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A fast method for low-level actinide measurement in concrete

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Abstract

During the dismantling of nuclear installations much building material must be disposed of. The material may be potentially contaminated by low-levels of α radiation-emitting radionuclides. Because the contamination levels are often around the legal activity levels, an accurate and quick method is essential to determine whether or not this material can be treated as radioactive or non-active waste. A direct measurement of α radiation in the building material is impossible because the α particles are absorbed by the concrete. Chemical analysis of the α contaminants included many time-consuming analytical steps and is therefore rather unsuitable. We use direct α-spectrometry after only mechanical preparation of the concrete samples. The method is very efficient and requires only 1 day. © 1998 Elsevier Science S.A.

Keywords: Actinide; α-Spectroscopy; Concrete; Low-level radiation

1. Preparation and measurement

Because the α radiation has only a short range and is absorbed in concrete layers, extremely thin and large area sources are necessary to directly measure α radiation in concrete.

Samples were collected according to screening radiation measurement guidelines laid down by the Ministry of the Environment in the German state were the building is located.

We obtained particles with an average diameter of 0.5 μm by crushing the concrete in a two-step process. First the concrete is broken with a jaw breaker into particles with diameters about 0.2 mm. Another possibility for the first fragmentation is pulsed electrical discharge [1] which produces particles as small as 0.02 mm. During this crushing step the entire sample is homogenized. In a second step, the concrete is wet milled with special hard metal milling spheres. The milling suspension is diluted with water and the measuring sources are prepared by pouring or spraying the suspension onto plates and subsequent drying. To prevent the particle agglomeration during these preparation steps, the suspension has to be stabilized. Measurement of the pH dependence of the electrochemical potential (ESA signal) and determination of the isoelectrical point (pH_{IEP}) provides information about the working range to be adjusted. This should be at a pH value far from the pH_{IEP}, in these experiments more than 2 pH units above the pH_{IEP}. We achieved layers of about 1 μm thickness.

The α spectra of these sources were measured using a large grid ionization chamber. The diameter of the sources was 200 mm.

2. Results

Using standard concretes with added actinides, we determined the peak shape and the self absorption of the α radiation as the function of the sample layer's thickness. Fig. 1 shows the direct comparison of standard actinide concrete samples of various layer thicknesses with 'massless' samples containing the same activity.

The spectral resolution of the 1-μm sources are comparable to ideal 'massless' spectra.

We fitted and deconvoluted the spectra of unknown samples with the peak shape of standard samples having the same thickness. The measured α spectra of the actinides in a contaminated sample are shown in Fig. 2. Fig. 2a shows the spectrum of a concrete from a water collection tank for contaminated waters at the Rossendorf site. The total specific activity of this concrete is (1.18 ± 0.04) Bq g^{-1}. Fig. 2b depicts the spectrum of 'inactive' concrete without actinide contamination having a

