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Geological Age Determination by Lead : Alpha-ray Method

I. Determination of Alpha-ray Activity of Zircon

By

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Abstract

Determination of age of granite by lead : alpha-ray method involves (1) preparation of samples, (2) determinations of lead and (3) those of alpha-ray activity. This paper includes the third procedure followed by the writers in determining the ages of the zircons separated from some granites using a 4π gas-flow counter. The formula for calculating alpha-activity of the sample from the observed counting rate is newly deduced taking into consideration of the absorption by the foil which serves as sample holder as well as a cover preventing the jumping off of the pulverized sample. The results of some check examinations are also given.

Introduction

Geochronology is a problem recently much interested by most geologists in Japan and some of them are planning or already getting into the study on this subject. Usually argon-potassium method is adopted which is presumably one of the best methods now available. The present writers, however, have been engaged in the investigation of the lead : alpha-ray method of age determination, which was devised by LARSEN, E. S. Jr. *et al.*[1] for the first time in 1952 and followed by a few other authors in various countries. Though the fundamental principle remains the same, the apparatus and procedure adopted by the writers are different from those used by them, it will be adequate to describe them somewhat in detail as well as the deduction of the formula fit for the present apparatus.

General principle

During the decay of uranium-238 to the final daughter element lead-206, eight alpha-particles are emitted. The similar relation may be observed with thorium-232 and uranium-235, and these relationships may be expressed as

$$U^{238} \longrightarrow Pb^{206} + 8\alpha + 6\beta$$
$$U^{235} \longrightarrow Pb^{207} + 7\alpha + 4\beta$$
$$U^{232} \longrightarrow Pb^{208} + 6\alpha + 4\beta .$$

If the member elements of each series are in radioactive equilibrium, the number of alpha-particles produced per year per gram of mineral is $8\lambda_U Z_U + 7\lambda_{AcU} Z_{AcU} + 6\lambda_{Th} Z_{Th}$, Z being the number of atoms existing and λ decay constant (yr^{-1}) of the parent element of the respective series. As uranium-238 and uranium-235 are chemically isotopic and the relative abundances of Z_U and Z_{AcU} are found nearly constant, the ratio Z_{AcU}/Z_U being very small (0.007), the effect of actinium series is usually treated as including in that of the uranium series. On the other hand, so far as the radioactive equilibrium is maintained, the production of radiogenetic lead corresponds to the decay of the parent elements uranium-238 (+ uranium-235) and thorium-232. In very old mineral, however, decrease of the parent element must be taken into consideration.

Assuming that no common lead was present initially in the mineral, the lead found by analysis may be solely attributable to the disintegration products, and if divided by the rate of disintegration of the parent elements deduced from the counting of alpha-rays, it will give the age of the mineral. Thus the general formula giving the age of the mineral t is:

$$t = C \cdot \frac{Pb}{\alpha} \quad \text{in million years} \qquad (1)$$

where Pb is the amount of lead determined by spectro-chemical or any other method usually expressed in 10^{-6} gram per gram of sample and α is the number of alpha-particles emitted in one milligram of the sample per hour. As for the constant C, slightly different values are assigned by various authors. For example, QUINN, A. W. (1957)[2] gave 2632 for the mineral containg uranium (+actinium) family alone, 2013 for one containing exclusively thorium family of the radioactive series elements and 2485 for zircon of which Th/U ratio is taken as 1. Assuming the ratio to be 0.46, WEBBER, G. R., *et al.* (1956) also gave $C=2580$ for zircon.

Deduction of number of alpha-ray emission in a sample from the observed counts in unit time.

An alpha-particle is emitted from the nucleus of the alpha-rayer without fail as it disintegrates into the next daughter element, so that alpha-particle emission is equal to the disintegration of alpha-rayers in number. Of the emitted alpha-particles only a portion can run out of the surface of the sample, leaving the remainder absorbed in the body of the sample itself. All of the alpha-particles thus escaped out of the sample in turn, cannot be caught by a counter as pulse counts, owing to the following factors: (i) geometrical relation between

alpha-ray source and receiver (ionization chamber in the present case), (ii) sensitivity of the counter, (iii) counting loss due to recovery time of the counter, (iv) absorbers intervening between source and effective space of the ionization chamber. The last means layer of air or gas and metal foil or metallized thin film which is used as a cover for protection against jumping up of powder of the sample and also as a holder of the sample in case of 4π counter. For the sake of simplicity, let us confine the problem in the case of 4π counter from now.

