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Nuclear energy — Waste-packages activity measurement —

Part 1:
High-resolution gamma spectrometry in integral mode with open geometry

Énergie nucléaire — Mesurage de l'activité de colis de déchets —

Partie 1: Spectrométrie gamma haute résolution en mode intégral et géométrie ouverte

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Foreword

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ISO 14850-1 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Subcommittee SC 5, *Nuclear fuel technology*.

ISO 14850 consists of the following parts, under the general title *Nuclear energy — Waste-packages activity measurement*:

- *Part 1: High-resolution gamma spectrometry in integral mode with open geometry*
- *Part 2: Gamma-ray spectrometry using HPGe detectors*

Introduction

Several non destructive methods may be used after calibration to determine the radioactive characteristics of a waste package:

- gamma spectrometry;
- passive neutron counting, with or without discrimination of neutrons originating from (α,n) reactions;
- active neutron counting, with detection of neutrons resulting from induced fission reactions (prompt or delayed neutrons).

This part of ISO 14850 describes one procedure for measuring the activity contained in waste packages by gamma spectrometry and points out recommendations for the calibration of a measurement chain.

Nuclear energy — Waste-packages activity measurement —

Part 1:
High-resolution gamma spectrometry in integral mode with open geometry

1 Scope

This part of ISO 14850 describes a procedure for measurements of gamma-emitting radionuclide activity in the following homogeneous objects:

- unconditioned waste, including process waste (filters, control rods, etc.), dismantling waste, etc.;
- waste conditioned in various matrices (bitumen, hydraulic binder, thermosetting resins, etc.), notably in the form of 100 l, 200 l, 400 l or 800 l drums, and test specimens or samples, (vitrified waste);
- waste packaged in a container, notably technological waste.

It also specifies the calibration of the gamma spectrometry chain.

The gamma energies used generally range from 0,05 MeV to 3 MeV.

2 Terms, definitions and symbols

For the purposes of this document, the following terms and definitions apply.

2.1
reference source
radionuclide sealed in a suitable containment, of which the radioactive characteristics are determined by comparison with a reference material

2.2
source volume
volume in m^3 taken up by the matrix (or by the waste) in which the activity is distributed

2.3
source weight
weight in kg of the matrix in which the activity is distributed

2.4
package
object to be characterized, comprising an outer shell (container or canister) surrounding the (conditioned or unconditioned) source volume

2.5
mockup
package consisting of a container and of well-known materials representative of a matrix

2.6
reference package
mockup containing reference sources in a well-known configuration

2.7
apparent density of the source
ratio of the mass of the source to its volume

2.8
container
envelope of source volume

2.9
matrix
structural material immobilizing the radioactivity

2.10
radioprotection-shield
material of suitable nature and thickness placed around a package to attenuate the photon emission flux

2.11
gamma ray attenuator
material of suitable nature and thickness placed between the package and detector to attenuate the photon flux

2.12
detector
any type of high-purity germanium semiconductor

2.13
efficiency
ε
ratio between the number of detected events and the number of emitted gamma photons

2.14
collimator and background shield
protective devices for the detector to decrease background by limitation of the solid angle and gamma background (collimator) and reduction of the ambient background incident (background shielding)

2.15
background noise
B_e
number of counts in s^{-1} recorded at energy e in the background spectrum in the absence of any source, sample or package

2.16
decision threshold
T_D
value (in s^{-1}) above which an observed quantity is considered true, within the risk α

NOTE This limit corresponds the risk α of affirming the presence of the true quantity when it is in fact not present. The recommended value of α is 2,5 %

2.17
detection limit
L_D
value (in Bq) of the measured quantity that can be detected with a given probability $(1 - \beta)$, i.e. capable of providing a corrected result exceeding the decision threshold

NOTE The risk β corresponds to the risk of affirming the absence of the true quantity when it is in fact present. The recommended β value is 2,5 %

2.18
combined standard uncertainty

$u_{\mathrm{c}x}$

sum-of-the-squares combination of standard uncertainties arising from a Type A evaluation (applying statistical methods, expressed as a standard deviation s_i) and a Type B evaluation (non-statistical methods, expressed as a standard deviation u_j):

$$u_{\mathrm{c}x} = \left[\sum_i (s_i)^2 + \sum_j (u_j)^2 \right]^{1/2}$$

2.19
coverage factor

k

user-defined value, depending on the probability law, the level of confidence, and the precision of the estimated standard deviation, with

$k = 1$ for standard deviation calculations, and

$k = 2$ for the normal law, for a 95 % confidence level assuming a known standard deviation

2.20
expanded uncertainty

product of the standard deviation by the coverage factor, k

3 Principle

This part of ISO 14850 describes a non-destructive method implemented using fixed or mobile equipment, which is based on measuring the photon emission rate at different energies by gamma spectrometry:

- to identify detectable radionuclides in the objects in Clause 1, either by direct measurement or by using decay schemes;
- to determine the activity of the identified radionuclides from the counting rates recorded under each total absorption peak, weighted by a calibration factor.

