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DETERMINATION OF Th- AND U-ABUNDANCES IN ROCKS BY GAMMA-RAY SPECTROSCOPY

by

P. HOLMBERG and R. RIEPPO

Department of Physics
University of Oulu

Abstract

A method for determining the abundances of natural thorium and uranium in rocks by means of gamma-ray spectroscopy is discussed. Th/U-ratios as well as absolute abundances of Th and U are obtained using Ge(Li)- and NaI(Tl)-spectroscopy methods.

Introduction

Several rocks contain small amounts of thorium and uranium. The tracing of these elements is of interest, and there have been developed different methods for this purpose. In the present report we discuss the use of gamma-ray spectroscopy for determining thorium and uranium abundances in rocks.

There are two long-lived isotopes in natural thorium and uranium, namely ^{232}Th and ^{238}U, with half-lives of 1.41×10^{10} y and 4.51×10^{9} y, respectively. In natural material these isotopes and their decay products are in radiative equilibrium. The main decay processes of the Th- and U-series are shown in Figs. 1 and 2. In the Th-series, the radioactive

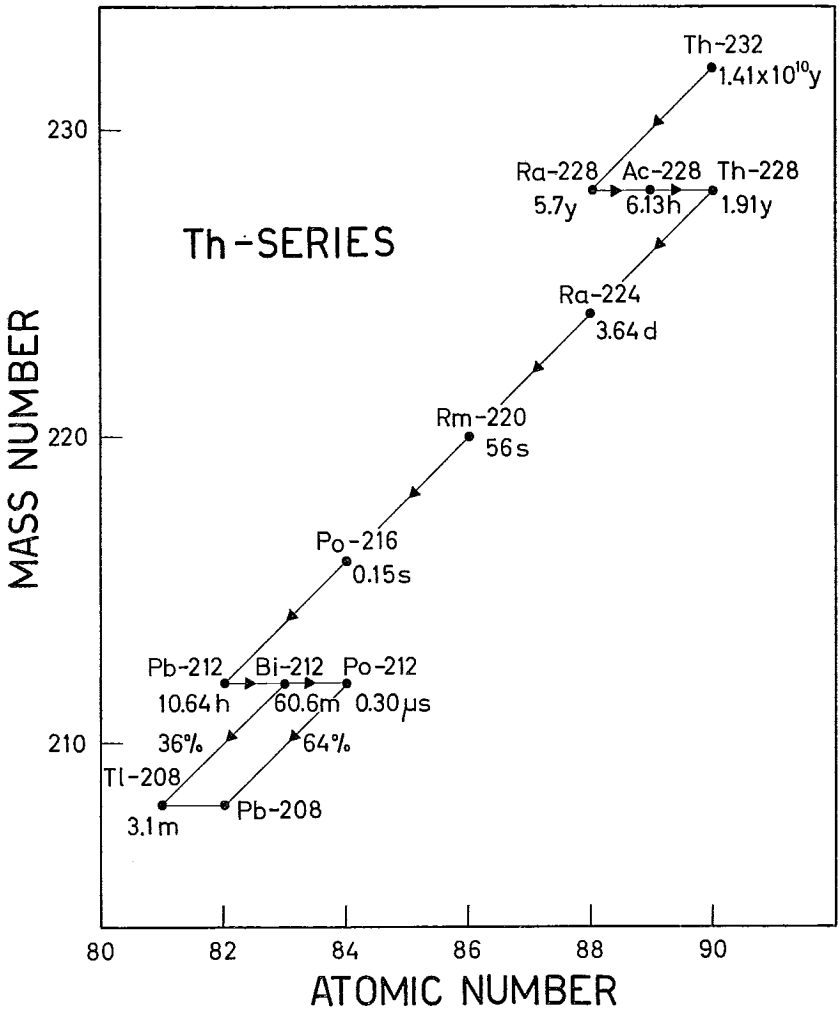

Fig. 1. The main properties of the Th-series, starting from the long-lived ^{232}Th-isotope and ending up with the stable end-product ^{208}Pb.

isotopes suitable for gamma-ray spectroscopy are ^{228}Ac (emitting the 1588 keV gamma-ray) and ^{208}Tl (emitting the 585 keV and 2614 keV gamma-rays); in the U-series the ^{214}Bi-isotope is the most suitable (emitting the 610 keV, 1760 keV and 2200 keV gamma-rays).