The pulverized sample to be tested is sandwiched as a 'thick source' with metallized film (Myler) or aluminium foil, and put on the center of the equatorial plane of the spherical ionization chamber. By thick source is meant any thickness exceeding the range of the most energetic alpha-ray. At the poles of the upper and lower hemisphere of it are provided with loop-shaped collecting electrodes (cf. Fig. 1).

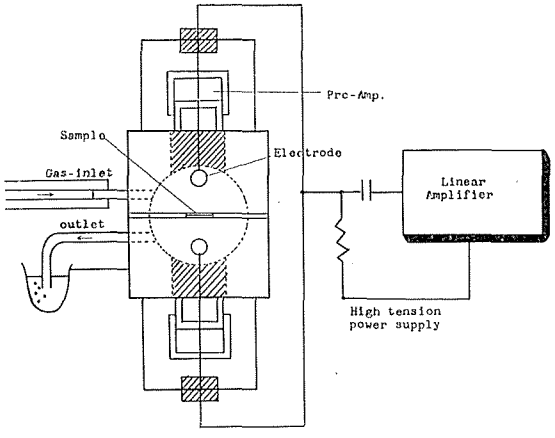

Fig. 1. Schematic diagram of 4π gas-flow counter.

Referring to Fig. 2, if SP is the path of an alpha-ray emitted from radioactive source S $(dx\,dy\,dz)$ within the sample covered with aluminium foil, leaving a residual range BP which is just equal to the minimum detectable range ρ for the counter, then the alpha-rays with residual ranges equal to or greater than ρ enter the detecting chamber within a cone of a half-angle θ, θ being the angle between SP and the direction SA normal to the surface. The proportion of such rays to the total is $(1-\cos\theta)/2$. For the case of 4π counter, however, both up and down sides must be taken into consideration, so the proportion become to be $(1-\cos\theta)$, the edge effect being neglected. Let R be the range (cm) of alpha-ray in air; μ, μ' the ratios of ranges in source and covering foil to that in air respectively; y the depth of the source (cm); a' the thickness of the foil (cm), and a the same expressed in terms of range of

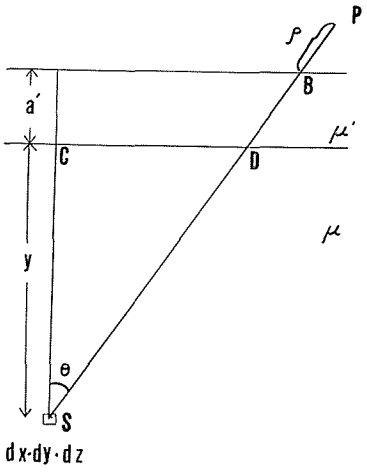

Fig. 2.

alpha-ray in air; $a=a'/\mu'$, then the number of counts per cm^2 of the surface of the source per unit time is:

$$n' = N(1-\cos\theta)dxdydz$$

where N is equal to the number of alpha-rays per cm^3 of the source per unit time, and the total number of the counts n is obtained by integrating n' between the limits; $y=0$ and $y=\mu(R-\rho-a)$. Assuming $dx\cdot dz=1$, we have

$$\begin{aligned} n &= N\int_0^{\mu(R-\rho-a)} (1-\cos\theta)\cdot dy \\ &= N\int_0^{\mu(R-\rho-a)} \frac{(R-\rho-a)-y}{(R-a)}\cdot dy \\ &= \frac{1}{2}\mu N\frac{(R-\rho-a)^2}{R-\rho} \end{aligned} \qquad (3)$$

Eq. (3) was deduced under the assumption that the radioactive elements whose alpha-ray having definite energy were uniformly distributed in the sample. But this is not the case in natural sample. Therefore, the term including R in Eq. (3) is to be calculated for each alpha-rayers in the radioactive series. As a development of Eq. (3), we have

$$n_{\mathrm{UI}} = \frac{\mu}{2} N_{\mathrm{UI}} \Sigma_{\mathrm{UI}} \frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho} \qquad \text{for uranium series,}$$

$$n_{\mathrm{AcU}} = \frac{\mu}{2} N_{\mathrm{AcU}} \Sigma_{\mathrm{AcU}} \frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho} \qquad \text{for actinium series,}$$

$$n_{\mathrm{Th}} = \frac{\mu}{2} N_{\mathrm{Th}} \Sigma_{\mathrm{Th}} \frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho} \qquad \text{for thorium series,}$$

where n_{UI} etc.: numbers of counts per cm^2 in unit time due to uranium series etc.; N_{UI} etc.: numbers of disintegration of the parent element UI per cm^3 in unit time, etc.; and R_{UI} etc.: ranges of alpha-rays from each members of uranium series etc. The summation is made for each alpha-rays from each series, respectively.