The method implies optimization of the equipment parameters, energy calibration and efficiency calibration (by modelling the transfer function or by fabricating mockups):

- choice of detector(s), electronic circuitry and shielding;
- choice of measurement geometry;
- choice of calibration geometry.

The method may be validated:

- by comparison with destructive examination results on representative samples;
- by measurement of reference packages for which the activity, the nature of the radionuclides, the nature of the elements composing the waste and homogeneity are accurately known.

4 Detectors and ancillary equipment

The measuring station usually comprises the following equipment.

a) Mechanical equipment:

- a package-positioning system (rotation, with optional vertical movement);
- a detector-positioning system (vertical, horizontal and distance);
- a weighing station (optional);
- a turntable;
- collimator, background shielding, gamma ray attenuators,

b) Detector and signal-processing electronics:

- a detector and preamplifier;
- an amplifier;
- an analog-digital converter,
- a “stand alone” module or a computer interface card,

c) Computer with measurement processing and interpretation software.

4.1 Mechanical equipment

4.1.1 Package-positioning system

The package-positioning system is designed to support the package to be measured, and to move it in rotation about its vertical axis. It may also allow for vertical moving of the package, and may be equipped with a weighing system.

4.1.2 Detector-positioning system

This system carries the detector with its protective shielding, and positions it in elevation and azimuth with respect to the package to be measured. It may be equipped with a system to determine the relative position of the package with respect to the detector.

4.2 Detector and preamplifier

The method covers only high-purity germanium semiconductor detectors. Two types of detectors may be selected depending on the energy of the radionuclides to be measured:

- planar or flat coaxial detectors provide better resolution at low energy (below 400 keV),
- coaxial detectors give higher efficiency at high energies.

The semiconductor crystal requires a cryogenic system. The detector signal is collected by a charge sensitive preamplifier; this can be of either the resistive feedback type, transistor reset type or pulsed optical feedback type depending upon the application.

4.3 Amplifier

The amplifier implements Gaussian or triangular pulse shaping with a time constant adjustable from 0,25 µs to 15 µs. A pileup rejector is generally used, and in some cases the amplifier is equipped with a gated integrator.

Various types of amplifiers may be used in conjunction with this method. The choice of an amplifier depends on the other components in the counting system.

4.4 Analog-digital converter (ADC)

Two types of ADC converters are used in gamma spectrometry:

- Wilkinson ADCs, with a variable dead time; counting losses depend on the conversion frequency and the signal amplitude;
- successive approximation ADCs, with a fixed dead time independent of the signal amplitude.

4.5 Multi-channel analyzer (MCA) and data processing system

The analyzer stores the encoded data in a basic memory array available to the computer.

NOTE A digital signal processing module may replace the functions described in 4.2, 4.3 and 4.4. It quantizes the preamplifier output signal, allowing higher counting rates

4.6 Background shielding

The unit is shielded against external radiation by (preferably low-activity) lead or other high-density materials. The dimensions depend on the characteristics of the measurement environment.

4.7 Collimator

The collimator is made of a high-density material (Pb, W, Ta, Cu) with a geometry corresponding to the desired detection solid-angle acceptance. The collimator geometry is designed to maximize the ratio of the gamma activity signal from the material of interest, to the gamma activity signal arising from the surrounding area, i.e the ambient background.

4.8 Gamma ray attenuators

Gamma ray attenuators can be placed in front of the detector to attenuate the incident photon flux. The material and thickness are selected according to the flux characteristics.

5 Calibration

Calibration consists of determining the efficiency (or yield) versus energy curve(s) of each detector (or of the complete measuring unit). The curve(s) is used to evaluate the ratio between the number of detected events and the number of gamma photons emitted from several single-energy sources or from a few multiple-energy sources with well-spaced energy lines covering the gamma ray region of the radionuclides present in the measured samples or in the measured packages.

The energy/channel relation shall first be established by means of several single-energy (or multiple energy) sources covering the energy band relevant to the measured samples or packages.

