

Age of Hiei Granite determined with Zircon and Lead-Alpha Method

By

Jirô NAGAI

Geological and Mineralogical Institute, University of Kyoto

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Résumé

Les principales opérations ont été effectuées pour déterminer l'âge de la granite par des mesures de la radioactivité et la teneur en plomb radiogénique des zircons. L'évaluation du rapport d l'uranium au thorium a été y considéré. Le granite Hiei serait âgé $111 \cdot 10^6$ ans.

Introduction

Revived from "chemical lead-uranium method", the "lead-alpha-ray method" to determine the age of rock, devised by LARSEN, E. S. JR.¹⁾ *et al.* has been followed in this country by HATUDA, Z.^{2), 3)} and by his co-workers since 1958. The method offers great promise because it is relatively brief in process and good in result.

In the process of measurement of alpha-activity of sample, the variation of the ratio of thorium to uranium contained, being different in each sample, produces some error in results, if it is neglected as done by LARSEN. The writer has determined the ratio in each sample of zircon by utilizing the difference in energy spectra of alpha-rays from thorium and uranium series.

In the lead-alpha-ray method the chief equipments are alpha-counter and spectrographic instrument which are now somewhat easily available in this country.

Fundamental relation of radiogenetic lead and its parent elements

If N_0 is the number of radioactive atoms initially existed in 1 gr of mineral at the time 0, and N the number at the time t , then

$$N = N_0 e^{-\lambda t}$$

where λ is equal to the disintegration constant of the said element. The num-

ber of atoms of the radiogenetic lead formed during the period t , is equal to that of the initial atoms disintegrated during the same time.

$$N_{\text{Pb}} = N_0 - N$$

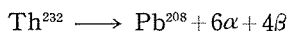
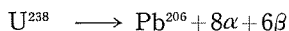
Therefore,

$$\frac{N_{\text{Pb}}}{N} = \frac{1 - e^{-\lambda t}}{e^{-\lambda t}} = e^{\lambda t} - 1 = \lambda t + \frac{\lambda^2 t^2}{2!} + \frac{\lambda^3 t^3}{3!} + \dots$$

If $\lambda t \ll 1$ (λ in yr^{-1} ; t in yr), i. e., t is less than 3×10^8 years, the second and its descendent terms can be negligible, then we have the approximate formula;

$$t = \frac{N_{\text{Pb}}}{\lambda N}$$

Denoting the number of alpha particles emitted per hour from the uranium series or thorium series in radioactive equilibrium contained in 1 mg of mineral, by $\bar{N}_{\text{UI}}$, $\bar{N}_{\text{Th}}$ and remembering the relationships



the number of atoms disintegrated in the uranium or thorium contained in 1 gr of mineral during one year, λN is

$$\lambda N(\text{U}^{238}) = \frac{\bar{N}_{\text{UI}}}{8} 10^3 \cdot 24 \cdot 365$$

$$\lambda N(\text{Th}^{232}) = \frac{\bar{N}_{\text{Th}}}{6} 10^3 \cdot 24 \cdot 365$$

Replacing N_{Pb} with m_{Pb} : the mass of radiogenetic lead in 1 mg of mineral, we have

$$\begin{aligned} t &= \frac{6.06 \cdot 10^{23} \cdot m_{206\text{Pb}} \cdot 8}{\bar{N}_{\text{UI}} 10^3 \cdot 24 \cdot 365 \cdot 206} = \frac{6.06 \cdot 10^{23} \cdot m_{208\text{Pb}} \cdot 6}{\bar{N}_{\text{Th}} 10^3 \cdot 24 \cdot 365 \cdot 208} \\ &= \frac{m_{206\text{Pb}} \cdot 2670 \cdot 10^{12}}{\bar{N}_{\text{UI}}} = \frac{m_{208\text{Pb}} \cdot 1970 \cdot 10^{12}}{\bar{N}_{\text{Th}}} \end{aligned}$$

Using 10^6 years and ppm Pb for the unit of time and content of lead, and neglecting the slight influence to the coefficient by the actinium series,

$$t = \frac{m_{\text{Pb}} (\text{total})}{\bar{N}_{\text{UI}} + 1.35 \bar{N}_{\text{Th}}} \cdot 2670 \quad (1)$$

The age of the suitable mineral may be determined by use of this equation⁴⁾.

N. B. KEEVIL⁵⁾ (1939) has presented a correction formula for a mineral older than 300 million years.

$$t = t_1 - \frac{1}{2} K t_1^2 \quad (1')$$

where t_1 is an age obtained from equation (1) and K is equal to

$$K = 1.27 \cdot 10^{-10}.$$

The use of equation (1') with the value mentioned above for K results in error less than six per cent, even for two and a half billion years, in the range of Th/U from zero to ten.

