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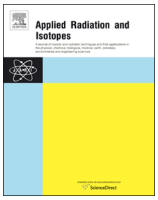

Calculation of the decision thresholds in gamma-ray spectrometry

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HIGHLIGHTS

- Decision thresholds were calculated for gamma-ray emitters by inducing type-I errors.
- Peaks were supposed to appear at energies where the emitters radiate.
- In case of a peaked background the peak area was obtained from the background peak area uncertainty.
- Decision thresholds were calculated from the uncertainties of the average activities.
- The peaked background increases the decision threshold considerably.

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ABSTRACT

A method was developed for calculating the decision thresholds for gamma-ray spectrometric measurements. At the energies where gamma-ray emitters that are present in the nuclide library, but were not identified in the spectrum, radiate, peaks are supposed to appear. The peak areas are calculated by fitting, using the method of least squares, the spectral region of the supposed peaks with a continuous background and the spectrometer response function at the gamma-ray energies where the supposed peaks are positioned. The null measurement uncertainty of a gamma-ray emitter is obtained as the uncertainty of the weighted average of the activities calculated from the areas of the supposed peaks in a spectrum where the specified activity of the gamma-ray emitter is zero. For the calculation of the decision threshold the null measurement uncertainty is used. These decision thresholds overestimate the critical limits calculated with the Currie formula by about 10% in the case of single gamma-ray emitters. For multi-gamma-ray emitters the decision thresholds yield smaller values than the Currie formula. The presence of a peaked background or peaks that are near the supposed peaks increases the decision threshold considerably.

1. Introduction

In gamma-ray spectrometry the presence of gamma-ray emitters in samples is recognized by identifying their characteristic peaks in the spectrum. Then, the activity of a gamma-ray emitter is calculated from the areas of the peaks associated with it. Frequently, for gamma-ray emitters of interest, but not identified in the spectrum, the decision threshold is reported as the upper limit of the interval containing the true activity (ISO, 2010, p. 15).

The ISO 11929 (ISO, 2010, p.10) standard defines the decision threshold in terms of the uncertainty of a measurement result obtained from a measurement on a blank sample or a sample with an activity having a reference quantity value, which is much smaller than the measurement uncertainty, referred to as the null measurement uncertainty (ISO, 2007, p. 45). Here, besides the statistical uncertainties of the channel contents, the uncertainties of the corrections of the peak areas due to the peaked background, the presence of interfering radionuclides in the sample and the activity of the blank also have to be taken into account. In addition to that, in the case of overlapping peaks, the uncertainties of the peak areas are not only given by the statistical uncertainties of the channel contents but also by the conditioning of the system of equations used for calculating them.

On the other hand, in calculations of the critical limit (Gilmore, 2008, pp. 114–116, Canberra, 1998, pp. 53–55), only statistical uncertainties of the channel contents lying within the peak region are taken into account. In reports on measurement results the critical limit is often used instead of the decision threshold as the upper limit of the activities for radionuclides that were not identified in the sample. To prevent any possible underestimation of the decision threshold reported, a method has been developed for calculating the decision threshold using a similar measurement model to that used in calculations of the activities of gamma-ray emitters that are identified in the sample. In this way all the

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sources of uncertainty that enter into the calculation of the activities of the radionuclides identified in the spectrum are taken into account also here.

Since in gamma-ray spectrometry the activities are usually calculated by computerized peak-evaluation and activity-calculation procedures, the easiest way to evaluate the uncertainty of the activity of gamma-ray emitters that were not identified in the spectrum is to induce type-I errors. Then, for these unrecognized radionuclides the activities, together with their uncertainties, are also calculated. Here, the calculated activities lie near zero and from their uncertainty it is possible to calculate the decision threshold.

Type I errors that occur spontaneously in gamma-ray spectra analyses performed with a low value of the sensitivity parameter, defining the criterion for differentiating between statistical fluctuations of the continuous background and small peaks, result in positive observed values having a large relative uncertainty. Usually, these results are converted to single-sided intervals, if their relative uncertainty exceeds the predetermined level defining the determination limit (Currie, 1968). These results are interpreted as “observed, but not quantified”. To reduce the frequency of type I errors in these results a narrow energy window for associating peak energies with energies appearing in the nuclide library is used and for multi-gamma-ray emitters more peaks are used for calculation of the activity (Canberra, 1998, pp. 39–42).

