Procedure for determining the specific activities of radionuclides in building materials by gamma spectrometry

K-γ-SPEKT-BAUST-01

Author:
U.-K. Schkade

Federal coordinating office for questions of monitoring of the radioactivity at enhanced natural radioactivity (ENORM)

(Leitstelle für Fragen der Radioaktivitätsüberwachung bei erhöhter natürlicher Radioaktivität (ENORM))

ISSN 1865-8725 Version April 2008

Procedures manual for monitoring of radioactive substances in the environment and of external radiation
(Messanleitungen für die „Überwachung radioaktiver Stoffe in der Umwelt und externer Strahlung")

Procedure for determining the specific activities of radionuclides in building materials by gamma spectrometry

1 Scope

The procedure described serves to determine the specific activities of mainly natural radionuclides in building materials and forms the basis for their radiological evaluation in accordance with the recommendation of the European Commission "Radiological protection principles concerning the natural radioactivity of building materials" (Radiation Protection 112) (1) and for the evaluation of residues in accordance with Part 3, Chapter 3 of the StrlSchV (2). This procedure is used to record the minimum expected specific activities for Ra-226 and Th-232 (Ra-228) of 10 $Bq \cdot kg^{-1}$ and for K-40 of 50 $Bq \cdot kg^{-1}$.

2 Sampling

The sample to be analysed must be representative of the building material or residue. The sample mass made available for testing should be at least 1 kg.

3 Analysis

3.1 Principle of the procedure

After drying and coarse crushing of the sample material, the specific activities of the radionuclides contained therein are determined by gamma spectrometry, preferably using high purity germanium detectors.

3.2 Sample preparation

3.2.1 Drying

Drying the sample material to a constant weight may be achieved at temperatures of up to 105 °C. Bricks and other voluminous materials should be crushed coarsely before drying.

3.2.2 Crushing and grinding

The coarsely crushed and dried building materials are milled with a suitable milling device to a grain size of less than 4 mm or crushed with a jaw crusher. If several milling processes are required as a result of a limited capacity of the milling vessel, the individual batches are to be combined and homogenised once milling has been completed.

ISSN 1865-8725 Version April 2008

3.2.3 Preparation of the counting source

The sample material must be filled into the measuring vessel in a way that the volume of the sample material cannot change until the end of the measurement and the measurement geometry specified by the calibration is retained. Measurements of short-lived daughter radionuclides of Rn-222 additionally require the sample material to be filled into gas-tight measuring vessels and the radioactive equilibrium to be maintained. The dead volume in the measuring vessels between the filled sample and the lid should be as small as possible.

3.3 Radiochemical separation

No radiochemical separation is required.

4 Measuring the activity

For basics of gamma spectrometry, it is referred to the chapters IV.1.1 through IV.1.4 of this procedures manual, with chapter IV.1.4 explicitly discussing particulars relevant to determining natural radionuclides in sample materials, e. g. establishing the radioactive equilibrium. These chapters also contain notes on summation and self-absorption corrections, which might need to be taken into consideration when measuring soil samples. If the densities of building materials, e. g. insulating materials, are significantly below 1 $g \cdot cm^{-3}$, the density may be approximated to the calibration conditions by addition of distilled water.

For measuring, preference should be given to a high purity germanium detector whose efficiency relative to a 3" x 3" NaI(Tl) crystal lies between 20 % and 60 % and whose full width at half maximum is smaller than 2,0 keV relative to the 1332-keV-gamma line of Co-60. For measuring natural radionuclides, it will be of advantage to employ an ultra-low level measuring configuration (detector and shielding), because it ensures a very low background for analysing the required energy lines of a number of natural radionuclides (e. g., U-238, U-235, Ra-226, Pb-210, Ac-228).

Calibrating the gamma spectrometer is described in detail in procedure F-γ-SPEKT-MILCH-01.

5 Calculation of the results

For calculating the gamma spectra, commercial high-performance software is available that comply with the requirements detailed in chapters IV.1.1 through IV.1.4 of this procedures manual.

Procedure C-γ-SPEKT-SEDIM-01 contains a portrait of how to calculate the specific activity of an individual nuclide, which may be applied analogously for calculating those of natural radionuclides, e. g., Ac-228 (Th-232, Ra-228) or K-40, too.

Gamma spectrometric determination of natural radionuclides is complicated by the fact that some radionuclides have identical gamma lines or at least lines that are so close to each other that they cannot be analysed separately. These cases call for corrections by means of other gamma lines or by other methods.

For example, the radionuclides Ra-226 and U-235: their gamma lines with an energy of 186,1 keV and 185,72 keV, respectively, cannot be determined separately. Even though the specific activity of U-235 in building materials accounts only for some $^1/_{22}$ of the specific activity of Ra-226, while the emission probability of the line of U-235 is higher by a factor of 16 than the one of Ra-226. This overlap does not present a problem if a radioactive equilibrium in the uranium-radon decay chain can be supposed, i. e., if the activity of U-238 equals that of Ra-226 and thus renders the activity of U-235 known. This does not necessarily apply to most soil samples, however, so that a correction is possible only if the activity of U-235 can be determined with the aid of other lines or derived from the activity of U-238.

