Gamma-spectroscopic analysis of NORM samples

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Gamma-spectroscopic analysis of NORM samples

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Abstract   Two samples of Naturally Occurring Radioactive Materials (NORM) were studied using gamma-spectrometry in order to determine their radioactivity levels. The samples consisted of phosphate ore and phosphogypsum, a by-product of the phosphate fertilizer industry. Customized containers of standard geometry were filled with the above materials and then sealed with epoxy resin in order to prevent gaseous isotopes from emanating. The samples were measured by means of gamma spectrometry using a High-Purity Germanium detector. The full energy peak efficiency calibration of the detector was performed experimentally, using reference volume sources of the same geometry and density as those of the samples. To account for the true coincidence summing effect and the self-attenuation of gamma-rays within the samples volume, appropriate correction factors were calculated using the TrueCoinc software and the MCNP code, respectively. In both samples, ^{228}Ac, ^{214}Bi, ^{235}U, ^{214}Pb, ^{212}Pb, ^{234}Th, ^{208}Tl and ^{40}K were detected, with activity levels varying per isotope. The results of the present study provide information on the activity levels of NORM, which is important for Occupational Radiological Exposure assessments of workers in industries involving NORM, such as the phosphate fertilizer industry, as well as for the protection of the environment.

Keywords   NORM, phosphate ore, phosphogypsum, gamma spectrometry, activity determination

INTRODUCTION

Naturally Occurring Radioactive Materials (NORM) have been the subject of interest and research for many years, due to their presence in various natural resources and industrial processes. The interest in these materials stems from the need to prevent any potential hazards and impact on human health and the environment. Therefore, knowledge of NORM activity levels contributes to the development of safety measures, regulatory frameworks and best practices for managing these materials in a responsible and sustainable way. The purpose of this work was to study, in terms of activity concentration, two samples of phosphate ore and phosphorus gypsum, two NORMs produced in industrial areas related to the extraction and processing of phosphorus. The work provides information on the preparation, experimental measurements and simulations which were carried out to determine the activity levels in each sample.

MATERIALS AND METHOD

Samples Preparation

Two containers of standard geometry were filled with the sample materials (Fig. 1, left). The humidity of the samples was determined by drying amounts of the materials examined. Two samples of 5 g and 10 g from each material were dried at 80°C overnight in a drying oven (Fig. 1, right) and sample humidity was determined by weighting the samples before and after drying.

All three natural radioactive series are expected to be detected in the studied materials, i.e. the ^{238}U, the ^{232}Th and the ^{235}U series with all their daughter isotopes. Among them, of crucial importance is Radon, the immediate decay product of radium isotopes, which is a noble gas and can escape from the place where it is produced. Namely, ^{222}Rn has a sufficiently long half-life of 3.825 d and can be

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released from the soil and rock, where it is generated, resulting in the disturbance of the radioactive series equilibrium. To avoid this, the samples were sealed with an epoxy resin, which prevents all gases to escape the sample containers. The samples were then analyzed, after adequate time of 6-7 half-lives of ^{222}Rn, to allow for the restoration of the radioactive equilibrium.

Figure 1. *Samples in the containers used (left) and during the drying procedure (right)*

Gamma spectrometry

Both samples were measured by means of gamma spectrometry using a coaxial High-Purity Germanium (HPGe) detector of 85% relative efficiency. Efficiency calibration was performed using a combination of reference volume sources, of the same geometry as that of the samples, and reference point sources. In particular, due to the decay of several isotopes of those initially included in the reference volume sources, their use provided efficiency data only for four energies, namely not sufficient for the calibration curve. Thus, in order to cover the full energy range of interest and due to lack of other reference volume sources, additional reference point sources were used to provide efficiency values for a variety of energies. These values were subsequently used in conjunction with efficiency transfer, using Monte Carlo code MCNP [1], to derive the full energy peak efficiency (FEPE) curve. Figure 2 shows the derived FEPE as a function of photon energy for the volume source geometry.
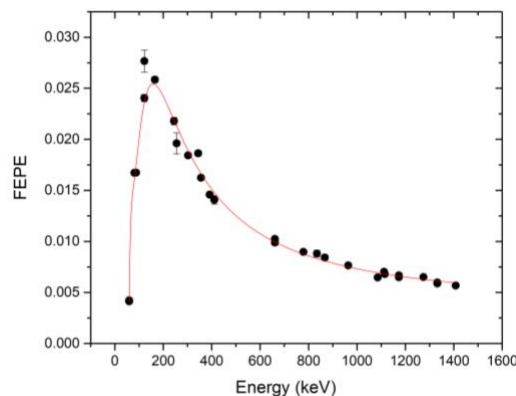

Figure 2. *FEPE as a function of photon energy for the volume source geometry*

The samples were measured 90 days after the sealing at 1 cm distance from the detector endcap for 24 h. Background measurements of the same duration were collected to account for the contribution of background radiation. Spectral analysis was performed with GammaVision™ software, and the activity was calculated using the following formula:

$$A = \frac{N_{net}}{t_c \times FEPE \times yield \times f_{TCC} \times f_\gamma} \tag{1}$$

where

- N_{net} : Number of net counts under the photopeak of interest

- t_c : Measurement duration
- $yield$: Emission probability
- f_{TCC} : Correction factor for true coincidence phenomenon
- f_γ : Correction factor for gamma self-attenuation within the samples