The total number of alpha-rays to be counted is therefore,

$$\begin{aligned} n &= n_{\mathrm{UI}}+n_{\mathrm{AcU}}+n_{\mathrm{Th}} \\ &= \frac{\mu}{2}\left\{N_{\mathrm{UI}}\Sigma_{\mathrm{UI}}\frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho}+N_{\mathrm{AcU}}\Sigma_{\mathrm{AcU}}\frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho}+N_{\mathrm{Th}}\Sigma_{\mathrm{Th}}\frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho}\right\} \end{aligned} \qquad (4)$$

Since $N_{\mathrm{AcU}}/N_{\mathrm{UI}}=0.045$ as calculated below,

$$n = \frac{\mu}{2}\left[N_{UI}\left\{\Sigma_{UI}\frac{(R_{UI}-\rho-a)^2}{R_{UI}-\rho}+0.045\Sigma_{AcU}\frac{(R_{AcU}-\rho-a)^2}{R_{AcU}-\rho}\right\}+N_{Th}\Sigma_{Th}\frac{(R_{Th}-\rho-a)^2}{R_{Th}-\rho}\right] \quad (5)$$

Determination of ρ and "a"

For the determination of ρ and "a", alpha-ray source of a constant range such as polonium source is preferable. But as such one was not available, chemically prepared uranium oxide was used as a thick source. Successive countings were made by increasing the number of sheet (m) of the absorber foil. In this case, in the assumption that radioactive equilibrium is nearly attained between uranium I and uranium II and $\mu'/\mu \fallingdotseq 1$, μ' being the ratio of alpha-ray from uranium II in the sample and that in air, the counts to be obtained are

$$n_m = \mu\frac{N}{2}\left\{\frac{(R_{UI}-\rho-a)^2}{R_{UI}-\rho}+\frac{(R_{UII}-\rho-a)^2}{R_{UII}-\rho}\right\}$$

where $m=0,\ 1,\ 2,\ 3,$ etc.

Hence,

$$\frac{n_m}{n_0} = \frac{\dfrac{(R_{UI}-\rho-ma)^2}{R_{UI}-\rho}+\dfrac{(R_{UII}-\rho-ma)^2}{R_{UII}-\rho}}{(R_{UI}-\rho)+(R_{UII}-\rho)} \quad (6)$$

The results obtained for n_m/n_0 are shown in Table 1, in which aluminium foil (A) (shown in Table 2) was used.

Table 1. Counting rate *vs* number of sheet of Al-foil

No. of sheets of Al-foil (A)	$m=0$	$m=1$	$m=2$	$m=3$
Counts/min.	3950.83 ±19.16	1650.05 ±10.18	348.08 ±2.57	9.28 ±0.13
n_m/n_0	1 ±0.000485	0.41765 ±0.00029	0.0881 ±0.00065	0.00235 ±0.00003

With the four observation equations for different values of n_m/n_0, the most probable values of ρ and a were determined by means of the method of least squares. The similar processes were taken for other two Al-foils and one myler film, and the results are summarized in Table 2.

Table 2

Minimum detectable range : ρ (cm)	Thickness of abosrbers in terms of alpha-ray range in air : a (cm)			
	Myler	Al-foil (A)	Al-foil (B)	Al-foil (C)
0.20	0.84	0.97	0.50	0.34

The summations in Eq. (5) were calculated as shown in Table 3 by using the values thus obtained for ρ and a, in which values of ranges of alpha-rays from each alpha-emitter summarized by YAGODA, H. (1949)[3] were adopted, and also members of very small branching ratio were neglected.