Note
The specific activities of U-238 and U-235 in a sample may also be known from other measurements (e. g., determined by alpha spectrometry) or calculated from the results obtained for uranium analyses (e. g., by means of fluorimetry, phosphorimetry, X-ray fluorescence analysis, mass-spectrometry).

5.1 Equations

The following example is meant to illustrate this problem. The net count rate of the 186-keV-line is calculated according to equation (1), in which the radioactive decay need not be considered due to the long half-lives of U-235 (t_r = 7,037 · 10^8 years) and Ra-226 (t_r = 1,600 · 10^3 years).

$$R_n = (a_{Ra-226} \cdot p_{Ra-226} + a_{U-235} \cdot p_{U-235}) \cdot \varepsilon \cdot m_{DM} \quad (1)$$

From this follows equation (2) for determining the specific activity of Ra-226, $a_{Ra\text{-}226}$, as:

$$a_{Ra-226} = \frac{R_n}{\varepsilon \cdot p_{Ra-226} \cdot m_{DM}} - \frac{a_{U-235} \cdot p_{U-235}}{p_{Ra-226}} = \frac{R_{Ra-226}}{\varepsilon \cdot p_{Ra-226} \cdot m_{DM}} \quad (2)$$

in which

$$R_{\mathrm{n}} = R_{\mathrm{g}} - R_{\mathrm{T}}$$

$$R_{\mathrm{Ra-226}} = R_{\mathrm{n}} - R_{\mathrm{U-235}}$$

$$R_{\mathrm{U-235}} = a_{\mathrm{U-235}} \cdot p_{\mathrm{U-235}} \cdot \varepsilon \cdot m_{\mathrm{DM}}$$

In equations (1) and (2):

$a_{\mathrm{Ra-226}}$ specific activity of Ra-226, in Bq·kg^{-1} (DM);
$a_{\mathrm{U-235}}$ specific activity of U-235, in Bq·kg^{-1} (DM);
R_{n} net count rate, in s^{-1};
R_{g} gross count rate, in s^{-1};
R_{T} background count rate determined by the trapezoidal method, in s^{-1};
$R_{\mathrm{Ra-226}}$ net count rate of Ra-226, in s^{-1};
$R_{\mathrm{U-235}}$ calculated net count rate of U-235, in s^{-1};
ε detection efficiency at 186 keV, in Bq^{-1}·s^{-1};
$p_{\mathrm{Ra-226}}$ emission probability per disintegration for Ra-226;
$p_{\mathrm{U-235}}$ emission probability per disintegration for U-235;
m_{DM} dry mass (DM), in kg.

If the activity is measured with an ultra-low level measuring configuration, the contribution to the measuring effect by the background line at the gamma energy of 186 keV will be so small that it can be neglected. The standard uncertainty of the net count rate of Ra-226 is calculated according to chapter IV.5, section 4.7 of this procedures manual, applying equation (3) as follows:

$$s(R_{\mathrm{Ra-226}}) = \sqrt{\frac{R_{\mathrm{Ra-226}}}{t_{\mathrm{m}}} + \frac{1}{t_{\mathrm{m}}} \cdot \left[R_{\mathrm{T}} \cdot \left(1 + \frac{b}{2 \cdot L} \right) + R_{\mathrm{U-235}} + t_{\mathrm{m}} \cdot s^2(R_{\mathrm{U-235}}) \right]} \qquad (3)$$

in which

$$s^2(R_{\mathrm{U-235}}) = R_{\mathrm{U-235}}^2 \cdot \left(\frac{s^2(a_{\mathrm{U-235}})}{a_{\mathrm{U-235}}^2} + \frac{s^2(\varepsilon)}{\varepsilon^2} \right) \qquad (4)$$

For the standard uncertainty of the specific activity of Ra-226, equation (5) applies:

$$s(a_{\mathrm{Ra-226}}) = s(R_{\mathrm{Ra-226}}) \cdot \frac{a_{\mathrm{Ra-226}}}{R_{\mathrm{Ra-226}}} \qquad (5)$$

Aside from the symbols already defined, in equations (3) through (5):

$s(R_{\mathrm{Ra-226}})$ standard uncertainty of the net count rate of Ra-226, in s^{-1};
$s(R_{\mathrm{U-235}})$ standard uncertainty of $R_{\mathrm{U-235}}$, in s^{-1};

t_m duration of sample measurement, in s;

b base width of the gamma line in channels;

L number of channels for determining the background.

5.2 Worked example

Measuring the specific activity of Ra-226 from the gamma line shared by Ra-226 and U-235 at 186 keV produces the following data:

$a_{U\text{-}235}$	=	1,8 Bq·kg^{-1} (DM);	R_n	=	0,01339 s^{-1};
R_T	=	0,000954 s^{-1};	ε	=	0,026 Bq^{-1}·s^{-1};
$p_{Ra\text{-}226}$	=	0,0351;	$p_{U\text{-}235}$	=	0,572;
m_{DM}	=	0,230 kg (DM);	$R_{Ra\text{-}226}$	=	0,00723 s^{-1};
$R_{U\text{-}235}$	=	0,00616 s^{-1};	t_m	=	60000 s;
$s(R_{Ra\text{-}226})$	=	1,5·10^{-5} s^{-1};	b	=	2·L.

