

## A METHOD FOR RAPID DETERMINATION OF ACTIVITY CONCENTRATIONS IN NORM USING GAMMASPECTROSCOPY

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### ABSTRACT

A method for rapid determination of activity concentrations in Naturally Occurring Radioactive Materials (NORM) using gamma spectroscopy is developed. A box of 250 mL is filled with NORM and its spectrum is counted for about five minutes. In the measured spectrum, the number of counts in five major peaks is used to calculate the activity concentrations of the  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  series. Assumptions are made with respect to equilibrium and composition. The overall maximum error is estimated to be about ten percent.

Special attention must be paid to abundances or deficits of  $^{210}\text{Pb}$  which can lead to an underestimation or an overestimation of the total activity concentrations, respectively.

### INTRODUCTION

In the earth's crust, trace amounts of the naturally occurring radioactive elements uranium and thorium are present. Products extracted directly or produced using minerals from the crust (i.e. scales, sludge, ores) will therefore contain uranium ( $^{238}\text{U}$ ,  $^{235}\text{U}$ ), thorium ( $^{232}\text{Th}$ ) and their decay-products.

In Dutch Legislation, a difference is made between solid natural radioactive substances and other radioactive substances. For solid natural radioactive substances an exemption limit of  $500 \text{ Bq}\cdot\text{g}^{-1}$  is given, while for all other radioactive substances a limit of  $100 \text{ Bq}\cdot\text{g}^{-1}$  is considered. Ores that did not undergo any chemical treatment (uranium and thorium series are in secular equilibrium) are therefore screened using the  $500 \text{ Bq}\cdot\text{g}^{-1}$  exemption level, while e.g. for sludges and scales the  $100 \text{ Bq}\cdot\text{g}^{-1}$  is used.

When NORM has to be investigated on location, the external radiation levels of substances or objects are screened and those that exceed a preset level (for example three times the background radiation level) are selected. Subsamples are taken (if possible at all) and analysed in the laboratory. The radionuclides from the  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  series in those materials (i.e. ores, scales, sludges) are not always in secular equilibrium. Therefore, to determine the total specific activities, several radionuclides from each series have to be taken into account. Moreover, exact knowledge of the composition and the density of the sample is necessary to calculate corrections (often lengthy) for the difference between the composition and density of the calibration source and the sample to be analysed, assuming an equal geometry.

This procedure cannot be applied for rapid in situ analyses. However, it is often needed to indicate on the spot a possible exceeding of limits. Therefore, a method is developed for screening purposes, to give within minutes an estimation of activity concentrations in ores, sludges and scales.

### METHOD

To perform analyses on the spot, RIVM has a van equipped with — among other things — a germanium detector and analysis software. A subsample of 250 mL of the material is taken for analysis. In case of large pieces of material (e.g. slags) it is grinded first. If the amount of material available is insufficient, quartz-sand is added and mixed with the material to make a total of 250 mL. The total mass of the 250 mL sample is determined, and in case of additional quartz-sand the mass of the original sample is determined. A polyethylene sample container (diameter 44 mm) is filled with the 250 mL material (filling height 41 mm) and a spectrum of this sample is recorded for five minutes on top of a high-purity germanium detector with an effective energy window of 80 to 2000 keV. The resulting spectrum is analysed and the peak areas for the peak energies 295 ( $^{214}\text{Pb}$ ), 351 ( $^{214}\text{Pb}$ ), 583 ( $^{208}\text{Tl}$ ), 609 ( $^{214}\text{Bi}$ ) and 911 keV ( $^{228}\text{Ac}$ ) are determined.

Because the detector is calibrated for a water sample ( $\rho=1$ ), the calibration must be corrected for the composition and density of the NORM-sample [1]. However, in the method presented here only corrections for density differences are calculated. The correction factor  $C_r$  is calculated as a function of the energy  $E$  and the density  $\rho$  of the sample using the following relation:

$$C_r(E, \rho) = \varepsilon(E, \rho) / \varepsilon(E, \rho=1) ,$$

where  $\varepsilon(E, \rho)$  is the efficiency of the detector for a gamma-ray with energy  $E$  emitted from a sample with density  $\rho$ .

