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Activation Analysis of Soils and Rocks in Norway

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A method is described to identify elements giving the greatest contributions to thermal-neutron induced gamma activities. The complex gamma spectrum is analysed for soils and rocks from different parts of Norway.

The purpose of this work was to obtain information about elements in rocks and soils contributing significantly to the gamma radioactivity induced by thermal neutrons.

There are few chemical analyses of Norwegian rocks and soils suitable for estimating neutron-induced radioactivity in which the abundances of particular elements are a primary consideration. The decisive factors are the thermal neutron cross sections and the abundance of the stable isotopes of the elements in question, and the decay characteristic of the activation products. It is important that an element present in tracer amount, not easily detectable by chemical methods, may contribute significantly to the induced gamma activity.

To obtain the required information neutron activation and subsequent gamma analysis of soils and rock samples was initiated.

EXPERIMENTAL

The neutron activation was carried out in the Jeep reactor (IFA, Kjeller, Norway), in two steps. A 2 g part of the sample was activated in low neutron flux for a short period (10^8 neutrons cm^{-2} sec^{-1}, 5 min) and counted in the gamma spectrograph from about 5 min after exposure. We were then able to obtain the spectra from isotopes with half lives in the range from about 1 min to about 15 h, by counting at various intervals.[1] A typical series of spectra is given in Fig. 1.

Another 2 g part of the sample was activated in high neutron flux for a longer period (10^{12} neutrons cm^{-2} sec^{-1} for 30 min). By waiting 4–5 days before counting we were able to measure isotopes with half lives of more than about 15 h. A typical series is shown in Fig. 2.

In all neutron activations a known amount of sodium chloride was bombarded together with the samples. By measuring the induced ^{24}Na activity, the product flux × time could be standardized.

The gamma spectrograph consisted of a 3″ × 3″ NaI crystal and a 256 channel pulse height analyser. The crystal was mounted in a lead chamber to reduce the background.

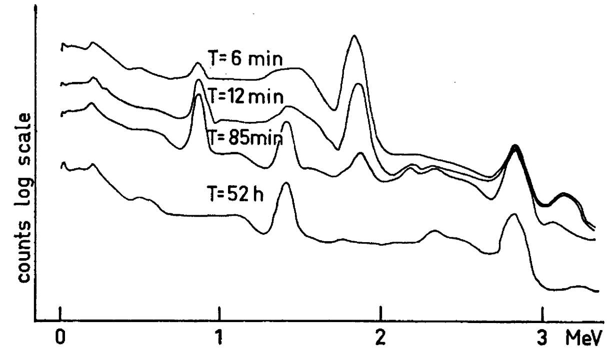

Fig. 1. Gamma spectra of a soil sample (Namsos) at various times T after the neutronactivation for 5 min in a flux 10^8 neutrons cm^{-2} sec^{-1}.

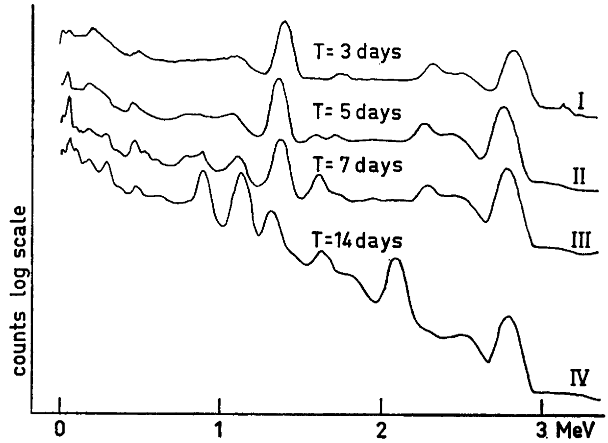

Fig. 2. Gamma spectra of a soil sample (Sola, Stavanger) at various times T after the neutronactivation for 30 min in a flux 10^{12} neutrons cm^{-2} sec^{-1}.

ANALYSIS OF THE GAMMA SPECTRA

Gamma spectroscopy is much used for activation analysis. Several computation methods for resolving complex gammaspectra have been described.[2-4] In our case we are concerned with a large energy range with great complexity that makes the so called "strip off" method unsatisfactory. This method was, however, used to unscramble the contribution from the high energy gamma rays to detect the hidden photopeaks of lower energy. This was performed by electronic computation and subsequent outprint of the resultant spectra. In this way most of the elements contributing to the gamma spectra were identified.

In the quantitative analysis the standard spectra of the contributing elements were first taken by neutron activation of pure chemicals. The number of recorded pulses c in the channel i is given by the matrix equation

$$[c_i] = [\mathrm{A}_{ij}]\,[\mathrm{S}_j]^* \tag{1}$$

where

$$[\mathrm{S}_j]^* = \begin{bmatrix} \mathrm{S}_1 \\ \mathrm{S}_2 \\ \cdot \\ \cdot \\ \cdot \\ \mathrm{S}_j \end{bmatrix} \tag{2}$$

gives the amount of the contributing elements, and A_{ij} is the contribution to the i'th channel from the standard of the j'th element: The A_{ij} matrices are then the normalised standard spectra recordings.

If we know all the elements j, we can solve the equation

$$[c_i^1] = [c_i]$$

c_i^1 is the number of recorded pulses in channel i. This equation can be solved for the S_j values.[2]

An exact solution is of course impossible. The most often used solving criterion is the least square criterion

$$\sum_{i=1}^{k} (c_i^1 - c_i)^2 = \text{minimum} \tag{3}$$

Partial differentiation will give an equation in S_j with an exact solution.