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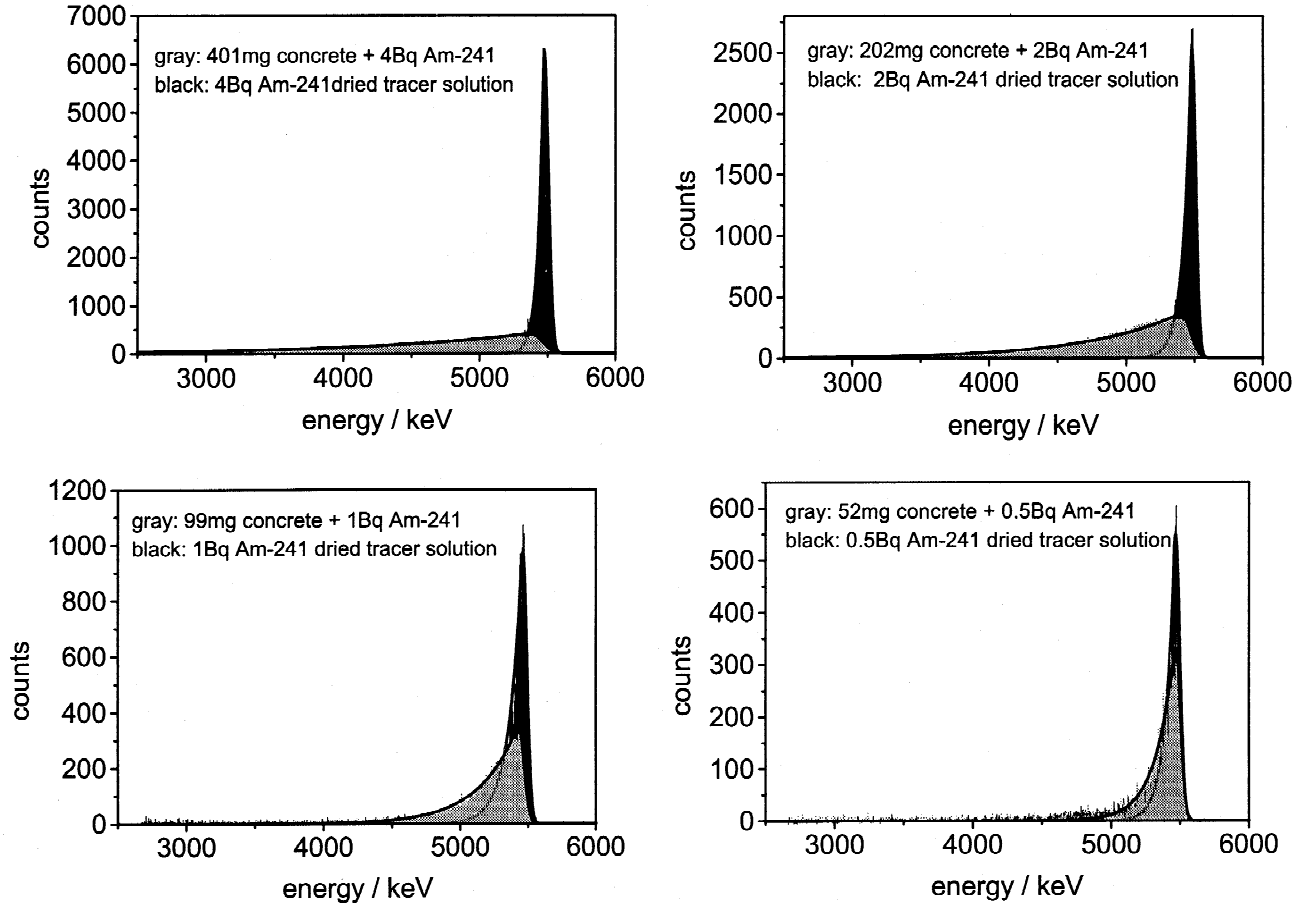

Fig. 1. Comparison between standard concrete and 'massless' samples with the same activity and geometry.

total activity of 0.43+0.03 Bq g^{-1}. The spectrum in Fig. 2c was generated by subtracting the spectrum of the 'inactive' concrete from the spectrum of the actinide-contaminated sample; the difference between the spectra in Fig. 2a and Fig. 2b. The total activity of 0.72 Bq g^{-1} is distributed in five different energy peaks. The spectral resolution is about 100 keV. To distinguish the spectra of americium-241 and plutonium-238, a better resolution than 100 keV is required. We can only measure the sum of both nuclides.

The specific activity of 'inactive' concrete was between 0.14±0.01 and 0.78±0.01 Bq g^{-1}. This α activity is caused by natural uranium and thorium and their decay products (which are also depicted in Fig. 2b). Because different 'inactive' concrete samples may contain a varying amount of natural radioactivity, it is advantageous to know the exact background activity for each sample to improve the accuracy of the net activity calculation by background subtraction. With this method α-emitting radionuclides as low as 0.01 Bq g^{-1} can be detected.

3. Summary

Direct α activity measurements of actinides with extremely thin and large-area sources can be done within 1 day and neither require thermo-chemical treatment of the concrete nor chemical separation and electrochemical deposition.