In the previous measurements [2] of the gamma-ray spectra obtained in the decay of the isotopes in the Th- and U-series, use has been mainly

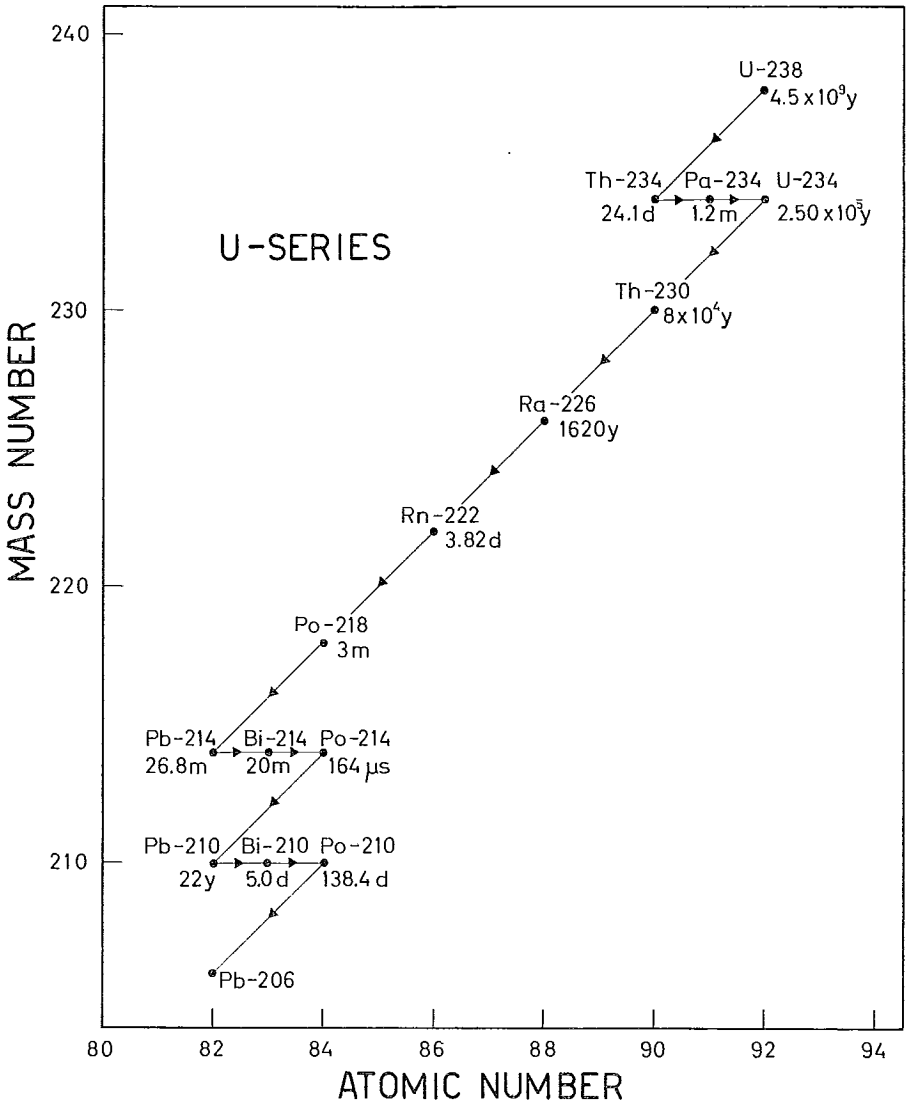

Fig. 2. The main properties of the U-series starting from the long-lived ^{238}U-isotope and ending up with the stable end-product ^{206}Pb.

made of NaI-scintillation detectors. The analysis of these spectra is difficult because of their complexity. With the advent of the Ge(Li)-detectors, however, it has become possible to distinguish even between close-lying lines in the gamma-ray spectra. Several authors [1, 3—7, 9—10] have recently used Ge(Li)-detectors to determine accurate gamma-ray energies and intensities in the decay of the Th- and U-series.

In the present report we demonstrate a method for determining the relative intensities of the gamma-rays from the ^{228}Ac, ^{208}Tl and ^{214}Bi isotopes by using Ge(Li)-spectroscopy. With the aid of the strongest gamma-ray pairs from the Th- and U-series, the ratio between the Th- and U-abundances is determined.

The absolute Th-abundance is obtained by measuring the intensity of the 2614 keV gamma-ray following the decay of ^{208}Tl.

Experimental technique

The Th—U-samples were made by grinding the rocks and putting the powder into thin-walled plastic cylinders. The dimensions of the cylinders were either 3 cm in diameter and 5 cm in height or 7 cm in diameter and 7 cm in height.