Zircon as suitable mineral

The conditions required for a mineral to be suitable for this method are that the mineral should have only radiogenetic lead, have lost no lead, have only primary radioactive elements and lost none of them. In the fresh igneous rock, the radioactivity is frequently concentrated in the accessory minerals. Among these radioactive minerals, zircon has been considered to be adequate for this method of age determination.

Surely, little chance would be left for the primary lead to come into zircon, because the ionic radii of zirconium ($0.8 \text{ \AA}$) and lead ($1.32 \text{ \AA}$) are remarkably different and the close packing of zircon is excellent. In the fresh igneous rocks the zircon seems to be well protected from the alteration process that would introduce or remove radioactive material or lead.

Many results, which were carried out by LARSEN *et al.*¹⁾ and G. R. WEBBER *et al.*⁶⁾ with pre-Cambrian or Paleozoic zircon, show good agreements with the stratigraphical study. Therefore, the fact seems to affirm indirectly at least the consideration concerning the zircon mentioned-above.

Nevertheless there were occasions when grave discrepant results were reported which appeared to be more than simple precision error in analysis. For example M^{me} A. HÉE *et al.*⁴⁾ reported about the zircon from the granite d'Andlau, and G. R. WEBBER *et al.*⁵⁾ about the zircon from the Northbridge granite gneiss near Uxbridge, and also that from Fletcher granite gneiss.

HURLEY *et al.*⁷⁾ made it clear that there are three ways in which the radioelements are distributed in the zircon crystals.

1) Zircon has a limited tolerance for uranium and thorium atoms at the time of its crystallization and might take into its structure a uniformly distributed proportion of these elements.

2) If conditions changed during the crystallization of the zircon, it might be zoned with various proportions of the radioelements in concentric zones in the individual crystals.

3) It was found that some zircon crystals contain other radioactive crystals as inclusion. In this case, the overall zircon crystal might have a rather uniform level of activity and distinctly separate phase representing a spheroidal region about the radioactive inclusions, that is much more highly metamict.

Those which have too strong radioactivity would be unsuitable. Hurley studying metamict of zircon put the upper limitation. As a general rule any

material with an activity greater than 500 alphas/mg/hr should be suspected of having either inclusion of other radioactive minerals or else, particularly in the case of accessory zircons from granites, a highly radioactive skin layer. But according to HÉE, even the zircon of 730 alphas/mg/hr is used in success.

Furthermore, an interesting type of zircon was recently reported in this country.

Y. KARAKIDA⁸⁾ found some overgrown zircon in the granitized gneiss and granitic rocks of Ryôke metamorphic zone in the Yanai area. He describes that such rocks containing the overgrown crystals may be originated by mixing of sediments, which had detrital zircon now seen as relict cores of one characteristic zircon overgrowth (called type A) and granitic liquids, which were crystallizing euhedral zircons now seen as relict cores of the other type (B and C) as well as parts of type A.

This seems to be very important for the age determination of using zircon by the next two reasons: 1) Some zircon overgrowths might have two geologically different ages of the shell and core in the same zircon grain. 2) The apparent ages obtained from the zircon overgrowth may have some petrological significances. Nevertheless the abundance and locality of such overgrown zircon may be limited in general. Consequently, it has become clear that homogeneous and euhedral zircon is the most secure as the sample for the age determination and zircon from the metamorphic rock may have a danger to fail. It must be attentively treated with the zircon from the granite suspected its origin of metasomatism. In practice, it will be sufficient to make use of colourless or light-coloured transparent crystals in the case of ordinary granite.

Matters to be determined

As apparently seen from the equation (1), there are only three matters to be determined for this method, i. e., (1) content of lead (ppm), (2) intrinsic emissions of alpha-particles from the uranium series in 1 mg of zircon per hour, and (3) that from the thorium series.

1. *Relation between apparent counts and intrinsic emissions*

The basic relation between the number of counts obtained by some measuring equipment and that of the intrinsic alpha-emissions within a radioactive solid has been indicated by the analytical expression by FINNERY and EVANS⁹⁾ (1935) for the first time. Thenceforth, some different forms of it are reported by CURIE, I.¹⁰⁾ (1946) and NOGAMI *et al.*¹¹⁾ (1940) etc., corresponding to respective counting method. HATUDA¹²⁾ (1961) *et al.* have recently made a formula suitable for 4π gas-flow counter. The number of α -counts per second (or hour) per cm^2 of the surface of a "thick" solid source in radioactive equilibrium is given by

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

where N_{UI} , N_{Th} ; intrinsic alpha-emissions from uranium I or thorium in one cm^3 of source in radioactive equilibrium, excepting the emissions from its daughter elements, alphas/ cm^3/sec , (or hr),
 μ ; ratio of ranges in mineral to ranges in air of alpha-particle,
 Σ ; summations are made over the members of the U, Th and AcU series,
 a ; thickness of aluminium or Myler foil covering the sample in terms of range of alpha-particles in air (cm),
 ρ ; minimum detectable residual range of alpha-ray in air (cm).