Table 3

	Al-foil (A) $a=0.97$ cm	Al-foil (B) $a=0.50$ cm
$\Sigma_{UI}\frac{(R_{UI}-\rho-a)^2}{R_{UI}-\rho}$	16.75	22.67
$\Sigma_{AcU}\frac{(R_{AcU}-\rho-a)^2}{R_{AcU}-\rho}$	19.75	24.67
$\Sigma_{Th}\frac{(R_{Th}-\rho-a)^2}{R_{Th}-\rho}$	17.38	18.82

Putting these values in Eq. (5), it was reduced to the following simple form:

$$\left.\begin{aligned} n &= \frac{\mu}{2}(17.62N_{UI}+17.38N_{Th}) \quad \text{for Al-foil (A)} \\ n &= \frac{\mu}{2}(23.78N_{UI}+22.00N_{Th}) \quad \text{for Al-foil (B)} \end{aligned}\right\} \tag{7}$$

If the mineral whose age is to be determined lacks either uranium or thorium, Eq. (7) is again simplified as $n=(\mu/2)\cdot 17.38N_{Th}$ and $(\mu/2)\cdot 17.62N_{UI}$. Usually for a mineral such as zircon, the relative abundance may be assumed constant, say K.

Now, let $\bar{N}_{UI}$, $\bar{N}_{AcU}$, $\bar{N}_{Th}$ be the numbers of alpha-rayers disintegrating in 1 mg of sample per hour, then we have

$$N_{UI}=\frac{1}{8}\cdot d\cdot\bar{N}_{UI}\cdot 10^3;\quad N_{AcU}=\frac{1}{7}\cdot d\cdot\bar{N}_{AcU}\cdot 10^3;\quad N_{Th}=\frac{1}{6}\cdot d\cdot\bar{N}_{Th}\cdot 10^3,$$

d being the density of the sample.

On the other hand,

$$\frac{N_{AcU}}{N_{UI}}=\frac{6.06\cdot 10^{23}\cdot\lambda_{AcU}\cdot AcU/W_{AcU}\cdot d}{6.06\cdot 10^{23}\cdot\lambda_{UI}\cdot UI/W_{UI}\cdot d}\fallingdotseq 0.045$$

and

$$\frac{N_{Th}}{N_{UI}}=\frac{6.06\cdot 10^{23}\cdot\lambda_{Th}\cdot Th/W_{Th}\cdot d}{6.06\cdot 10^{23}\cdot\lambda_{UI}\cdot UI/W_{UI}\cdot d}=0.334\frac{Th}{UI}$$

where W is atomic weight; UI, AcU and Th are respectively content (in gram) of uranium, actinium and thorium.

Hence,

$$\frac{\bar{N}_{AcU}}{\bar{N}_{UI}}=\frac{N_{AcU}\cdot(7/d)}{N_{UI}\cdot(8/d)}=0.039$$

$$\frac{\bar{N}_{\mathrm{Th}}}{\bar{N}_{\mathrm{UI}}} = \frac{N_{\mathrm{Th}}\cdot(6/d)}{N_{\mathrm{UI}}\cdot(8/d)} = 0.25\,K$$

From Eq. (4),

$$n = \frac{\mu}{2}\cdot N_{\mathrm{UI}}\left\{\Sigma_{\mathrm{UI}}\frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho} + \frac{N_{\mathrm{AcU}}}{N_{\mathrm{UI}}}\Sigma_{\mathrm{AcU}}\frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho} + \frac{N_{\mathrm{Th}}}{N_{\mathrm{UI}}}\Sigma_{\mathrm{Th}}\frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho}\right\} \tag{9}$$

$$= \frac{\mu}{2}\cdot 10^3\cdot\frac{d}{8}\cdot N_{\mathrm{UI}}\,(16.75+0.045\times 19.34+0.334K\times 17.38)$$

So that,

$$\bar{N}_{\mathrm{UI}} = \frac{2.8n}{(17.62+5.80K)\cdot\mu\cdot d\cdot 10^3}$$

$$\bar{N}_{\mathrm{Th}} = \frac{2.8n\times 0.25K}{(17.62+5.80K)\cdot\mu\cdot d\cdot 10^3}$$

Similarly.

$$\bar{N}_{\mathrm{AcU}} = \frac{2.8n\times 0.039K}{(17.62+5.80K)\cdot\mu\cdot d\cdot 10^3}$$

Finally, we have as the total number of alpha-rays emitted per hour per milligram of sample,

$$\alpha = \bar{N}_{\mathrm{UI}}+\bar{N}_{\mathrm{AcU}}+\bar{N}_{\mathrm{Th}} = \frac{60\times 16n\,(1+0.039+0.25K)}{(17.62+5.80K)\cdot\mu\cdot d\cdot 10^3} \tag{10}$$