5.1 Principle of the calibration

5.1.1 The activity (in becquerels) of radionuclide i measured at an energy e at a reference date is defined by the following relation:

$$A_{i,\mathrm{e}} = \left(N_{i,\mathrm{e}} - B_\mathrm{e}\right) \cdot \frac{1}{\rho_{i,\mathrm{e}}} \cdot \frac{1}{\varepsilon_\mathrm{e}} \cdot K_i \cdot f_{i,\mathrm{e}}\left(k_1, k_2 ...\right) \quad (1)$$

where

$A_{i,\mathrm{e}}$	is the activity, in becquerels, of radionuclide i at energy e;
$N_{i,\mathrm{e}}$	is the number of counts expressed in s^{-1} in the total absorption peak of radionuclide i at energy e;
B_e	is the number of counts expressed in s^{-1} recorded at energy e in the background spectrum;
$\rho_{i,\mathrm{e}}$	is the probability of photon emission by radionuclide i at energy e;
ε_e	is the detection efficiency at energy e;
K_i	is the correction factor for the radioactive decay during the measurement for radionuclide i (generally equal to 1);
$f_{i,\mathrm{e}}(k_1, k_2, \ldots)$	is the correction factor for variations in self-attenuation, attenuation and solid angle for radionuclide i at energy e.

Radionuclide i is the radionuclide to be determined in the measured sample or package. $f_{i,\mathrm{e}}(k_1, k_2,\ldots)$ represents calibration parameters related to the differences in geometry and matrix between the measurement standard and the measured object. The self-attenuation, attenuation and solid angle variations taken into account in this factor depend on the specific absorption coefficients of the elements found in the measured object (and the screens, if any), as well as on the object density and geometric dimensions.

The calibration of the measurement device consists of determining the product

$$\frac{1}{\varepsilon_\mathrm{e}} \cdot f_{i,\mathrm{e}}\left(k_1, k_2 ...\right)$$

5.1.2 This can be determined by one of following methods.

a) The calibration of the detectors (associated to their collimators) using reference sources; which allows the determination of the efficiency (ε_e). In this case, the corrective factor $f_{i,\mathrm{e}}(k_1, k_2,...)$ is the result of a calculation (simulation) taking into account the different parameters.

b) Measurements on reference packages representative of the package to be characterized (in terms of geometry, activity and matrix characteristics).

The radioactive sources are reference materials of which the activity value and the associated uncertainty are sufficiently well defined to allow the evaluation of a measurement method. It is important to keep in mind that

- the method of the “reference package” is adapted to the measurement devices treating a large number of packages (of limited type), respecting accurate specifications for which a mockup is easy to achieve (for example, set geometry, only slightly varied matrices, limited activity range), and
- the method of the “reference sources” is adapted to the measurement devices with a varied use range not allowing a simple mockup (for example, varied geometries and matrices, extensive activity range). This method can however require the implementation of complex calculation codes.

5.1.3 To be representative, the parameters to take into account for the calibration, normally defined in the specifications, are listed below:

a) characteristics of the “container” (drum, shell, etc.):
   - dimensions,
   - nature and composition of materials,
   - shielding thickness,
   - presence of biological protection,
   - mass when empty;

b) characteristics of the waste (or of the matrix):
   - apparent density (mass and volume of the source),
   - composition in mass fraction of the constitutive materials,
   - spatial distribution of materials,
   - nature, activity and distribution of radionuclides;

c) measurement geometry:
   - relative position source-detector,
   - collimation (nature of materials, dimensions),
   - possible presence of gamma ray attenuator (nature of materials, dimensions).

5.2 Calibration apparatus

The mockups or simulations cannot take into account all the different combinations of the parameters mentioned in 5.1.3.

To be sure that the calibration is representative of all the cases considered, we define an “upper limit” corresponding to the extreme variations accepted by the characteristics of the measurement station.

5.2.1 Characteristics of the mockup container

For the calibration of the assay system, using the reference package, the same “container” as the one for the packages to be assayed (including the additional biological protections, in some cases) is used.

For the calibration of the detectors by “reference source” and calculated simulation, the real characteristics of the “container” and those of the biological protections are described.

5.2.2 Characteristics of the mockup matrix

The “source volume” shall be representative, especially for the filling height.

The “source mass” shall be known through weighing.

The “apparent density” is a variable parameter, a function of the mass and volume of the source. The representation or the description of this variation requires taking into account the mockup matrices of different densities (see Table 1).

The nature, activity and distribution of radioelements: for the range 50 keV to 2 MeV the range of sources in Table 2 can be used. The reference sources used for the calibration shall have the gamma energies which surround those of the radionuclides looked for.

The volume distribution of the materials: the “reference packages” or the calculation take into account the variations of the measurement station characteristics.

The efficiency curve of a package of given apparent density is obtained by interpolation, using the reference matrix yield curves. The number of reference mockups and the chosen interpolation function should be checked to be sufficient so as to not generate errors incompatible with the performances of the device.