The samples were placed in the measuring position at a distance of 1 cm from the front face of the detector system. The detectors were a 2″ × 3″ NaI(Tl)-detector and a 50 cc Ge(Li)-detector, both well shielded by lead to diminish the background radiation. The gamma-ray spectra were recorded on a 400-channel pulse height analyzer, and each sample was measured for 24 hours.

Results

In Fig. 3 we show one of the Ge(Li)-gamma-ray spectra obtained from a natural Th—U-sample. Inserted is also a background spectrum. The most suitable gamma-ray pairs for determining the Th/U-ratio are the following: 585 keV, 610 keV; 1588 keV, 1760 keV and 2614 keV, 2200 keV. The relative intensities of these gamma-rays have been measured earlier [1, 3—7, 9—10], and a compilation of these measurements is given in Table 1, together with the value adopted in this work.

A relative efficiency curve for detecting gamma radiation is constructed for the Ge(Li)-spectrometer by plotting the $I_{meas.}/I_{aver.}$ as a

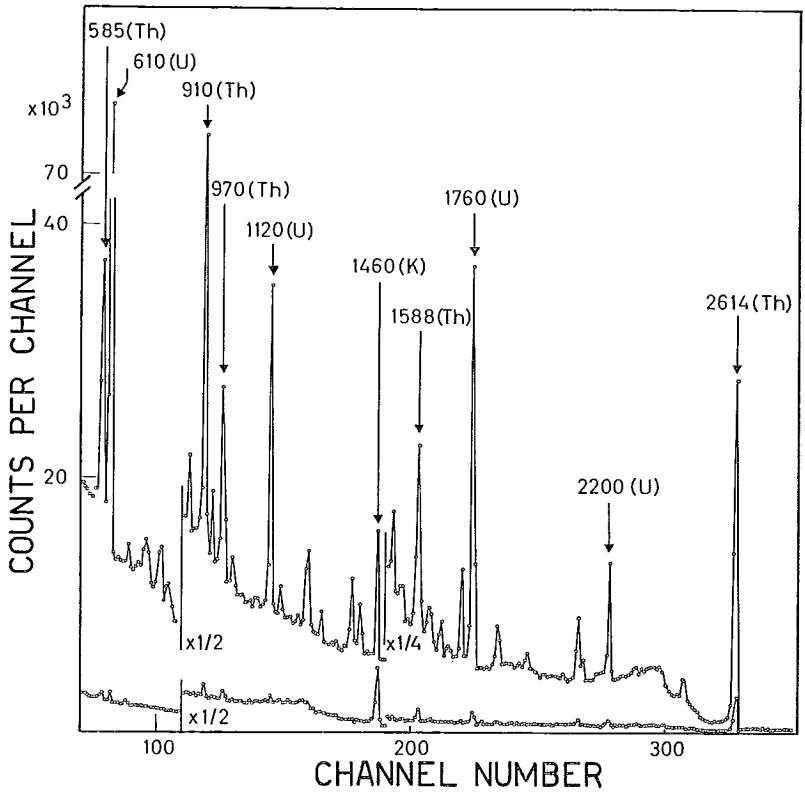

Fig. 3. Gamma-ray spectrum from a natural Th—U-sample registered with the Ge(Li)-detector. Indicated are the prominent gamma-rays from the decays: Th-thorium, U-uranium, K-potassium. The measuring time was 24^h. A background spectrum is also shown.

Table 1. The energies of the prominent gamma-rays in the decay of the Th-series and the U-series are listed together with the corresponding average intensity values ($I_{aver.}$) and the measured relative intensities (I_{meas}).

Th-series			U-series		
E (keV)	$I_{aver.}$	$I_{meas.}$	E (keV)	$I_{aver.}$	$I_{meas.}$
2614	120	32.5	2200	12.1	3.8
1588	40	16.9	1760	36.3	14.5
970	60	48.7	1120	34.1	21.2
910	83	66.2			
585	100	100.0	610	100.0	100.0

function of the gamma-ray energy. Here $I_{meas.}$ is the intensity of the gamma-ray line in the spectrum and $I_{aver.}$ is the corresponding adopted intensity value, as given in Table 1. The relative efficiency for the 585 keV and 610 keV gamma-rays is set as equal to unity. The relative efficiency curve is given in Fig. 4.