Monte Carlo simulations

The Monte Carlo code MCNP [1] was used to predict the detector efficiency, where no experimental data were available, and for self-attenuation calculations. For the efficiency calibration, the reference sources were described in detail and an experimentally validated model of the HPGe detector was used for the calculation of the full energy peak efficiency of the point and volume sources. The ratio of these values was then used as the efficiency transfer factor, in order to calculate the efficiency of the volume sources in the energy range of interest. For the self-attenuation study, the experimental set-up was simulated first with the plastic container empty and then with the plastic container filled with the sample. Gamma self-attenuation correction factors were calculated as the ratio of the FEPE with the sample over the FEPE without the sample. Figure 3 shows the calculated self-attenuation correction factor as a function of photon energy for the phosphate ore sample.
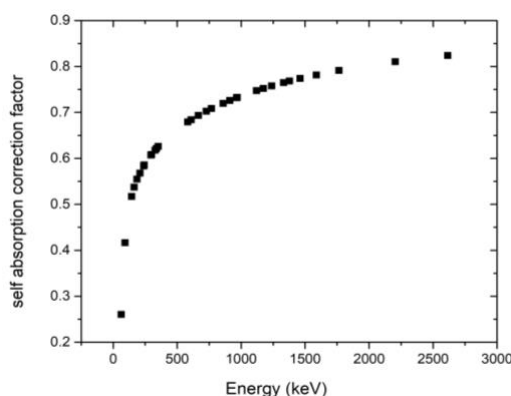

Figure 3. *Self-attenuation correction factor as a function of photon energy for the phosphate ore sample*

True coincidence corrections

Several of the detected isotopes of interest are cascade emitters, thus, the probability of two or more photons to be recorded by the detector as one, with energy equal to the summation of the individual photons, creating a sum-peak, is very high. True coincidence correction factors are necessary in such cases and in the present study they were calculated using the TrueCoinc program [2]. The program requires the user to provide the FEPE and the total efficiency curves and to select the isotope of interest through the program's database. In this work, FEPE curve was calculated as described above, using both volume and point sources, while the total efficiency curve was calculated by means of Monte Carlo simulation using the MCNP code. To avoid uncertainties due to geometry inaccuracies between the model and the experiment, the total efficiency curve was introduced in the program as a function of the full energy peak efficiency (total-to-peak ratio).

RESULTS AND DISCUSSION

In both samples studied, several isotopes were detected originating from all natural radioactive series. Namely ^{234}Th, ^{214}Bi and ^{214}Pb were detected from the ^{238}U series, ^{235}U from the respective series, ^{228}Ac, ^{208}Tl and ^{212}Pb from the ^{232}Th series, as well as ^{40}K. An indicative gamma-ray spectrum of the phosphate ore sample, measured 90 days after the sealing, is shown in Fig. 4.
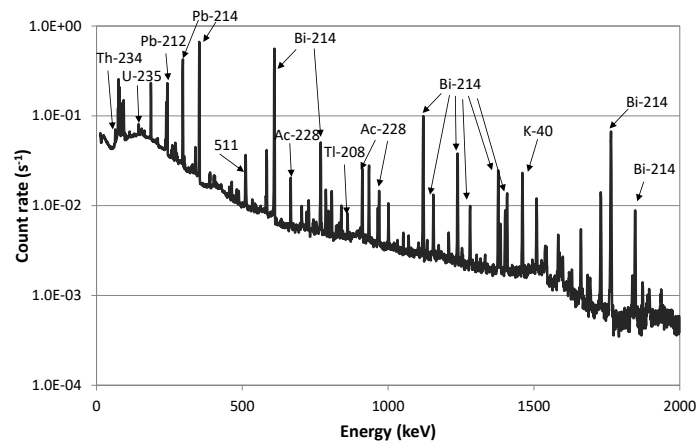
Figure 4. *Gamma-ray spectrum of phosphate ore sample, measured 90 days after the sealing*

In Table 1 the activity concentration results for all detected isotopes of interest are presented for both samples. In case several photopeaks of the same isotope were detected, the weighted average activity was calculated taking into consideration both the uncertainty of the measurements and the standard deviation of the different results. As shown in Table 1, the activities of the isotopes vary between the two samples, which is attributed to the different origin of the samples. In both samples, however, the results are within the anticipated activity range and confirm the expected radioactive equilibrium.

Table 1. *Activity concentration results for the two samples*

Isotope	Activity Concentration (Bq/kg dry mass)	
	Phosphate ore	**Phosphogypsum**
Ac-228	47.7 ± 2.6	22.4 ± 1.4
Bi-214	628.5 ± 18.7	311.4 ± 20.0
U-235	46.9 ± 3.0	19.9 ± 3.5
Pb-214	643.2 ± 26.0	310.2 ± 17.3
Pb-212	52.2 ± 5.7	18.0 ± 3.2
Tl-208	14.9 ± 0.8	5.9 ± 0.4
K-40	42.3 ± 2.6	50.0 ± 3.1
Th-234	417.5 ± 21.1	147.2 ± 16.6

CONCLUSIONS

In both samples several isotopes were detected, all of which are naturally occurring. The activity concentration values are within the expected range and relatively low in the studied samples of phosphate ore and phosphorus gypsum. However, high levels of radioactivity may occur in NORMs, so it is of outmost importance to monitor their activity levels, aiming to restrict radiation exposure of the employees and other persons who may be affected by their work with such materials and to ensure that the derived working limits in the industry are successfully met.

References

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