The number of necessary yield curves depends on

- the range of the materials to be measured,
- the required accuracy, and
- the interpolation possibilities of the treatment program.

For the measurement by gamma spectrometry of the wastes from the nuclear industry, ^{133}Ba, ^{152}Eu and ^{241}Am (sometimes complemented by ^{239}Pu, ^{137}Cs and ^{60}Co) generally cover the energy range involved.

For the calibration of the measurement device by reference packages, if verifications or inter-comparisons are periodically made, it is necessary that the package keeps all its initial physical characteristics over time.

The calibration of the detector using the reference sources shall be performed within the validity range of the measurement chain.

Table 1 — Examples of matrices proposed for the calibration

Nature of the “source volume”	Possible material of the mockup	Apparent densities $kg \cdot m^{-3}$
Plastic wastes	Polyvinyl chloride	100 to 300
Cellulose wastes	Wood	150 to 400
Steel parts (metal tubes)	Stainless steel	~ 700
Dismantling concrete waste	Concrete	~ 1 000
Concrete	Concrete	~ 2 300

Table 2 — Main gamma ray characteristics of radionuclides used for the elaboration of the yield curves

Radionuclide	Half-life	Emission energy keV	Probability of photonic emission %
^{60}Co	5,271 ± 0,001 years	1 173,24 ± 0,004	99,89 ± 0,02
		1 332,50 ± 0,005	99,98 ± 0,001
^{88}Y	106,62 ± 0,02 days	898,04 ± 0,004	94,10 ± 0,5
		1 836,06 ± 0,013	99,36 ± 0,05
^{137}Cs	30,15 ± 0,02 years	661,66 ± 0,003	85,20 ± 0,2
^{133}Ba	10,5 ± 0,1 years	53,16 ± 0,001	2,20 ± 0,04
		79,62 ± 0,01	2,63 ± 0,08
		81,00 ± 0,005	34,10 ± 0,5
		276,40 ± 0,002	7,17 ± 0,04
		302,85 ± 0,001	18,32 ± 0,07
		356,02 ± 0,002	62,00 ± 0,3
		383,85 ± 0,003	8,93 ± 0,06
^{152}Eu	13,53 ± 0,03 years	121,78	28,40 ± 0,15
		244,70 ± 0,001	7,54 ± 0,05
		344,28 ± 0,002	26,52 ± 0,18
		411,12 ± 0,005	2,25 ± 0,016
		443,98 ± 0,007	3,10 ± 0,02
		778,90 ± 0,006	12,94 ± 0,07
		867,39 ± 0,008	4,23 ± 0,03
		964,13 ± 0,009	14,60 ± 0,08
		1 085,91 ± 0,013	10,09 ± 0,04
		1 089,70 ± 0,015	1,74 ± 0,008
		1 112,12 ± 0,017	13,56 ± 0,06
		1 212,95 ± 0,012	1,42 ± 0,01
		1299,12	1,63 ± 0,01
		1 408,01 ± 0,015	20,80 ± 0,12
^{235}U	7,04E+08 ± 1E+06 years	143,76 ± 0,02	10,96 ± 0,08
		163,33 ± 0,02	5,08 ± 0,04
		185,72 ± 0,005	57,20 ± 0,5
		205,31 ± 0,01	5,01 ± 0,05
^{238}Pu	87,74 ± 0,1 years	99,85 ± 0,001	7,4E-03 ± 8E-05
		152,72 ± 0,002	9,4E-04 ± 1E-05

Table 2 (*continued*)

Radionuclide	Half-life	Emission energy keV	Probability of photonic emission %
^{239}Pu	2,41E+04 ± 30 years	51,62 ± 0,001	2,7E-02 ± 4E-04
		94,66	4,8E-03 ± 2E-04
		98,44	7,8E-03 ± 2E-04
		129,30 ± 0,001	6,2E-03 ± 6E-05
		203,55 ± 0,005	5,7E-04 ± 4E-06
		332,85 ± 0,005	5,0E-04 ± 1E-05
		345,01 ± 0,004	5,6E-04 ± 8E-06
		375,05 ± 0,003	1,6E-03 ± 1E-05
		413,71 ± 0,005	1,5E-03 ± 3E-05
^{240}Pu	6560 ± 10 years	160,31 ± 0,003	4,0E-04 ± 4E-06
^{241}Pu	14,4 ± 0,1 years	148,57 ± 0,01	1,9E-04 ± 3E-06
		164,61 ± 0,02	4,5E-05 ± 2E-06
		208,01 ± 0,03	5,3E-04 ± 2E-05
^{241}Am	432,7 ± 0,6 years	59,54 ± 0,001	35,90 ± 0,4

5.2.3 Measurement geometry

Relative position of the source and of the detector.

a) For the case of a detector calibration using point “reference sources”, the source-detector distance, d, shall be assessed and reproduced accurately. This distance shall be located in the zone where the received gamma flux varies according to a law in

$$\frac{1}{d^2}$$

b) For the case of the calibration of the measurement device by “reference package”, it is necessary to be able to position the sources radially and axially inside the package. The positioning device used must have a negligible absorption of the gamma radiation versus the absorption of the mockup or the geometric effect.