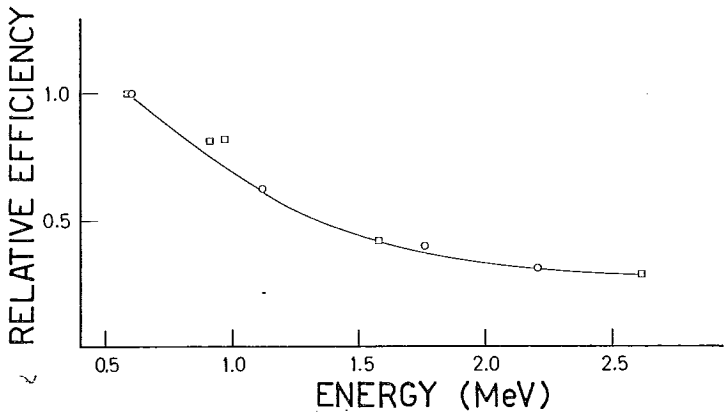

Fig. 4. The relative efficiency curve for the Ge(Li)-spectrometer is displayed. The relative efficiency values are normalized as a unity at the energy 0.6 MeV.

The Th/U-ratio is calculated with the aid of the gamma-ray pairs mentioned above. The net peak areas obtained from the spectra are corrected for the half-lives and atomic masses of the ^{232}Th- and ^{238}U-isotopes, decay branches and relative detecting efficiences. The Th/U-ratio for six samples is given in Table 2. Included are also the results obtained by the X-ray fluorescense and activation methods.

In Fig. 5 we display a spectrum obtained with the NaI(Tl)-detector. Inserted is also a background spectrum. It is obvious that, apart from

Table 2. The Th/U-ratios obtained by gamma-ray spectroscopy methods as compared with the results obtained by other methods.

Sample	1	2	3	4	5	6
$(Th/U)_{present}$	27.5	163	1.97	5.1	1.20	0.31
X-ray-fluor. meth.	—	—	2.65	—	1.15	0.34
Activ. meth.	—	—	2.80	9.4	1.23	0.48

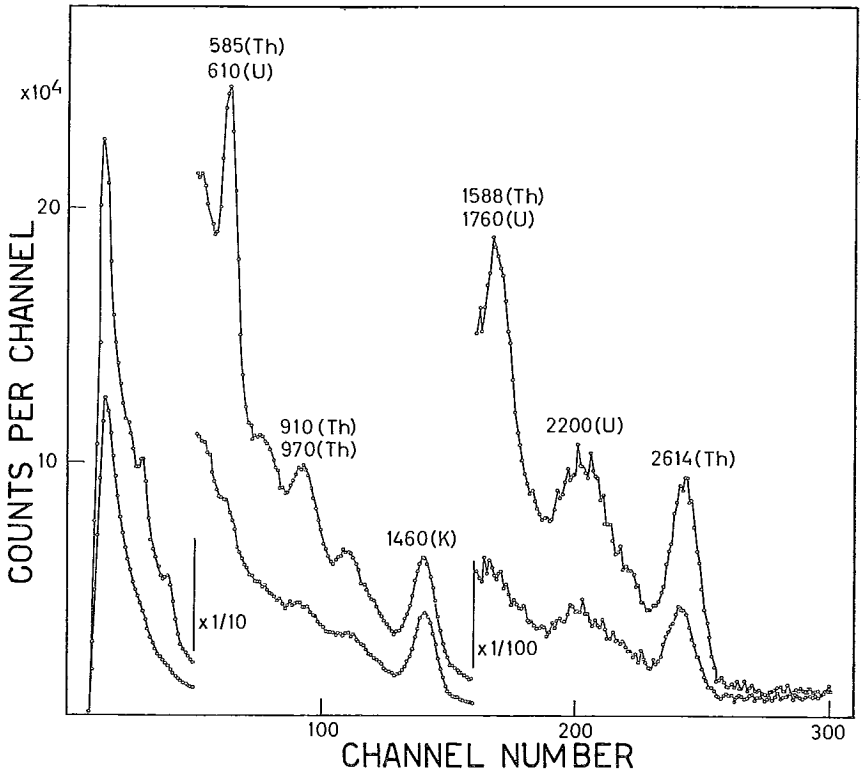

Fig. 5. Gamma-ray spectrum from a natural Th—U-sample registered with the NaI(Tl)-detector. The prominent gamma-rays from the decays are indicated and a corresponding background spectrum is given.

the 2614 keV gamma-ray peak, net peak areas are difficult to determine. Therefore only the absolute Th-abundances can be obtained from NaI-spectra with the aid of the 2614 keV gamma-ray. The Th-abundance in the sample is then obtained from the equation

$$\text{Th(ppm)} = \frac{T_{1/2}}{0.693} \cdot \frac{A_{\text{Th}}}{N_0} \cdot \frac{w}{m} \cdot \frac{1}{\varepsilon} \cdot \frac{dN}{dt} \cdot 10^{+6} \qquad (1)$$

