution corresponding to the probability $1-\beta$ and $u(y^{\#})$ is the uncertainty in the measured value if it equals $y^{\#}$. Eq. (1) has to be solved for the value of the measurand $y^{\#}$ (ISO, 2010). According to the measurement model this value is given in terms of the net indication $n_n{}^{\#}$ (ISO, 2007), corresponding to the detection limit and the conversion factor w as $y^{\#} = w \cdot n_n{}^{\#}$ (ISO, 2010). The uncertainty $u(y^{\#})$ depends on the uncertainty in the conversion factor as well as on the uncertainty in the net indication, which is a function of the value of the net indication. Since the function $u(n_n)$ describing the uncertainty in the indication as a function of the value of the indication n_n depends on the kind of measurement for which the detection limit is calculated, the equation for the detection limit cannot be solved directly for the general case in a closed form, but only by iteration (ISO, 2010). However, because the measurement model is linear, it is possible to

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https://doi.org/10.1016/j.apradiso.2017.11.031
Received 8 June 2017; Received in revised form 20 November 2017; Accepted 28 November 2017
Available online 11 December 2017
0969-8043/

rewrite the equation for the detection limit in the equation for the net indication corresponding to the detection limit

$$n_n^{\#} = n_n^{*} + k_{1-\beta'} \cdot u(n_n^{\#}), \quad (2)$$

where n_n^* denotes the net indication corresponding to the decision threshold, $n_n^* = y^*/w$, and $k_{1-\beta'}$ denotes the quantile for the net indication, which depends on the uncertainty in the conversion factor $u(w)$ (Glavič-Cindro et al., 2017):

$$k_{1-\beta'}^2 = k_{1-\beta'}^2 \cdot [1 + u_{rel}^2(w) \cdot n_n^{\#2}/u^2(n_n^{\#})]. \quad (3)$$

Then, the equation for the net indication corresponding to the detection limit is as follows:

$$(n_n^{\#} - n_n^{*})^2 = k_{1-\beta'}^2 \cdot [u^2(n_n^{\#}) + u_{rel}^2(w) \cdot n_n^{\#2}]. \quad (4)$$

To calculate the indication corresponding to the detection limit explicitly, $u(n_n)$ must be known in terms of n_n and Eq. (4) must be solved for $n_n^{\#}$ in an appropriate approximation. Then, the detection limit is calculated from the net indication $n_n^{\#}$ using the measurement model.

Since the net indication is given as the difference between the gross indication and the background indication, in counting experiments it is usually supposed that, according to the Poisson distribution of counts, the uncertainty in the net indication is

$$u^2(n_n) = u^2(n_g) + u^2(n_0) = n_g + n_0 = 2 \cdot n_0 + n_n, \quad (5)$$

where n_g and n_0 denote the gross and background indications, respectively. For this case the detection limit $y^{\#}$ can be derived in a closed form (Kirkpatrick et al., 2013). In gamma-ray spectrometry, where the gross number of counts and the number of counts in the continuous background can be obtained from overlapping spectral regions, because of the correlations Eq. (5) does not apply (Korun et al., 2016a). In gamma-ray spectrometry the net indication is represented by the difference between the indication, i.e. the number of counts in the peak (the peak area) and the blank or background indication (the area of the background peak) (ISO, 2007). It is the purpose of this contribution to present the calculation of the detection limits for gamma-ray spectrometric measurements using the peak areas and its a-priori uncertainty calculated by the peak analysis procedure, assuming a case of overlapping spectral regions.

2. Methods

In gamma-ray spectrometric measurements of activities the indications, i.e., the peaks, are associated with gamma-ray emitters on the basis of the agreement between the energies of the peaks occurring in the spectrum and the energies from the nuclide library, comprising the radionuclides and the gamma-ray energies where they radiate. If an agreement is found, the corresponding radionuclide is believed to be present in the sample. In computerized gamma-ray spectra analyses, the peaks are automatically located, evaluated and associated with radionuclides tabulated in the nuclide library. If several peaks are associated with a radionuclide, its activity can be calculated from the data corresponding to all the peaks associated with it.