(n: cpm)

Ratio of ranges of alpha-ray in minerals to that in air: μ

The data of the observed ranges of alpha-particles in minerals are very scarce, and usually they are calculated from the ranges in air by the formula based upon the Bragg-Kleeman rule. If ψ is the permeability of the medium to alpha-particles defined in terms of its atomic composition, which is composed of atoms A, B, C, $\cdots$ with the relative proportions a, b, c, $\cdots$, then the permeability of the medium is

$$\psi = a_1\sqrt{W_A}+b_1\sqrt{W_B}+c_1\sqrt{W_C}+\cdots = \Sigma n_1\sqrt{W_N}$$

where W designates atomic weight.

Let the ranges of alpha-particle in air and in any other medium be R and R_s respectively, then

$$\mu = \frac{R_s}{R} = \frac{d_a\psi_s}{d\,\psi_a} \fallingdotseq \frac{0.001226\,\psi_s}{d\times 3.839} \quad \begin{pmatrix}\text{density of air: } d_a=0.001226,\\ \text{permeability of air: } \psi_a=3.839\end{pmatrix}$$

or

$$\mu d \fallingdotseq 3.194\cdot 10^{-4}\,\psi_s$$

Assuming the composition of zircon to be $ZrSiO_4$, we have $\psi=6.94$ and consequently

$$\mu d = 3.194\cdot10^{-4}\times6.94 = 22.17\cdot10^{-4}$$

Using this value, Eq. (10) becomes:

$$\alpha = \frac{(1.039+0.25K)\times9600}{(17.62+5.80K)\times22.17}n \tag{11}$$

Hence, by counting the number n of alpha-particles per minute per cm^2 of the surface of the thick source of zircon, of which $K=\frac{Th}{U}$ is known, we can calculate the total emission $\alpha\,(mg^{-1}hr^{-1})$.

Assuming $K=1$, finally we have

$$\alpha_{(\alpha's/mg\cdot hr)} = 23.81\,n_{(cpm)} \tag{12}$$

This relation folds under the conditions that the measurement is made by 4π gas-flow counter whose minimum detectable range ρ is 0.20 cm, the sample being sandwiched by foils of thickness 0.97 cm in air-equivalent of alpha-rays.

Some experiments proving the appropriety of the assumed values of ρ and "α"

(i) *Pulse-height analysis by means of discriminator*: Pulse-height analysis of alpha-rays were made with the ionium standard of which facility was due to

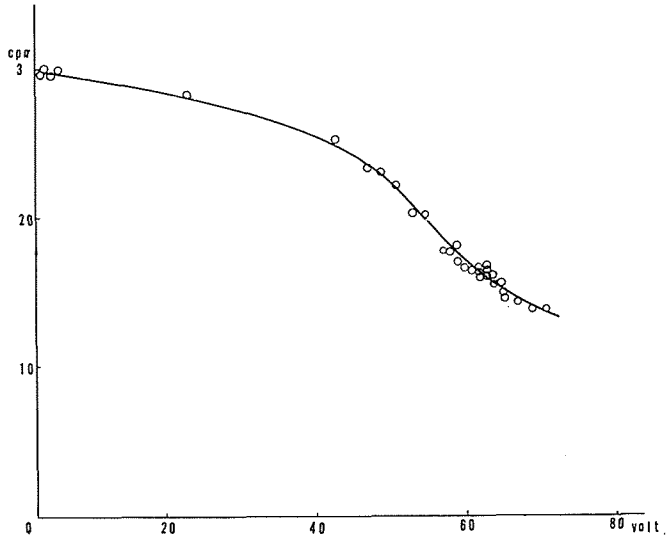

Fig. 3.

Dr. T. MURANO of the Japan Atomic Fuel Corp.. The standard is very thin ionium film deposited on a platinum plate in a circle of 1.5 cm in diameter. The result, counts *vs* input pulse-height in terms of volt, is shown in Fig. 3. That the alpha-rays from the interior show self-absorption, in spite of the film being very thin, can be perceived by sloping of the left half part of the curve. Besides, that of the right half part shows the suspectable existence of the longer range alpha-rayers. Anyhow, this standard is proved to be not so good as to be used as a mono-energy alpha-ray source.

