

in the absence or in the presence of added cytochrome C, as measured in the Warburg apparatus (1).

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Determination of Radioactive Content of Rocks by Means of Geiger-Müller Counters

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Geiger-Müller counters are widely used in the search for radioactive substances, but their use for immediate quantitative determination has not been elaborated. During a survey of the Velence Mountains in Hungary we have developed a direct approximating method.

Brass G-M counters of the self-quenching type (45 mm in diameter, 88 mm in length and 1.8 mm in wall thickness), filled with 100 mm argon and 10 mm alcohol vapor, were used in connection with all battery amplifiers. The soft components of the γ -radiation were absorbed by a 2 mm-lead shield. In this way only the known penetrating γ -components of the known radioactive bodies (uranium, thorium, their decay products, and potassium) could reach the counter.

In most cases the counter was placed touching the rock wall to be investigated, but in one instance, we drove a bore, 5 cm in diameter and 50 cm deep, into the granite wall and placed the counter within. The counting rate (number of impulses/min) was determined and compared with the counting rate of the cosmic radiation, the last amounting to 40/min. We subtracted this value from the observed total counting rate, and divided the remaining rock-activity counting rate by this factor. We observed that the counting rate was about 6 times higher in the bore and 2-3 times higher immediately at the wall than the cosmic radiation. The activity of the rocks can be expressed approximately by this factor, and this kind of expression is, within some limits, independent of the counter dimensions. It can be used for comparison of measurements made by various brass counters. However, it is not independent of the material of the counter, because the γ -sensitivity of a G-M counter depends upon the atomic weight of its substance. As is known, the intensity of the cosmic radiation depends upon the geographical latitude. In this case it amounted to about 1 impulse/min $\cdot$ cm² of maximum square area of the counter (length $\times$ diameter).

The observed γ -radiation of rocks may originate from K, U, and Th, and their decay products in equilibrium.

ThC'', RaB + C and K are the sources of the hard γ -radiations, which affect the counter under such conditions. Softer components may have a minor part only, for they are absorbed by the lead shield, and the sensitivity of a brass counter is small for soft γ -rays.

Now it must be realized, that the direct determination of the proportion of the existing radioactive substances to each other in the rock is hardly possible with a G-M counter without any chemical separation. It is possible, however, to determine the total radioactivity of the rocks expressing it in Th- γ -equivalents, as measured by a brass G-M counter behind a 2-mm lead shield. We achieved it in the following way: It is possible to calculate the total amount of the hard γ -radiation that reaches the counter from the surrounding rock substance. In Fig. 1 we have

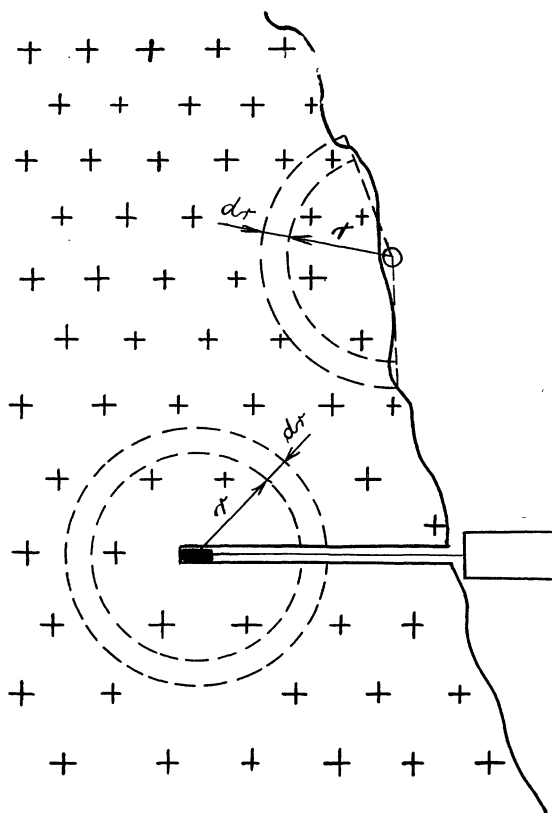


FIG. 1. Determination of Th and U concentration in rocks by a G-M counter tube.

taken the center of our coordinate system as the middle of the counter in the bore. (The counter may be regarded as small in comparison with the surrounding rock masses.) A small volume, dv , of the rock containing c g-Th-equivalents of radioactive substance/cm³ rock, will send a penetrating γ -radiation from r distance to the counter, giving the counting rate, dJ . It is obvious that $dJ = cAdv/r^2$, where A is the sensitivity of the given counter, that is, the rate of counting when 1 g Th (in radioactive equilibrium) is placed 1 cm from the counter.