This criterion is not the best when there is an equal interest for photopeaks over a large energy range, as the amount of recorded pulses often drops off with a factor of 10^3 from the low to the high energy gamma rays.

In this work was used a criterion

$$\sum_{i=1}^{k} \frac{(c_i^1 - c_i)^2}{c_i^1} = \text{minimum} \tag{4}$$

which gives an equation that has an exact solution in S_j

$$\left[\frac{c_i^1 - c_i}{c_i^1}\right]^* [\mathrm{A}_{ij}] = 0 \tag{5}$$

where the asterisk indicates that the matrix is transformed.

This criterion for a minimum matches the theoretical spectrum, constructed by known standards, to the experimental spectrum at all energies according to the experimental accuracy of the recordings. As is seen from eqn. (4) the contribution from the different c_i is proportional to the standard deviation.

Table 1. Amounts of some elements in a collection of soil samples given in grams of the element in a gram soil.

Samples	Al	Mn	Cu	K	Na	Br	La	Fe	Sc
Rock, Namsos Gneiss	6.05×10^{-2}	1.21×10^{-3}	$\lesssim 10^{-4}$	4.2×10^{-2}	2.50×10^{-2}	—	1.40×10^{-4}	1.4×10^{-1}	1.8×10^{-5}
Rock, Hinnøy Gneiss	1.0×10^{-1}	6.75×10^{-4}	$< 3 \times 10^{-4}$	5.9×10^{-2}	3.15×10^{-2}	—	—	—	—
Rock, Hinnøy Glimmerslate	8.0×10^{-2}	1.16×10^{-3}	$< 4 \times 10^{-4}$	$\approx 4 \times 10^{-2}$	3.9×10^{-2}	$< 5 \times 10^{-6}$	6.6×10^{-5}	1.0×10^{-1}	2.1×10^{-5}
Soil, Stjørdal Sandy, pine forest	4.3×10^{-2}	7.8×10^{-4}	$< 10^{-4}$	$\approx 1 \times 10^{-2}$	1.7×10^{-2}	6×10^{-6}	5.5×10^{-5}	7.7×10^{-2}	1.7×10^{-5}
Soil, Porsangerfjord Calcareous sand, leaf forest	5.9×10^{-2}	4.63×10^{-4}	$\lesssim 10^{-4}$	$\approx 1 \times 10^{-1}$	3.0×10^{-2}	$< 5 \times 10^{-6}$	4.7×10^{-5}	5.2×10^{-2}	1.1×10^{-5}
Soil, Sjørdal Sandy, corn field	5.1×10^{-2}	1.32×10^{-3}	$\lesssim 10^{-4}$	5×10^{-2}	1.85×10^{-2}	$< 10^{-6}$	7.2×10^{-5}	1×10^{-1}	1.6×10^{-5}
Soil, Leksdalsvann Sandy, field	8.5×10^{-2}	1.54×10^{-3}	$< 2 \times 10^{-4}$	5×10^{-2}	2.02×10^{-2}	2×10^{-5}	8.7×10^{-5}	1.2×10^{-1}	2.1×10^{-5}
Soil, Målselv Mould, field	3.2×10^{-2}	2.40×10^{-4}	$\lesssim 10^{-4}$	1×10^{-2}	7.8×10^{-3}	—	—	—	—
Soil, Namsos Sand, brushwood	5.45×10^{-2}	7.25×10^{-4}	$< 2 \times 10^{-4}$	3×10^{-2}	2.25×10^{-2}	$< 5 \times 10^{-6}$	5.7×10^{-5}	5.7×10^{-2}	1.13×10^{-5}

As the c_i^1 does not necessarily mean pulses registred in one or a certain number of channels, the spectrum can be divided into groups "*i*" so that each group contains one photopeak. If there is a wide energy range without any photopeak this can be omitted from the computation in eqn. (4).

RESULTS

In Table 1 are tabulated the amounts of the detected elements together with the sampling locations. The results given are uncertain in the last figure. The uncertainity for any one element depends upon the amounts of others. In general the amount of sodium limits detection of the shorter lived isotopes.

The approximate limit for detection of copper is Cu:Na $\approx 10^{-2}$ and for potassium K:Na ≈ 1. For detection of bromine the amount of lanthanum is the limitation Br:La $\approx 10^{-1}$. The limits depend upon the frequency of spectral recordings for a given sample. The measurements times are not optimal for all elements in the considered samples.

Another problem not considered here is the representativity of samples. Broadly, this question might be answered for a particular location by evaluating the geological structure in the area *etc*. Beside the listed elements, there are others that can be determined easily, but the standards for *e.g.*, chromium, europium and samarium could not be prepared during the short period of the measurements. The elements zinc, caesium and cobolt can be found in an amount of about 10^{-6} g per g soil, but the neutronactivated sample needs a decay time of about 100 days before the radioactive isotopes of these elements are detectable.

CONCLUSIONS

The method described was found to be useful for resolving spectra where some of the peaks could hardly be seen by the eye.[1]

As there is a growing interest in tracer elements, such as the rare earths, scandium, bromine, zinc, *e.g.* in agriculture and biological research, the method can be a useful tool in these fields. The low limit for detection quoted can be reduced considerably and can thus meet special requirements. It can be used in routine analysis of a large number of samples.

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