Here

$T_{1/2}$ = half-line of ^{232}Th
A_{Th} = atomic mass of Th
N_0 = Avogadro's number

w = branching ratio of the decay of the ^{212}Bi isotope
m = mass of the sample
ε = photopeak efficiency value for the NaI-detector
dN/dt = counting rate

For the calculation, $T_{1/2}$, A_{Th}, N_0 and w are obtained from isotope tables [5], ε can be calculated [8], and m and dN/dt are measured. We also observe that for a given geometry

$$\frac{T_{1/2}}{0.693} \cdot \frac{A_{\mathrm{Th}}}{N_0} \cdot w \cdot \frac{1}{\varepsilon} \cdot 10^{+6} = \text{constant} \tag{2}$$

The value of the constant can be experimentally obtained by measuring the term $\frac{1}{m} \cdot dN/dt$ for samples with known Th(ppm)-abundances. This method is demonstrated in Fig. 6, where the measured results for $\frac{1}{m} \cdot dN/dt$ are plotted against the known Th(ppm)-abundances. The result is a straight line through the origo. The Th(ppm)-abundances for unknown samples are then obtained graphically from Fig. 6.

The advantage of the graphic method is that the influence of minor

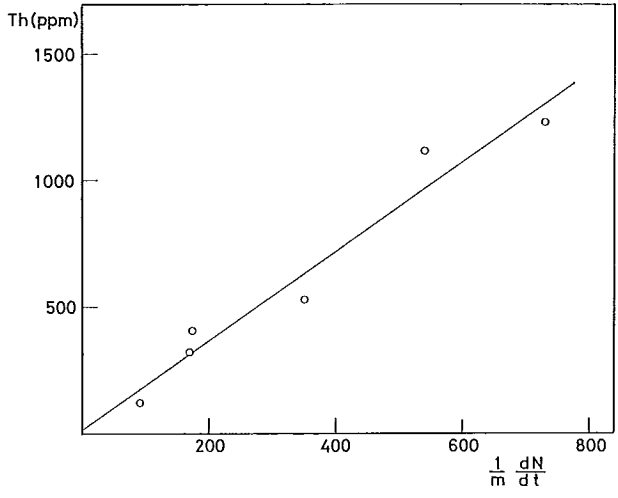

Fig. 6. Measured $m\frac{dN}{dt}$-values plotted against the known Th(ppm)-abundances. A least-square fit of a straight line to the points is also given.

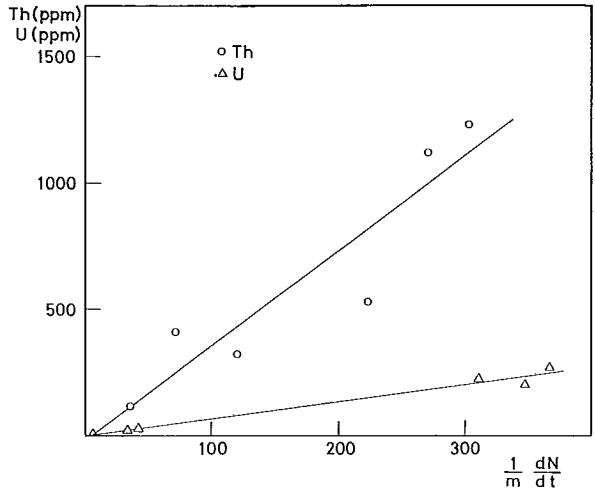

Fig. 7. Measured $m\frac{dN}{dt}$-values (Ge(Li)) plotted against known Th(ppm)- and U(ppm)-abundances. Least-square fits of straight lines to the points are also given.

unhomogenities in the powder samples is diminished and the effect of multiple scattering of gamma-rays in the detector system is eliminated.

In Fig. 7 we plot the Th(ppm)- and U(ppm)-values against the measured values of $\frac{1}{m} \cdot dN/dt$ for the Ge(Li)-spectrometer. Use is made of the 585 keV and 610 keV gamma-rays. This graphic representation can similarly be used for determining abundances.

The present method for determining the Th/U-ratio and the Th- and U-abundances is somewhat slow as compared with some other existing methods (X-ray fluorescence, activation analysis), because it demands long runs for registering the gamma-ray spectra. It is, however, sensitive to even small amounts of Th and U, and may prove useful in tracing these elements.

Also, the apparatus is compact and easily transportable into different places.

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