Correction in determination of specific activity of radionuclides by gamma spectrometry in building materials

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Abstract. For more accurate determination of specific activities of radionuclides by gamma spectrometry in materials, it is necessary to take into account two important factors. These are the effect of true coincidences and self-absorption of gamma radiation directly in the measured sample. The corrections for these factors are determined for the specific measurement geometry defined by the detector, the measuring vessel and their relative positions. Corrections are calculated by Monte Carlo simulation. The results show a significant effect of material bulk density on the correction values.

1. Introduction

The content of natural radionuclides in building and solid materials is determined by gamma spectrometry [1, 2]. The detection device of the spectrometric assembly registers gamma radiation, which is emitted from the material during the radioactive decay of the contained radionuclides. The energy of the detected gamma radiation is characteristic for individual radionuclides. The output is an energy spectrum, from which it is possible to determine the specific activities of the radionuclides contained in the material after calibration of the measuring device.

Some radionuclides have more complicated decay pattern and they can emit gamma radiation of more energies over a very short period of time, which is detected by the device as a single event with the value of the sum of their energies. Therefore, it is appropriate to make corrections for true coincidences in the activities of these radionuclides [3, 4].

To quantify the specific activity of individual radionuclides, it is necessary to perform an efficient calibration of the spectrometer with a certified standard [5]. The different composition of the standard with which the spectrometer is calibrated and of the measured samples affects the values of the measured specific activity of radionuclides. A correction is necessary to obtain the correct value. The specific activities determined for the individual gamma energies of the given radionuclides are corrected.

At lower energies, the correction can reach tens to hundreds of percent [2].

The corrected specific activity $A_{cor}$ is obtained by multiplying the measured specific activity $A$ by corrections for self-absorption $K_s$ and by correction for true coincidences $K_c$.

$$A_{cor} = A \cdot K_s \cdot K_c$$

(1)

The size of the corrections is influenced mainly by material composition, material bulk density, the energy of the emitted gamma radiation and the geometry of the measurement.

For the normal operation of a laboratory that determines the specific activities of several radionuclides in many different materials, it is necessary to find a fast, inexpensive and sufficiently
accurate method for determining corrections. According to [6] and [7], where the authors compared several methods, the Monte Carlo simulation computational method seems to be a suitable approach.

2. Methods and materials

2.1. EFFTRAN
The program EFFTRAN [8] was chosen to calculate true coincidence summing correction and self-absorption correction. The software calculates self-absorption correction and true coincidence summing correction using Monte Carlo simulation.

The input data is the material composition of the detector, measuring vessel and measured sample. It is necessary to load the material bulk density into the program, the contained elements or compounds in the form of chemical formulas (e.g. SiO₂, Ge, H₂O) and their representation in the material in wt. %. The program creates a file with specified parameters and a file with calculated material attenuation coefficient for different energies of gamma radiation [3, 9, 10].

Furthermore, the detector parameters and measuring geometry are defined. The detector is characterized by dimensions and material composition, which is stated in the documents of the detector producer. The geometry of the measurement is determined by material and dimensions of the measuring vessel, the mutual position of the detector and the sample. Polypropylene cylindrical measuring vessel with a volume of 0.5 l was placed directly on the detector head.

Other parameters for calculating the attenuation of gamma radiation in the sample and self-absorption correction for selected radiation energies are the material of the calibration standard, the material of the measured sample and the efficiency of the detector [3].

While maintaining the same measurement geometry, the relation applies to the correction for self-absorption \( K_s \) as follows:

\[
K_s = \frac{\eta_{et}}{\eta_{sa}},
\]

where is:

\( \eta_{et} \) … detector efficiency for standard,
\( \eta_{sa} \) … efficiency for the measured sample.

2.2. Standard
When verifying the spectrometer, an efficiency calibration is performed with an activity standard of CBSS 2 type. The radioactive material is homogeneously dispersed in silicone rubber with a bulk density of 0.98 g/cm³. The material composition is given in table 1.

Table 1. Composition of standard.

| composition | Si  | C  | O  | H  |
|-------------|-----|----|----|----|
| wt. %       |     |    |    |    |
| SiO₂        | 37.9| 32.4| 21.6| 8.2|

2.3. Definition of materials
The most common building materials used in laboratory practice were selected. The material composition was found in the literature or chemistry analysis was performed. The chemical composition and bulk density of selected materials are given in table 2. The meaning of bulk density in this article is the ratio of the sample weight and the volume of measuring vessel. Samples of building materials are dried and crushed under 8 mm.
Table 2. Composition of building materials.