For the calculation of the decision thresholds and the detection limits for radionuclides believed to be present in the sample the data from the peak-analysis report can be used. To perform the calculation for a specific peak, from the peak-analysis results the peak area n_p and its uncertainty $u(n_p)$ are retrieved for post-processing. The total number of counts in the peak region, n_g, is retrieved directly from the spectrum, assuming a width of the peak region, given in terms of a predefined number of FWHMs at the energy where the peak appears. Usually, values of 1.25 or 2.5 are used for unexpressive and expressive peaks, respectively (ISO, 2010). The uncertainty of the total number of counts is $n_g^{1/2}$.

2.1. Isolated peaks

Supposing that all the counts belonging to the indication contribute to n_g, the number of counts in the continuous background within the peak region is $n_0 = n_g - n_p$. Because the total number of counts in the peak region is fixed (it is given by the spectral data as the sum of the channel contents within the peak region) the number of counts in the peak and in the continuum are correlated with a correlation coefficient of –1. This value of the correlation coefficients follows from the evaluation of the peak area with the method of least squares (ISO, 2010; Korun et al., 2016a), where the same region of the spectrum is used to determine the number of counts in the peak and the number of counts in the continuous region where it resides. It follows that with isolated peaks the uncertainty in the number of counts in the continuum is $u(n_0) = u(n_p) - u(n_g)$ (Korun et al., 2016a). It should be mentioned that, because the uncertainty of the background is expressed as a difference, it might assume a value smaller than zero. This may happen if an excessively wide spectral interval is used for calculation of n_g or because of shortcomings in the peak area uncertainty calculation. In this case the negative value of the difference should be used.

The number of counts in a peak is the sum of the counts belonging to the net indication and the counts belonging to the peaked background n_B

$$n_p = n_n + n_B. \quad (6)$$

The number of counts n_B in the peaked background and its uncertainty $u(n_B)$ are obtained from separate measurements or from spectral regions that do not overlap with the peak region. Therefore, they are not correlated statistically. It should be mentioned that not only the spectrometer background, but also the presence of interfering radionuclides in the sample and the activity of the blank sample, can contribute to n_B and $u(n_B)$. It should also be recognized that n_B and $u(n_B)$ are not directly related. For instance, in the case of a spectrometer background that is stable with time $u(n_B)$ can be arbitrarily small if the duration of the background measurements increases appropriately. On the other hand, if the background depends on time and the dependence is not related to any influencing quantity that is being measured, $u(n_B)$ is given by the dispersion of the background-measurement results.

The total number of counts in the peak region for the net indication $n_n^{\#}$ is $n_g^{\#} = n_{g0} + n_n^{\#}$, where n_{g0} denotes the total number of counts in the peak region in the null measurement, i.e., the gross background. Since $u(n_p) = u(n_0) + u(n_g)$ the uncertainty in the number of counts in the peak, corresponding to the net indication $n_n^{\#}$, is

$$u[n_p(n_n = n_n^{\#})] = u(n_0) + n_g^{\#1/2} = u(n_p) - n_g^{1/2} + (n_{g0} + n_n^{\#})^{1/2}. \quad (7)$$

The various quantities representing the number of counts in the region of interest (n_p, n_n, n_B, n_g, n_{g0} and n_0) are presented in Fig. 1. Since

$$n_{g0} = n_0 + n_B = n_g - (n_p - n_B), \quad (8)$$

the uncertainty in the number of counts in the peak comprising $n_n^{\#}$ counts that belong to the indication can be expressed in terms of the peak-analysis data, the peaked background data and the gross number of counts in the peak region as

$$u[n_p(n_n = n_n^{\#})] = u(n_p) - n_g^{1/2} + [n_g - (n_p - n_B) + n_n^{\#}]^{1/2}. \quad (9)$$

This equation replaces Eq. (5) in the derivation of the detection limit, which describes the uncertainty in the net indication in the counting experiments. It is evident that for $n_B = 0$ and $n_g = n_p$ (no background) the uncertainty in the peak area equals the uncertainty in the total number of counts and, consequently, the uncertainty in the peak area corresponding to the detection limit is $(n_n^{\#})^{1/2}$. In the case of a large and well-determined continuous background, i.e., when $n_p \ll n_g$ and $u(n_0) = 0$, it follows that $u(n_p) = n_g^{1/2}$ and the uncertainty in the peak area corresponding to the detection limit is $(n_g + n_n^{\#})^{1/2}$. It follows that the null-measurement uncertainty in the indication is