According to equation (2), the specific activity of Ra-226 amounts to:

$$a_{\mathrm{Ra-226}} = \frac{0{,}01339}{0{,}0026 \cdot 0{,}0351 \cdot 0{,}230}\,\mathrm{Bq \cdot kg^{-1}} - \frac{1{,}8 \cdot 0{,}572}{0{,}0351}\,\mathrm{Bq \cdot kg^{-1}} = 34{,}5\,\mathrm{Bq \cdot kg^{-1}}$$

The standard uncertainty of the net count rate of Ra-226 is calculated according to equation (3):

$$s(R_{\mathrm{Ra-226}}) = \sqrt{\frac{0{,}000723}{60000} + \frac{0{,}000954 \cdot 2 + 0{,}00616 + 60000 \cdot (1{,}5 \cdot 10^{-5})^2}{60000}}\ \mathrm{s^{-1}} =$$

$$= 5{,}05 \cdot 10^{-4}\,\mathrm{s^{-1}}$$

and according to equation (5), the standard uncertainty of the specific activity is calculated as:

$$s(a_{\mathrm{Ra-226}}) = 5{,}05 \cdot 10^{-4} \cdot \frac{34{,}5}{0{,}00723}\,\mathrm{Bq \cdot kg^{-1}} = 2{,}4\,\mathrm{Bq \cdot kg^{-1}}\ (\mathrm{DM})$$

5.3 Consideration of uncertainties

The uncertainty in determining the specific activity is mainly due to the uncertainty from counting statistics in the net count number and the uncertainty arising from determining the energy-dependent detection efficiency. The uncertainty in the U-235 count rate that has to be subtracted needs to be given particular attention. Other impact factors can be neglected. The total uncertainty must be expected to amount to ca. 10 %.

6 Characteristic limits of the procedure

6.1 Equations

The detection limit is calculated according to chapter IV.5 of this procedures manual, following equation (6):

$$g = \frac{\left(k_{1-\alpha} + k_{1-\beta}\right) \cdot k_{1-\alpha}}{2 \cdot t_{\mathrm{m}} \cdot \varepsilon \cdot m_{\mathrm{DM}} \cdot p_{\mathrm{Ra-226}}} \cdot \sqrt{1 + \frac{4 \cdot t_{\mathrm{m}}}{k_{1-\alpha}^2} \cdot \left[R_{\mathrm{T}} \cdot \left(1 + \frac{b}{2 \cdot L}\right) + R_{\mathrm{U-235}} + t_{\mathrm{m}} \cdot s^2(R_{\mathrm{U-235}})\right]} \tag{6}$$

Aside from the symbols already defined:

g detection limit of the specific activity of Ra-226, in Bq·kg^{-1} (DM);

$k_{1-\alpha}$, $k_{1-\beta}$ quantile of the normal distribution for considering errors of the 1st and 2nd kind.

6.2 Worked example

Using the example given above and inserting values of $k_{1-\alpha} = 3$ and $k_{1-\beta} = 1{,}645$, the detection limit of the specific activity of Ra-226 amounts to:

$$g = \frac{(3 + 1{,}645) \cdot 3}{2 \cdot 60000 \cdot 0{,}026 \cdot 0{,}23 \cdot 0{,}0351} \cdot \sqrt{1 + \frac{4 \cdot 60000}{3^2} \cdot [0{,}000954 \cdot 2 + 0{,}00616 + 60000 \cdot (1{,}5 \cdot 10^{-5})^2]}\ \mathrm{Bq \cdot kg^{-1}} =$$

$$= 8{,}5\ \mathrm{Bq \cdot kg^{-1}}\ (\mathrm{DM})$$

7 Catalogue of chemicals and equipment

7.1 Chemicals

No chemicals are required.

7.2 Equipment

— Gamma spectrometry measuring station consisting of:
  - Semiconductor made of high purity germanium with a relative efficiency relative to a 3 x 3" NaI(Tl) crystal of preferentially between 20 % and 60 % and a full width at half maximum of less than 2,0 keV relative to the 1332-keV-gamma line of Co-60.
  - Computer with software for analysing spectra;
  - Multi-channel analyser;
  - Measuring electronics;
— Drying cabinet;
— Ashing furnace;
— Freeze-drying installation;
— Jaw crusher or mill;
— Ring dishes or cans with screw-on lids;
— Basic laboratory equipment.

References

(1) European Commission: *Radiological protection principles concerning the natural radioactivity of building materials.* Radiation Protection 112, 1999.

(2) Verordnung über den Schutz vor Schäden durch ionisierende Strahlen (Strahlenschutzverordnung – StrlSchV), BGBl. I S. 1714 ber. I 2002 S. 1459, BGBl III 751-1-8, 20. Juli 2001.