|        | plaster | cement | concrete | slag | fly-ash | brick |
|--------|---------|--------|----------|------|---------|-------|
| wt. %  |         |        |          |      |         |       |
| CaO    | 62      | 64     | 11       | 5    | 2       | 10    |
| SiO₂   | 16      | 20     | 63       | 52   | 53      | 58    |
| Al₂O₃  | 4       | 5      | 13       | 23   | 30      | 15    |
| SO₃    | 3       | 3      | 1        | 2    | 1       | 2     |
| Fe₃O₅  | 3       | 3      | 11       | 9    | 5       |       |
| H₂O    | 12      | 4      | 2        | -    | 1       | 1     |
| MgO    | -       | 1      | 2        | 2    | 4       |       |
| K₂O    | -       | -      | 3        | 4    | 2       | 3     |
| Na₂O   | -       | -      | 2        | 2    | -       | 2     |
| bulk   |         |        |          |      |         |       |
| density [g/cm³] | 0.8–1.8 | 1.5–1.8 | 1.1–2.4 | 0.7–2.2 | 0.7–1.6 | 1.0–1.6 |

2.4. Parameters

Natural radionuclides emit gamma radiation in the range of energy values 46–1764 keV during their decay. The standard analysis of building materials determines the specific activity of the three radionuclides ⁴⁰K, ²²⁶Ra and ²²⁸Th and the activity concentration index I calculated from them [1, 11]. The specific activity of ⁴⁰K is determined directly from one energy line. The specific activity of radionuclides ²²⁶Ra and ²²⁸Th are calculated as weighted activities from the three energy lines of their decay daughter products, with which they are in radionuclide equilibrium. Table 3 shows the line energies for determining the specific activity of radionuclides [2, 4].

Table 3. Gamma-ray energies in building materials.

| Energy [keV] | Radionuclide | Source of radiation |
|--------------|--------------|---------------------|
| 238.63       | ²²⁸Th        | ²¹²Pb               |
| 295.21       | ²²⁶Ra        | ²¹⁴Pb               |
| 351.93       | ²²⁶Ra        | ²¹⁴Pb               |
| 583.19       | ²²⁸Th        | ²⁰⁸Tl                |
| 609.31       | ²²⁶Ra        | ²¹⁴Bi               |
| 727.33       | ²²⁸Th        | ²¹²Bi               |
| 1460.80      | ⁴⁰K          | ⁴⁰K                 |

Activity concentration index I is calculated according to the formula [1]:

\[
I = \frac{A_{K}}{300} + \frac{A_{Ra}}{300} + \frac{A_{Th}}{200}
\]  

(3)

where is:

\( A_{K} \) … specific activity of ⁴⁰K,

\( A_{Ra} \) … specific activity of ²²⁶Ra,

\( A_{Th} \) … specific activity of ²²⁸Th.
3. Results and Discussion

3.1. Self-absorption correction

The size of self-absorption correction \( K_s \) is influenced mainly by material composition, material bulk density and the energy of attenuated gamma radiation [3, 10].

A concrete sample was selected to monitor the effect of bulk density on the correction size. The concrete composition in the calculations was the same but the value of the material bulk density changed. The shape of the curves of the dependence of \( K_s \) on the radiation energy, see figure 1, is identical with the shape obtained by experimental methods [6] and simulation [10]. The value of the self-absorption correction increases steeply for low energies below 100 keV [9].

The linear dependence of \( K_s \) on the bulk density is illustrated in figure 2. The slope of the line increases with decreasing of the radiation energy. There is a steep increase in the values of the slopes for energies below 100 keV. The value of \( K_s \) increases with increasing bulk density [12–15]. This can be explained by the fact that lower energy gamma radiation is significantly absorbed in the sample and therefore is not registered by the detector. Likewise, the materials with a higher bulk density significantly attenuate gamma radiation.

The bulk density of different kinds of building material varies a lot. However, analysing a large number of samples, it was possible to choose different types of building materials with the same bulk density (as it is mention above, bulk density in this article means the ratio of the sample weight and the volume of measuring vessel). The bulk density was chosen 1.6 g/cm\(^3\) as it fits to most of measured types of building materials. For some materials, e.g. fly ash, this is a limit value.

For building materials with the same bulk density and different compositions, the \( K_s \) values differ within a one percent difference to energy values around 150 keV. Below an energy of 100 keV, a significant influence of different composition of materials starts to be seen. The self-absorption correction increases significantly with decreasing value of gamma radiation energy, see figure 3. Bulk density of 1.6 g/cm\(^3\) was chosen to illustrate the effect of the composition and energy of the emitted radiation on the correction coefficients.