The aim is to distribute the sources to achieve a distribution representative of the packages to be measured.

For the drums, the positioning is done using aluminium tubes (thin series) or PVC tubes, attached to the package.

Two models exist for the radial positioning:

- a model of parametering versus the radius R (in the centre, at $R/2$, at $R/\sqrt{2}$ and in the periphery). This model allows us to take into account (in some cases) the radioactive heterogeneities;
- a positioning model in spiral form with 8 positions of linear sources.

The axial positioning is done inside each tube (by visible stick and extension, for example). The accuracy and the representativity of the positioning (by 5 cm steps for example) are important.

5.2.4 Collimation geometry

The calibration shall be done, in all cases, with the collimation geometry set for the normal operation of the measurement station.

5.2.5 Gamma ray attenuators

In the case of the measurement station calibration, the determination of the efficiency curves shall be made in the configuration(s) of the device with or without a gamma ray attenuators.

In the case of the detector calibration, the attenuation by gamma ray attenuators is taken into account in the simulation calculation.

5.3 Measurements to be performed

5.3.1 Setting of the gamma spectrometry chain

Before acquiring a spectrum for the efficiency calibration, the settings of the spectrometry chain must be similar to those used during the measurements, for example:

a) for a spectroscopy amplifier:

  - time constant,
  - gain,
  - correction devices;

b) for an Analogue to Digital Converter (ADC):

  - conversion slope,
  - offset,
  - upper and lower level discrimination,
  - setting of zero.

5.3.2 Control of resolution

With the counting electronic chain set correctly it shall be ensured that the resolution of the detector is within the manufacturer’s specification. This is usually defined in terms of the width of a defined peak at half of its height, i.e full width half maximum (FWHM). It shall be periodically checked that these values remain within the manufacturer’s specification. For the planar and coaxial detectors, the peak at 122,0 keV (^{57}Co) or 121,8 keV (^{152}Eu) and the peak at 1 332,5 keV (^{60}Co) can be used respectively for checking.

5.3.3 Control of the calibration in energy

The program shall recognize peaks of the radionuclide of the reference source with a research window of a width of ± 1 keV (it can be less for certain applications).

5.3.4 Counting times during calibration

The duration of counting is defined so that the statistic of the photoelectric peak selected for the calibration is correct (for example, number of events $>$ 10 000).

For each reference radionuclide, and for each detector associated to the geometry, a source spectrum is recorded, as well as a background noise spectrum, of time more or less equal to the measurement spectrum, with a frequency which depends on the installation (at least before each calibration campaign).

5.4 Error estimation of the calibration parameters

According to Equation (1), the calibration coefficient corresponds to the following formula:

$$\varepsilon_i = \left(N_{i,\mathrm{e}} - B_\mathrm{e}\right)\cdot\frac{1}{\rho_{i,\mathrm{e}}}\cdot\frac{1}{A_{i,\mathrm{e}}}\cdot K_i\cdot f_{i,\mathrm{e}}\left(k_1,k_2\ldots\right) \tag{2}$$

This coefficient can be considered as the product of five variables:

$$N_{i,\mathrm{e}} - B_\mathrm{e};\quad 1/\rho_{i,\mathrm{e}};\quad 1/A_{i,\mathrm{e}};\quad K_i \quad\text{and}\quad f_{i,\mathrm{e}}\left(k_1,k_2,\ldots\right).$$

By considering these variables as independent, the composed typical uncertainty affecting this calibration coefficient is estimated using the quadratic sum of the typical uncertainties affecting each of these variables: we thus have in the first order:

$$\frac{u_{\mathrm{c}\,\varepsilon i}}{\varepsilon_i} = \left[\frac{\left(\dfrac{N_{i,\mathrm{e}}}{t_1} + \dfrac{B_\mathrm{e}}{t_2}\right)}{\left(N_{i,\mathrm{e}} - B_\mathrm{e}\right)^2} + \frac{u^2_{\rho_i}}{\rho^2_{i,\mathrm{e}}} + \frac{u^2_{\mathrm{c}\,A_i}}{A^2_{i,\mathrm{e}}} + \frac{u^2_{K_i}}{K_i^2} + \frac{u^2_{\mathrm{c}\,f_{i,\mathrm{e}}(k_1,k_2,\ldots)}}{f^2_{i,\mathrm{e}}\left(k_1,k_2,\ldots\right)}\right]^{1/2} \tag{3}$$

where t_1 and t_2 are respectively the counting durations corresponding to $N_{i,\mathrm{e}}$ and B_e. The first four terms of Equation (3) can easily be estimated but this is more difficult for the last term. Indeed, the latter is a function of:

— the geometric characteristics of the whole detector, of the sample and of the possible collimation,

— the nature and the thickness of the screens placed between the detector and the sample, and

— the nature and characteristics of the matrix.

In the simplest case:

$$f_{i,\mathrm{e}} = C_\mathrm{geo}\cdot \mathrm{e}^{-\mu_{(\mathrm{ec,e})}\cdot x_{(\mathrm{ec})}}\cdot\frac{1-\mathrm{e}^{-\mu_{(i,\mathrm{e})}\cdot x_{(i)}}}{\mu_{(i,\mathrm{e})}\cdot x_{(i)}} \tag{4}$$

where

C_geo	represents the geometric correction factor;
$\mu_{(\mathrm{ec,e})}$ and $\mu_{(i,\mathrm{e})}$	are the attenuation coefficients recorded for the energy e of the shield and the element i of the matrix ,
$x_{(\mathrm{ec})}$ and $x_{(i)}$	are the thicknesses of the screen and the matrix element.

Therefore, the typical uncertainty affecting $f_{i,\mathrm{e}}$ can be referred:

- either to a simplified Equation (4):

$$\frac{u_{\mathrm{c}}\,f_{i,\mathrm{e}}}{f_{i,\mathrm{e}}} \approx \left[\frac{u^2_{C_{\mathrm{geo}}}}{C^2_{\mathrm{geo}}} + \mu^2_{(\mathrm{ec,e})}\cdot u^2_{x_{(\mathrm{ec})}} + x^2_{(\mathrm{ec})}\cdot u^2_{\mu_{(\mathrm{ec,e})}} + \frac{\mu^2_{(i,\mathrm{e})}\cdot u^2_{x_{(i)}} + x^2_{(i)}\cdot u^2_{\mu_{(i,\mathrm{e})}}}{4}\right]^{1/2} \tag{5}$$

- or to the expression acquired during the use of a Monte Carlo code or of another simulation process:

$$\frac{u_{\mathrm{c}}\,f_{i,\mathrm{e}}}{\overline{f}_{i,\mathrm{e}}} = \frac{1}{\overline{f}_{i,\mathrm{e}}}\left[\sum_{j=1}^{n}\frac{\left(\overline{f}_{i,\mathrm{e}} - f_{j,i,\mathrm{e}}\right)^2}{(n-1)}\right]^{1/2} \tag{6}$$

where

$f_{j,i,\mathrm{e}}$ represents the j^{th} value obtained during n experiments;

$\overline{f}_{i,\mathrm{e}}$ is the average of the n measurements of $f_{i,\mathrm{e}}$.

6 Operating procedure

Gamma spectrometry measurement requires

- prior calibration valid for the type of package measured, in order to relate the measured signal to the activity,
- determination or verification of the background level under the measurement conditions, and
- verification (by means of a reference package or a source, such as ^{133}Ba or ^{152}Eu) that the measurement system is free from drift.

The validity time of each of these parameters depends on the measurement system specifications and the conditions under which it is used.

The waste package to be characterized is inserted into the measuring unit and placed on a mounting plate that allows for rotation and positioning of the package with respect to the detector (refer to 5.1).

The duration of the measurement is determined according to the measurement station characteristics (operating constraints, detection limit, uncertainty, etc.). It may be desirable to subdivide the counting time to allow statistical processing of untrue values.

7 Interpretation of results

7.1 Activity

The activity (in becquerels) of radionuclide i measured at an energy e at a reference date is defined by Equation (1) given in 5.1.1.