The described method is applied only to gamma-ray emitters not recognized in the spectrum. For single gamma-ray emitters, that are not present in the sample blank, it complies with the method described in the standard ISO 11919, since it takes into account the sources of uncertainty contributing to the indication and blank indication (peak count rate, background count rate and interfering count rates). For multi-gamma-ray emitters the decision threshold is calculated as the uncertainty of the weighted mean of the activities corresponding to different gamma-rays. For gamma-ray emitters present in the blank sample, the activity of the blank sample is subtracted from the total sample activity supposing homogeneous distribution of the gamma-ray emitters in the blank sample. Such subtraction simplifies the measurement model considerably, since it avoids calculation of count rates due to the blank sample and taking into account the correlations in calculation of the mean for multi-gamma-ray emitters.

The advantage of the proposed method over the Currie's method lies in the inclusion of the sources of uncertainty contributing to the blank indication including the peak area uncertainties that increase when overlapping peaks are decomposed. The advantage over the method described in the standard ISO 11929 is in the possible diminishing of the decision threshold for multi gamma-ray emitters because here the uncertainty of the mean activity is usually smaller than any of the uncertainties of the activities associated with specific gamma-rays. In this sense the method proposed is regarded as an extension of the method described in the standard ISO 11929. In addition to that, it eliminates the need to define the width of the peak region, used for calculating the decision threshold.

It is the purpose of this paper to describe the method of calculation of the null measurement uncertainty by inducing type-I uncertainties and to compare the decision thresholds calculated using the null measurement uncertainty with the minimum detectable activities calculated using the Currie formula (Currie, 1968).

2. Methods

Since the useful information retrieved from the spectra includes the peak properties, i.e. their positions, areas and widths, the peak location is normally the first operation that is performed in an automated gamma-ray spectrum-analysis procedure. Identifying the peak location usually involves searching for the maximums on the continuous spectral background. Here, a predetermined criterion is used to differentiate the statistical fluctuations of the continuous background from the small peaks. Maximums that fulfill this criterion are recognized as peaks. In spectral analyses that are performed in the framework of environmental monitoring programs of radioactivity the criterion is set low in order to prevent as many type-II errors as possible, i.e., to miss as few as possible signals of the presence of radioactivity in the sample. It should be mentioned that detecting small peaks, i.e., peaks with a large relative uncertainty of the peak area, such as those in excess of 50%, is subject to random influences because of the statistical fluctuations of the background within the peak region and its immediate proximity (Korun et al., 2012, 2013). It is therefore uncertain whether the small peaks are recognized as being present in the spectrum or not.

An alternative approach to spectrum analyses is to avoid the peak location and to analyze the spectrum shape in the energy regions where the gamma-rays emitted by the radionuclides of interest, which are the nuclides present in the nuclide library, may potentially appear. In these regions the spectrum shape is decomposed into the continuous background and the peaks having positions corresponding to gamma-ray energies from the nuclide library and peak widths given by the full width at half-maximum (FWHM) calibration of the spectrometer. From the fitting of the spectrum shape with a linear combination of the smooth continuous background and the peak shapes the areas of the peaks are calculated. It should be noted that in this approach the peak areas can be either positive or negative, depending on the shape of the background in the peak region. It should also be noted that because of the peaked background and the interferences among the gamma-ray emitters this approach only gives reliable results if the background data file comprises all the peaks that appear in the spectrometer background and the radionuclide library is extensive enough so that all the peaks located in the spectrum can be associated with their source.

This approach was adapted for the calculation of the decision thresholds for radionuclides that are present in the nuclide library, but were not identified in the spectrum. For each of the energies where these radionuclides radiate, areas of the supposed peaks are calculated and the uncertainties of the areas are used as the basis for the calculation of their decision thresholds.

In the case of the peaked background, i.e., in the case when the energy of the supposed peak is so close to the located peak that a reliable calculation of the contribution of the supposed peak is not possible, the information from the peaked background and the interfering nuclei is used for the calculation of the decision threshold. So, except in the case of the peaked background, the information for the calculation of the decision thresholds does not refer to the located peaks, but only to the continuous background. However, in the case of the peaks that escaped location or peaks with an erroneous peak area, the continuous background exhibits maxima or minima that are more pronounced than the statistical fluctuations and consequently increase the uncertainty of the background used in the calculation of the decision threshold.

Since the calculation of the decision thresholds is performed at the end of the spectrum analysis, the positions, the areas and the widths of the recognized peaks are known. For each gamma-ray energy in the nuclide library belonging to an unidentified radionucleus, the energy interval symmetric with respect to the energy of the supposed peak E_s and having a width of 6 FWHM is checked for the presence of localized peaks. If the energy E_s coincides with the energy of a located peak better than FWHM/2, no spectrum fitting is performed, because at a spacing of less than FWHM/2 the decomposition of a peak in two components does not yield reliable results. A minimal spacing of FWHM/2 for decomposition used from experience, although usually 1 FWHM is quoted for the