(ii) *Decrease of counting rate with increase of number of sheet of absorber foil*: Using the same standard, the decrease of counting rate with increase of number of sheet of absorber Al-foil (A) was observed. The results were shown in Table 4. On the other hand, in the last column of it was shown the similar results theoretically obtained by the following relation under the assumption that the self-absorption is negligible as the ionium source is very thin and alpha-rays from it have monorange R_{I0}.

Table 4. Counting rates *vs* numbers of sheet of absorber foil for alpha-particles from Io source.

Nos. of sheet of Al-foil (A) m	Observed rate of count cpm	Back-ground cpm	Net count cpm	Observed n_m/n_0	Calculated n_m/n_0 neglecting self absorption
0	29.54	0.57	28.97	1	1
1	20.13	0.57	19.56	0.6751	0.6655
2	10.28	0.57	9.71	0.3351	0.3344
3	1.26	0.57	0.69	0.0248	0

Let N be the rate of alpha-ray emission per cm^3 of the sample per minute, then the count per minute obtained by a 2π counter is

$$n = \frac{1}{2}N(1-\cos\theta) \qquad (n: \text{ cpm/cm}^2) \tag{13}$$

where $\cos\theta = a/(R_{I0}-\rho)$.

Denoting the number of sheet of the Al-foil as m, and counting rate (cpm) without and with absorber of m sheets as n_0, n_m respectively, then we get

$$n_m/n_0 = (R_{I0}-\rho-ma)/(R_{I0}-\rho) \tag{14}$$

Taking the range of alpha-ray from ionium source R_{I0} be 3.10 cm, we have as $(n_m/n_0)_{m=0,1,2,3,\ldots}$, the figures shown in the rightmost column in Table 4.

Considering from the fact that the used ionium alpha-ray source was not a good one as above-mentioned, the figures shown in the last two columns reveals fairly good agreements. This fact implies that the assumed values for ρ and "a" are adequate.

Test of reliability of Eq. (4) with a sample whose uranium and thorium contents are known.

As the sample for this purpose, Kobeite was used. This mineral was firstly found by TAKUBO, J.[4] in pegmatite at Kôbe, Ohmiya-chô in Kyoto Prefecture, and revised chemical analysis was given by NAGASHIMA and MASUTOMI[5].

About 20 mg of the sample obtained as the result of selection by heavy liquid (methylene-iodide) was put on a sample pan and covered with aluminium foil (A). The measurement gave as the number of count per cm^2 per sec.: $n=11.754$. The density of the mineral is 4.6 and the value of μd calculated to be $19.60\cdot10^{-4}$ after the formula of YAGODA (*loc. cit.*), based on the data of chemical analysis by NAGASHIMA and MASUTOMI.

Using these values, the total count deduced by the similar process as when Eq. (9) and Eq. (10) were deduced from Eq. (5), is given by

$$n=\mu d\cdot10^3\left[\mathrm{UI}\left\{6.21\,\Sigma_{\mathrm{UI}}\frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho}+0.28\,\Sigma_{\mathrm{AcU}}\frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho}\right\}+2.06\mathrm{Th}\,\Sigma_{\mathrm{Th}}\frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho}\right]$$
$$=19.60\cdot10^{-1}(109.44\mathrm{U}+35.80\mathrm{Th}) \qquad (15)$$

in which n is expressed as counts/sec.$\cdot cm^2$; U, Th: contents in g/g (UI≒U), also $a=0.97$ cm and $\rho=0.20$ cm.

On the other hand, the ratio Th/U is 0.24 according to the chemical analysis above-mentioned. From these, we get

$$\mathrm{U}=0.0498\ \mathrm{g/g};\qquad \mathrm{Th}=0.0120\ \mathrm{g/g}$$

or

$$\mathrm{U_3O_8}=0.0556\ \mathrm{g/g};\quad \mathrm{ThO_2}=0.0135\ \mathrm{g/g}.$$

The chemical analysis of Kobeite by NAGASHIMA and MASUTOMI gives

$$\mathrm{U_3O_8}=0.0539\ \mathrm{g/g};\quad \mathrm{ThO_2}=0.0125\ \mathrm{g/g}.$$

The differences are only ca. 3%. It may be said from this result that Eq. (5) and deduction thereafter as well as assumed values for a and ρ, and counting itself were proper.