![Figure 1. Correction \( K_s \) of concrete with different bulk density.](image-url)
3.2. True coincidence summing correction

The values of true coincidence summing correction $K_c$ for different building materials with the same bulk density of 1.6 g/cm$^3$ are practically unaffected by the material composition for the mentioned representatives of building materials or only very weakly, see table 4.

| Energy [keV] | concrete | cement | brick | plaster | fly-ash | slag |
|--------------|----------|--------|-------|---------|---------|------|
| 238.63       | 1        | 1      | 1     | 1       | 1       | 1    |
| 295.21       | 1        | 1      | 1     | 1       | 1       | 1    |
| 351.93       | 1.001    | 1.001  | 1.001 | 1.001   | 1.001   | 1.001|
| 583.19       | 1.093    | 1.093  | 1.093 | 1.093   | 1.093   | 1.093|
| 609.31       | 1.077    | 1.077  | 1.077 | 1.077   | 1.077   | 1.076|
| 727.33       | 1.024    | 1.024  | 1.024 | 1.024   | 1.024   | 1.024|
| 1460.80      | 1        | 1      | 1     | 1       | 1       | 1    |

Table 5. Correction $K_c$ of concrete with different bulk density.

| Energy [keV] | 1.0 | 1.2 | 1.3 | 1.4 | 1.5 | 1.6 | 1.7 | 1.8 | 1.9 | 2.0 | 2.4 |
|--------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 351.93       | 0.999 | 1.001 | 1.001 | 1.001 | 1.001 | 1.001 | 1.001 | 1.001 | 1.001 | 1.001 | 1.000 |
| 583.19       | 1.082 | 1.086 | 1.088 | 1.09 | 1.091 | 1.093 | 1.095 | 1.097 | 1.099 | 1.101 | 1.110 |
| 609.31       | 1.067 | 1.07 | 1.072 | 1.074 | 1.075 | 1.077 | 1.079 | 1.08 | 1.082 | 1.084 | 1.091 |
| 727.33       | 1.021 | 1.022 | 1.022 | 1.023 | 1.024 | 1.024 | 1.025 | 1.025 | 1.025 | 1.026 | 1.028 |

True coincidence summing correction $K_c$ for selected most commonly measured building material, i.e. concrete, with different bulk densities is given in table 5. It is not necessary to perform true coincidence summing correction for gamma energies 238.63 keV, 295.21 keV and 1460.80 keV. A more significant effect of material bulk density on $K_c$ value is manifested especially at an energy of 583.19 keV and 609.31 keV. Within the stated interval of real values of concrete bulk density, the value of $K_c$ increased by 0.015 for the line 583.19 keV and by 0.014 for the line 609.31 keV as the bulk density increased from 1.2 to 2.0 g/m$^3$. For the 727.33 keV line, the increase was lower, only by 0.004. As the bulk density of the material increases, the value of $K_c$ also increases, see figure 4. This is then
reflected in the values of the corrected specific activities calculated as a weighted average of the specific activities determined from different energy lines.

![Figure 4. Correction $K_c$ of concrete.](image)

**3.3. Influence of correction on the correctness of determined parameters**

The relative deviation of the corrected values from the measured value was chosen as a quantity illustrating the effect of corrections on the determined parameters. The relative deviations $\delta$ of specific activities of radionuclides and activity concentration index are calculated according to relation (4):

$$\delta = \left( \frac{A_{cor,i}}{A_{mea,i}} - 1 \right) \cdot 100\%,$$

where is:

- $\delta$ ........ deviation,
- $A_{cor,i}$ .... corrected parameter,
- $A_{mea,i}$ .... measured parameter,
- $i$ ........ specific activity of $^{40}$K, $^{226}$Ra, $^{228}$Th and I.

The effect of the corrections of specific activities of radionuclides and activity concentration index for different real materials of different composition and bulk density is shown in figure 5. For materials with higher bulk density, a higher deviation of the corrected values from the measured ones is evident.

![Figure 5. Deviation of parameters of real building materials.](image)
4. Conclusion

The results show that the sample composition is not a determining factor for true coincidence summing correction and for self-absorption correction for radiation energies above 100 keV. The advantage of this method is that for the energies of radionuclides used in gamma spectrometry of building materials it is not necessary to analyse the material composition, it is enough to know the bulk density of the material. The EFFTRAN software is a fast and efficient way to obtain the self-absorption coefficient for an energy of 100–1700 keV.

For photon energies lower than 100 keV, which are used to measure the activities of radionuclides in radioactive substances like waste from NORM workplaces, e.g. $^{210}$Pb with energy 46.54 keV, the influence of the material composition of the sample on the size of the self-absorption coefficient is significantly applied. At such low photon energies, EFFTRAN can only be used with knowledge of the chemical composition of the sample. If the exact composition is not known, it is necessary to use another method.

Making true coincidence summing corrections and self-absorption corrections significantly contributes to determining the correct values of specific activities and activity concentration index. For the measurement geometry used, the results show that when no corrections are performed, especially at higher material densities, the specific activities are underestimated by up to 30%.

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