7.2 Uncertainty evaluation

Equation (1) expresses the activity of radionuclide i determined from a peak at energy e on the assumption that the activity is the product of five independent variables:

$$N_{i,\mathrm{e}} - B_{\mathrm{e}};\quad 1/\rho_{i,\mathrm{e}};\quad 1/\varepsilon_{\mathrm{e}};\quad K_i \quad \text{et} \quad f_{i,\mathrm{e}}\left(k_1, k_2, \ldots\right).$$

Under these conditions, the combined standard uncertainty on the activity is estimated from the sum of the squares of the standard uncertainties corresponding to each of the variables:

$$\frac{u_{\mathrm{c}\,A_{i,\mathrm{e}}}}{A_{i,\mathrm{e}}} = \left[\frac{\left(\dfrac{N_{i,\mathrm{e}}}{t_1} + \dfrac{B_{\mathrm{e}}}{t_2} \right)}{\left(N_{i,\mathrm{e}} - B_{\mathrm{e}}\right)^2} + \frac{u^2_{\rho_{i,\mathrm{e}}}}{\rho^2_{i,\mathrm{e}}} + \frac{u^2_{\mathrm{c}\,\varepsilon_{\mathrm{e}}}}{\varepsilon^2_{\mathrm{e}}} + \frac{u^2_{K_i}}{K_i^2} + \frac{u^2_{\mathrm{c}\,f_{i,\mathrm{e}}(k_1,k_2,\ldots)}}{f_{i,\mathrm{e}}\left(k_1, k_2, \ldots\right)} \right]^{1/2} \tag{7}$$

In Equation (7), the t_1 and t_2 parameters represent the counting times for the sample spectrum and the background spectrum. The standard uncertainties for the first four components can easily be estimated. The standard uncertainty on the detection efficiency ε_{e} at energy e, in particular, is determined by smoothing the experimental points obtained using certified reference materials. For the last component, the estimate depends on the following:

- the geometric characteristics of the complete system comprising the package or sample, the detector and the collimator (if any);
- the nature of any screens inserted between the sample and the detector;
- the nature and properties of the matrix.

In the simplest hypothesis, $f_{i,\mathrm{e}}(k_1, k_2, \ldots)$ may be expressed as follows:

$$f_{i,\mathrm{e}} = C_{\mathrm{geo}} \cdot \mathrm{e}^{-\mu_{\mathrm{ec,e}} \cdot x_{\mathrm{ec}}} \cdot \frac{1 - \mathrm{e}^{-\mu_{i,\mathrm{e}} \cdot x_i}}{\mu_{i,\mathrm{e}} \cdot x_i} \tag{8}$$

where

C_{geo} represents the geometric factor, including the variation on waste height;

$\mu_{\mathrm{ec,e}}$ and $\mu_{i,\mathrm{e}}$ are the attenuation coefficients measured at energy e for the screens of thickness x_{ec} and for element i in the matrix of thickness x_i, respectively.

The standard uncertainty on $f_{i,\mathrm{e}}$ can thus be estimated as follows:

- either from simplified Equation (8), rewritten as follows:

$$\frac{u_{\mathrm{c}\,f_{i,\mathrm{e}}}}{f_{i,\mathrm{e}}} \approx \left[\frac{u^2_{\mathrm{c}_{\mathrm{geo}}}}{\mathrm{c}^2_{\mathrm{geo}}} + \left(\mu^2_{\mathrm{ec,e}} \cdot u^2_{x_{\mathrm{ec}}} \right) + \left(x^2_{\mathrm{ec}} \cdot u^2_{\mu_{\mathrm{ec,e}}} \right) + \frac{\left(\mu^2_{i,\mathrm{e}} \cdot u^2_{x_i} \right) + \left(x_i^2 \cdot u^2_{\mu_{i,\mathrm{e}}} \right)}{4} \right]^{1/2} \tag{9}$$

- or based on the experience acquired using modelling codes (Monte-Carlo method or other simulation methods):

$$\frac{u_{\mathrm{c}f_{i,\mathrm{e}}}}{\bar{f}_{i,\mathrm{e}}} \approx \frac{1}{\bar{f}_{i,\mathrm{e}}} \cdot \left[\sum_{j=1}^{n} \frac{\left(\bar{f}_{i,\mathrm{e}} - f_{j,i,\mathrm{e}} \right)^{2}}{(n-1)} \right]^{1/2} \qquad (10)$$

where

$f_{j,i,\mathrm{e}}$ represents the jth value obtained during n consecutive experiments;

$\bar{f}_{i,\mathrm{e}}$ is the mean value of the n measurements.

7.3 Detection limit

Spectrum-processing software calculates the decision threshold T_D and the detection limit L_D at a given energy.

The detection limit L_D, corresponding to the smallest value that the true measured quantity must have to be systematically detected with a given probability, can be calculated from the spectral data using a statistical approach.

The width of the region of interest that contain 99,7 % of the counts of a Gaussian peak is given by the relation:

$$I = 2{,}52\, R_{i,\mathrm{e}} \qquad (11)$$

where $R_{i,\mathrm{e}}$ is the FWHM at the energy i expressed in keV.