Now we must take into account the self-absorption of the γ -radiation within the rock substance itself. The

γ -quanta traverse the distance r in rock and not in space, thus the counting rate is diminished by a factor of $e^{-\mu r}$, where μ is the absorption coefficient, expressed in cm^{-1} . For simplicity of calculation of the volume element, dv , we choose the spherical shell of radius r ; the thickness of it being dr : $dv = 4\pi r^2 dr$. Taking the self-absorption into account, the counting rate is

$$dJ = \frac{cA4\pi r^2 e^{-\mu r} dr}{r^2} = 4\pi cAe^{-\mu r} dr.$$

We take the integral over the infinite space filled with rock and obtain the total counting rate:

$$J = \frac{4\pi cA}{\mu}.$$

We are actually interested in the concentration c , which is expressed here in g/cm^3 . It is more usual to express it c' in g/g of rock, if ρ is the specific density of the rock in g/cm^3 .

$$c' = \frac{c}{\rho} = \frac{\mu J}{4\pi A \rho}.$$

The absorption coefficient above is expressed in cm^{-1} and refers to the rock substance. It can be determined experimentally by using a known amount (about 10–50 g) of old Th-compound, or we can calculate it reliably, knowing its value in lead, $\mu_{\text{Pb}} = 0.46 \text{ cm}^{-1}$ (with 2 mm of lead). By dividing it by the specific density of lead we obtain the so-called mass-absorption coefficient; multiplying this by the density of the rock, we obtain the absorption coefficient in the rock

$$\mu = \frac{\mu_{\text{Pb}}}{11.3} \rho$$

If the absorption coefficient in lead is substituted, the formula takes a simpler form, in which the specific density of the rock does not play any role:

$$c' = \frac{\mu_{\text{Pb}} J}{4\pi 11.3 A}$$

The counter sensitivity, A , was determined experimentally by placing 10 g of old ThO_2 at 30 cm distance from the center of the counter. For our brass counter its value was $4.8 \cdot 10^3$ impulses/g of Th-min.

We calibrated the same counter in U-equivalents as well, using instead of U, about 0.01 mc of radium in radioactive equilibrium. As the penetrating γ -radiation originates from RaB and RaC , the γ -radiations of the decay products between U and Ra can be neglected (1 g of Ra is equivalent to $3 \cdot 10^6$ g of U in radioactive equilibrium). The value for μ_{Pb} is known to be 0.53 cm^{-1} in lead. We obtained a value for A of $9.3 \cdot 10^3$ impulses/min · g of U at 1 cm distance from the given counter.

As can be seen, the estimation gave for the Th-U equivalent a value of 1.9, that is, 1.9 g of Th gives, in this brass counter, the same rate of counting as 1 g U (in radioactive equilibrium with their decay products).

The method outlined above was checked experimentally by our measurements on a granite wall near the village of Velence in Hungary. We obtained, with the counter in the bore, $12 \cdot 10^{-5}$ g-Th-equivalents/g of granite for the total concentration of the radioactive bodies. Careful laboratory tests were carried out later on a sample taken

from the same granite wall. It was dissolved completely and the equilibrium of radon and thoron content determined separately, by means of an ionization emanometer. These measurements yielded $4.2 \cdot 10^{-5}$ g of Th and $0.87 \cdot 10^{-5}$ g of U/g of granite. The granite of these mountains contains about 3.9% K as determined by A. Vendl (5). The K-U equivalent was determined by F. Behounek (1) and more recently by E. Gleditsch and T. Graf (2). The γ -radiation of K is equivalent to $4 \cdot 10^{-4}$ g of U/g of K. Expressing the U and K content of the granite in Th-equivalents, we obtain good agreement of the two entirely different methods of determination. The difference lies in the fact that the counter gives a mean value for the concentration within a sphere of a diameter of about 60 cm surrounding the bore, but the emanometric measurements indicate the activity of the dissolved sample to be about 4 g of granite.

Now in a practical search for radioactive content, higher concentrations must be considered. The K content can be neglected. Any commercial G-M counter tube can be calibrated in the way outlined above by the use of about 10 g of old Th compound and/or about 0.01 mc, calibrated Ra source in equilibrium. With a brass counter, not differing too much from the dimensions above, calibration is not essential. One may use our calibration, expressed by the approximate numerical formula:

$$c = \frac{0.46 \cdot 36.4 J}{4\pi 11.3 \cdot 10^3 \cdot 11.3} = 2.4 \cdot 10^{-5} \frac{J}{LD}$$

where c is the concentration in g-Th-equivalents/g of rock; J , the counting rate (in min); L the length; and D the diameter (in cm) of the brass cathode of the counter tube. Similarly, we obtain the concentration in g-U-equivalents/g of rock by the approximate numerical formula:

$$c = 1.4 \cdot 10^{-5} \frac{J}{LD}$$

If the measurements are made without use of the bore, the counter is put as near as possible to the wall, and there is a loss of 50% or more in the counting rate, depending upon the geometry. The accuracy is reduced by the fact that the geometry is not so well defined as in the case of the bore, but the formula can be used, approximately, by multiplying the counting rate by a correction factor of about 2–2.2.

Further, the use of the following roughly approximating simple rule may be suggested for uranium explorers when extended territories must be surveyed within a short time: If a G-M counter of any commercial dimensions, shielded by 2 mm of lead, is placed against the rock wall and the observed counting rate (minus cosmic radiation) is x times the rate of cosmic radiation for the same counter, then the average U concentration in the rock is x times 25 g U/metric ton (1,000 kg) of rock.

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