Further check by the standard zircon

Standard zircon (R-1932) for alpha-ray was furnished by the courtesy of Dr. STERN, T. of the U. S. Bureau of Standard through Dr. HAYASE of our institute. The nominal alpha-ray emission per milligram per hour is 200, and Th/U ratio is assumed to be 1. Our measurements were practised twice with the interval of several days, each being two hours counting. The effective area of the sample pan was 0.495 cm^2 and Al-foil (A) was used. The obtained counting rate was 3.78 ± 0.02 cpm which corresponds to 7.64 α's/$cm^2\cdot$min. By Eq. (12), we get: $\alpha_{(\alpha\text{'s/mg}\cdot\text{hr})}=181.8$. Thus the present counter gave the counts 10.0% less

than that nominated with the alpha-particle standard. Hence, 1.10 is used as an efficiency correction factor for the present counter.

Summary

(1) The general formula giving the age t is: $t=C(\mathrm{Pb}/\alpha)$ in which Pb denotes the amount of the radiogenetic lead in ppm of the sample and α the numbers of alpha-particles produced in one milligram of it in an hour and C a constant depending upon Th/U in the sample.

(2) The total number of alpha-rays to be counted per cm^2 per minute is:

$$n=\frac{\mu}{2}\left\{N_{\mathrm{UI}}\Sigma_{\mathrm{UI}}\frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho}+N_{\mathrm{AcU}}\Sigma_{\mathrm{AcU}}\frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho}+N_{\mathrm{Th}}\Sigma_{\mathrm{Th}}\frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho}\right\}$$

$$=\frac{\mu}{2}\left[N_{\mathrm{UI}}\left\{\Sigma_{\mathrm{UI}}\frac{(R_{\mathrm{UI}}-\rho-a)^2}{R_{\mathrm{UI}}-\rho}+0.045\,\Sigma_{\mathrm{AcU}}\frac{(R_{\mathrm{AcU}}-\rho-a)^2}{R_{\mathrm{AcU}}-\rho}\right\}+N_{\mathrm{Th}}\Sigma_{\mathrm{Th}}\frac{(R_{\mathrm{Th}}-\rho-a)^2}{R_{\mathrm{Th}}-\rho}\right]$$

where N_{UI} etc. are numbers of alpha-rays produced from parent element UI per cm^3 per min. etc.; R_{UI} etc. ranges of alpha-rays from each member of uranium series, etc.; ρ minimum detectable range and a range (cm) of alpha-ray in air corresponding to the thickness of the absorber foil assuming the slight differences of μ for alpha-rays of different energies, μ being ratio of range in mineral and that in air.

(3) The values of ρ and a were determined by means of the method of least squares from observation equations obtained for different values of n_m/n_0 where the suffixes designate the numbers of sheet of the foil used as an absorber.

(4) The total number of alpha-rays produced per hour per milligram of sample is;

$$\alpha=\frac{0.96(1.039+0.25K)}{(17.62+5.80K)\cdot\mu\cdot d}n \qquad \left(n:\ \mathrm{cpm};\ K=\frac{\mathrm{Th}}{\mathrm{UI}}\fallingdotseq\frac{\mathrm{Th}}{\mathrm{U}}\right)$$

(5) Assuming the composition of zircon to be $ZrSiO_4$ and its density 0.46, we have $\mu d=22.17\cdot10^{-4}$, and consequently

$$\alpha_{(\alpha\text{'s/mg}\cdot\text{hr})}=23.81\ n_{(\mathrm{cpm})}$$

for $\rho=0.20$ cm and $a=0.97$ cm.

(6) The observed values of n_m/n_0 show fair agreements with the calculated values, proving the assumed values for ρ and a to be adequate.

(7) On the sample (Kobeite) of which results of the chemical analysis were given, the obtained values for n was

$$n=19.60\cdot10^{-1}(109.44\mathrm{U}+35.80\mathrm{Th})$$

Using Th/U=0.24 as given by the chemical analysis, we have found by this

relation $U_3O_8=0.0556$ g/g ; $ThO_2=0.0135$ g/g. Comparing these with the data of the chemical analysis, $U_3O_8=0.0539$ g/g ; $ThO_2=0.0125$ g/g, we found the difference ca. 3%. This results may prove the correctness of the deduction and assumed values for a and ρ as well.

(8) The check observation by use of the standard zircon for alpha-ray from the U. S. Bureau of Standard gave the counts 10.0% less than the nominated value of standard. Hence 1.10 was adopted as an efficiency correction factor for the present counter.

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