The peak width of the counting signal is directly proportional to the thickness of the sample layer, at nearly the same peak height. The spectra with the smallest peak width are comparable to the peaks in ideal 'massless' spectra. From a series of calibrations that relate the peak shape and width to the thickness of the sample layer, it is possible to fit and deconvolute spectra with an unknown

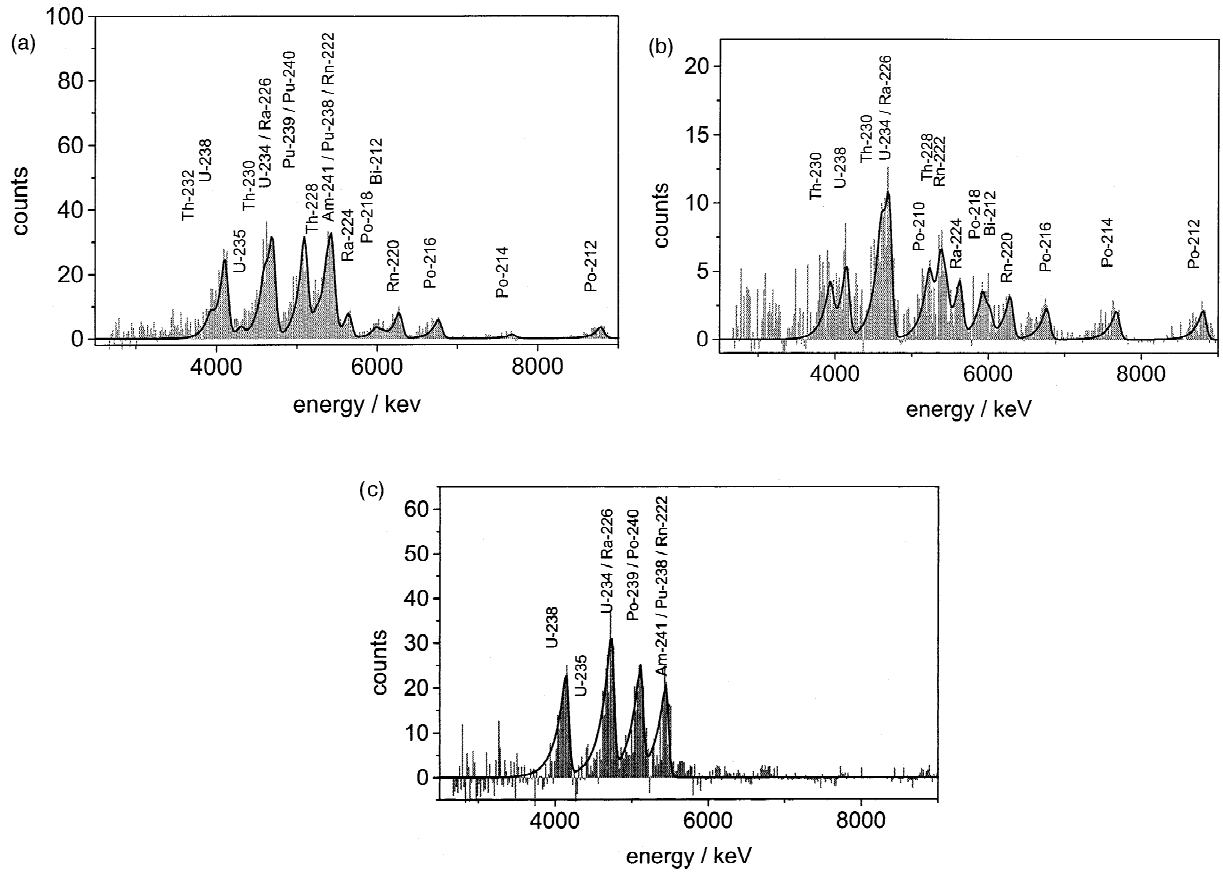

Fig. 2. Comparison of contaminated and 'inactive' concrete. (a) Contaminated concrete (background spectrum subtracted); (b) spectrum of 'inactive' concrete (background spectrum subtracted); (c) contaminated concrete (spectrum of 'inactive' concrete subtracted).

number and content of actinides up to 0.01 Bq g^{-1} per nuclide.

Acknowledgements

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