To reduce the time needed for the analysis, particularly the often lengthy calculation of the correction factor,  $C_r(E, \rho)$  is tabulated for a wide range of densities ( $0 \leq \rho \leq 5 \text{ g}\cdot\text{cm}^{-3}$ ) for each of the five energies used in the calculation of the total activity concentrations. The results are shown in figure 1. For intermediate densities interpolation is used.

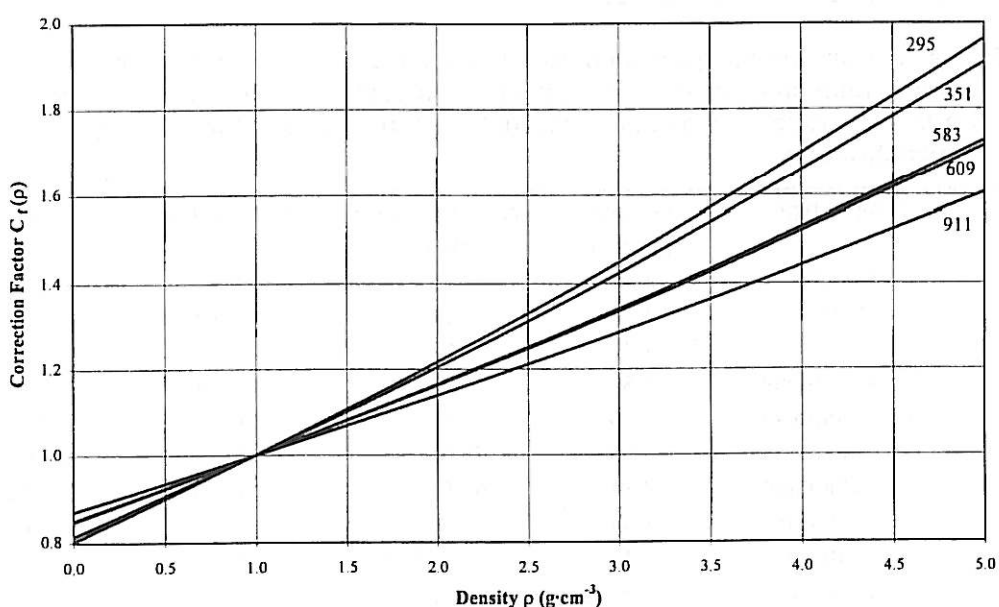


Figure 1. The correction factor as a function of the density of the sample for different gamma-ray energies.

The total activity concentration is calculated using the activity concentration of the nuclides  $^{214}\text{Pb}$  (295 and 351 keV),  $^{214}\text{Bi}$  (609 keV),  $^{228}\text{Ac}$  (911 keV) and  $^{208}\text{Tl}$  (583 keV). The nuclides  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  determine the total activity concentration of the  $^{238}\text{U}$  series and indirectly that of the  $^{235}\text{U}$  series, while the results of  $^{228}\text{Ac}$  and  $^{208}\text{Tl}$  determine the total activity concentration in the  $^{232}\text{Th}$  series. Furthermore the following assumptions are made:

- For ores, the  $^{238}\text{U}$  series is assumed to be in secular equilibrium, as well as the  $^{235}\text{U}$  series. The  $^{232}\text{Th}$  series is divided into subseries: from  $^{232}\text{Th}$  to  $^{228}\text{Ac}$  and from  $^{226}\text{Th}$  downwards, both in secular equilibrium.
- For scales, sludges and soils, the  $^{238}\text{U}$  series is assumed to be present only from  $^{226}\text{Ra}$  downwards and the  $^{235}\text{U}$  series from  $^{227}\text{Th}$  downwards. The  $^{232}\text{Th}$  series is present in two subseries: from  $^{228}\text{Ra}$  to  $^{228}\text{Ac}$  and from  $^{226}\text{Th}$  downwards. All subseries are assumed to be in secular equilibrium.
- The ratio between the  $^{238}\text{U}$  activity and the  $^{235}\text{U}$  activity is assumed to be 0.046, being the ratio for natural uranium. The contribution of  $^{235}\text{U}$  to the total activity concentration is a minor one altogether.