The net peak area S_n of the peak integrated in this interval is given by:

$$S_n = S_\mathrm{t} - S_\mathrm{b} \qquad (12)$$

where

S_t is the total peak surface area;

S_b is the continuous background under the peak ($S_\mathrm{b} = 2{,}52\, R_{i,\mathrm{e}} \cdot B_\mathrm{c}$, where B_c is the mean background amplitude, in events per keV, calculated over the interval I).

When S_n is very small compared with S_b, the detection limit is expressed as follows:

$$L_\mathrm{D} = \frac{C\sqrt{R_{i,\mathrm{e}} \cdot B_\mathrm{c}}}{t} \qquad (13)$$

where t is the package acquisition time and $C = 8{,}8$ if the α and β risks are equal to 2,5 %.

The detection limit is a function of the mean background level and of the acquisition time. In estimating L_D, the uncertainties of the $\frac{1}{\rho_{i,\mathrm{e}}}, \frac{1}{\varepsilon_\mathrm{e}}, K_i$ and $f_{i,\mathrm{e}}\left(k_1, k_2, \ldots\right)$ parameters are assumed to be negligible with regard to the counting uncertainty. The announced activity detection limit L_DA is then:

$$L_\mathrm{DA} = \frac{1}{\rho_{i,\mathrm{e}}} \cdot \frac{1}{\varepsilon_\mathrm{e}} \cdot K_i \cdot f_{i,\mathrm{e}}\left(k_1, k_2 \ldots\right) \cdot L_\mathrm{D} \qquad (14)$$

8 Validation of results

The absence of drift in the measurement system shall be verified regularly to guarantee the results obtained. This verification may be performed by periodically measuring a reference package or source, for example a sealed gamma-emitting source (e.g. ^{133}Ba or ^{152}Eu) for which a calibration certificate is available, or a reference package for which the constancy in time can be ensured (matrix, source position, source emission) or can at least be predicted and therefore corrected (e.g. source decay).

The verification interval is specified by the operator according to the specification characteristics: measurement rates, operating procedures, continuous or patch operation, type of operation (normal working hours, posts, etc.).

If a drift is detected by observing the reference measurements, (for example, through the use of inspection charts [7]), the cause shall be determined and corrected. After correction, all the packages measured between the last two reference measurements must be reinspected; in some cases, if the system fault condition is fully identified (date and consequences), the earlier results may be corrected.

9 Reporting of results

For traceability purposes, the measurement report must contain all the data necessary to validate the results. In particular, it must be possible to determine all the implicit measurement and interpretation conditions that do not directly affect the results obtained on the measured packages (background and calibration measurements used, reference library, etc.).

At least the following information should be recorded:

- time and date of measurement;
- package identification;
- package weight;
- calibration standard used;
- background measurement values used;
- nuclear data reference used;
- measurement parameters;
- raw measurement results;
- net measurement results corrected for background level;
- interpreted results (desired radioactive characteristic);
- corrections applied;
- uncertainties on results;
- detection limit per radionuclide considered;
- periodic inspections;
- reference spectrum;

- measurement anomalies;
- deviations with respect to input data.

The final results form intended for application of the measurement shall contain at least the following:

- package identification;
- time and date of measurement;
- activity (or L_{DA}) in becquerels, and related uncertainty values.

Annex A
(informative)

Radionuclide transformations — Energy and intensity of emissions (ICRP publication 38)

includes data for 817 radionuclides
See also revised and additional data on 162 radionuclides in references [2], [3] and [4]

FORMATION

RADIATION DATA

73	Tantalum	788	88	Radium	1061
74	Tungsten	822	89	Actinium	1070
75	Rhenium	834	90	Thorium	1082
76	Osmium	861	91	Protactinium	1096
77	Iridium	878	92	Uranium	1115
78	Platinum	911	93	Neptunium	1130
79	Gold	928	94	Plutonium	1153
80	Mercury	942	95	Americium	1174
81	Thallium	956	96	Curium	1194
82	Lead	982	97	Berkelium	1212
83	Bismuth	1004	98	Californium	1219
84	Polonium	1028	99	Einsteinium	1231
85	Astatine	1040	100	Fermium	1241
86	Radon	1047	101	Mendelevium	1249
87	Francium	1052			

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[14] NF M60-302:1999, *Nuclear energy — Nuclear fuel technology — Waste — High resolution gamma spectrometry activity measurement of waste packages*

[15] International vocabulary of basic and general terms in metrology BIPM, IEC, IFCC, ISO,IUPAC, IUPAP, OIML

[16] Guide to the expression of in uncertainty measurement BIPM, IEC, IFCC, ISO,IUPAC, IUPAP, OIML

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