The calculations of the total activity concentration of each of the series are summarized in table 1.

**Table 1: Formulas describing the method of calculation of the total activity concentration for each series using the activity concentrations of the measured nuclides.**

Series	Ores	Scales, sludges
$^{238}\text{U}$	$14 \times [^{214}\text{Pb}, ^{214}\text{Bi}]$	$9 \times [^{214}\text{Pb}, ^{214}\text{Bi}]$
$^{235}\text{U}$	$11/14 \times 0.046 \times [^{238}\text{U-total}]$	$8/9 \times 0.046 \times [^{238}\text{U-total}]$
$^{232}\text{Th}$	$3 \times [^{228}\text{Ac}] + 7 \times [^{208}\text{Tl}]$	$2 \times [^{228}\text{Ac}] + 7 \times [^{208}\text{Tl}]$

## RESULTS AND DISCUSSION

To test the method, several samples analysed extensively in the laboratory were analysed again following the procedure described above. A variety of samples have been used for this comparison: ores with densities between 3 and 4, sludges from the gas-industry and scales with various composition. Whenever the scale was just present as a thin film on the object under investigation, only a few grams of scale could be taken, and quartz-sand was used to complete the 250 mL. The results are given in table 2.

**Table 2: Results from the comparison between the same material analysed in the laboratory and analysed using the rapid method described in here. The error in the ratio is 1s and is calculated from the errors in both activity concentrations.**

sample	sample type	density ( $\text{g}\cdot\text{cm}^{-3}$ )	reported value ( $\text{Bq}\cdot\text{g}^{-1}$ )	this method ( $\text{Bq}\cdot\text{g}^{-1}$ )	ratio
1	sludge	1.46	$22 \pm 4$	$25 \pm 3$	$0.88 \pm 0.19$
2	sludge	1.38	$52 \pm 8$	$52 \pm 5$	$1.00 \pm 0.18$
3	sludge	1.47	$41 \pm 7$	$45 \pm 5$	$0.91 \pm 0.19$
4	ore (cassiterite)	3.89	$770 \pm 40$	$780 \pm 80$	$0.99 \pm 0.11$
5	ore (columbite)	3.16	$323 \pm 19$	$320 \pm 30$	$1.01 \pm 0.12$
6	ore (columbite)	3.08	$740 \pm 30$	$760 \pm 80$	$0.97 \pm 0.11$
7	ore (columbite)	3.14	$1030 \pm 50$	$1110 \pm 110$	$0.93 \pm 0.10$
8	scale (thick)	1.44	$64 \pm 7$	$34 \pm 4$	$1.9 \pm 0.3$
9	scale (thick)	1.46	$0.84 \pm 0.10$	$0.6 \pm 0.2$	$1.4 \pm 0.5$
10	scale (thin)	1.51	$16800 \pm 1000$	$15800 \pm 1600$	$1.06 \pm 0.12$
11	scale (thin)	1.51	$10900 \pm 600$	$12200 \pm 1200$	$0.89 \pm 0.10$

As can be seen from table 2, agreement is good, except for sample 8. The scale of sample 8 contains an abundance of  $^{210}\text{Pb}$  ( $^{210}\text{Pb}/^{214}\text{Pb} = 4$ ) and the activity of this scale is hence underestimated by a factor of two using the rapid method. An abundance of  $^{210}\text{Pb}$  with the ratio  $^{210}\text{Pb}/^{214}\text{Pb} = 10$  can even underestimate the activity by a factor of four.

Materials that undergo high temperature treatment (e.g. slags) may show a deficit of  $^{210}\text{Pb}$ . In that case the method will overestimate the activity concentrations.

## REFERENCES

- [1] Overwater R.M.W., Bode P. and De Goeij J.J.M., "Gamma-ray Spectroscopy of Voluminous Samples: Corrections for Source Geometry and Self-Attenuation", Nuclear Instruments and Methods A324, 1993, 209-218.