For the thickness of covering foil used, 0.97 cm in air equivalent and the sensitivity of the apparatus, *viz.* minimum detectable residual range 0.20 cm,

$$\Sigma_{\text{UI}} \frac{(R_{\text{UI}} - \rho - a)^2}{R_{\text{UI}} - \rho} = 16.75$$

$$\Sigma_{\text{AcU}} \frac{(R_{\text{AcU}} - \rho - a)^2}{R_{\text{AcU}} - \rho} = 19.34$$

$$\Sigma_{\text{Th}} \frac{(R_{\text{Th}} - \rho - a)^2}{R_{\text{Th}} - \rho} = 17.38$$

where the ranges of alpha-particles used in the calculation were quoted from the work by RANKAMA¹³⁾ (1954).

From Eq. (2),

$$n = \frac{\mu}{2} \cdot (17.62 N_{\text{UI}} + 17.38 N_{\text{Th}}) \quad (3)$$

Similarly, for 0.50 cm and 0.20 cm respectively

$$n = \frac{\mu}{2} \cdot (23.78 N_{\text{UI}} + 22.00 N_{\text{Th}}) \quad (4)$$

where any unit of time will do so long as it remains the same for both n and N_{UI} , N_{Th} .

Corresponding to the thickness of the foil or absorber covering the source in the sample-holder, and the sensitivity of the apparatus used, the numerical constants in Eq. (3) take different values. The foil thickness and sensitivity of the apparatus can be calibrated by standard emitter as shown in the above-mentioned paper.¹²⁾

2. The ratio of range in mineral to that in air of alpha-particles

The ranges of alpha-particles in air have been determined and reported by

many, but not so with those in minerals or the ratio of the latter to the former. Generally they are obtained by calculation following the empirical rule formulated by BRAGG and KLEEMAN¹⁴⁾ (1905). RUTHERFORD¹⁵⁾ (1930) stated that the additive rule of BRAGG and KLEEMAN though very convenient for rough calculation holds only in a first approximation, while NOGAMI *et al.*¹¹⁾ (1948) proved experimentally the rule of BRAGG and KLEEMAN to hold good in many minerals.

YAGODA¹⁶⁾ (1949) referring to the rule above-mentioned, calculated the range of alpha particles in varying substance by following form.

$$R_s = \frac{R_a d_a \psi_s}{d_s \psi_a} = 3.194 \cdot 10^{-4} \frac{R_a \psi_s}{d_s} \text{ (cm)}$$

where R_a, R_s ; ranges in air and substance, respectively,

d_a, d_s ; densities of air and substance, respectively,

ψ_a, ψ_s ; permeability of the medium to alpha particles defined in terms of its atomic composition of air and substance, respectively.

$$\psi = a\sqrt{W_A} + b\sqrt{W_B} + c\sqrt{W_C} + \dots$$

where W_A, W_B, W_C ; atomic weight of component elements A, B, C etc., of the substance, respectively,

a, b, c ; relative proportion by weight of component element A, B, C etc. to the whole.

Calculating the permeability ψ of zircon as ZrSiO_4 to be 6.94 and taking its density to be 4.60, we have as the ratio of range in zircon to that in air,

$$\mu = 4.82 \cdot 10^{-4}$$

3. Determination of Th/U in mineral

As described in the other paper¹²⁾, the total alpha-emissions can be calculated by use of approximate average ratio of thorium to uranium in mineral with the error less than 17%¹⁶⁾. The writer has devised a method for determining the ratio of uranium to thorium in minerals by the energy spectra of alpha rays¹⁷⁾.

The ratio of the number of counts $n_{\rho a}$ and n_0 obtained from the measurements under two different conditions (a, ρ) and (a_0, ρ_0) respectively; a being the thickness of the absorber and ρ the minimum detectable range,

$$\frac{n_{\rho a}}{n_0} = \frac{\left(\frac{n_{\rho a}}{n_0}\right)_U + \left(\frac{n_{\rho a}}{n_0}\right)_{\text{Th}} Y_0/X_0 \cdot \text{Th/U}}{1 + Y_0/X_0 \cdot \text{Th/U}} \quad (5)$$

where Th, U; content of thorium or uranium in sample mineral, gr/gr,

$\left(\frac{n_{pa}}{n_0}\right)_U, \left(\frac{n_{pa}}{n_0}\right)_{Th}$; the ratio of n_{pa} to n_0 in the case of uranium series alone (Th=0) in mineral and of thorium series alone (U=0), respectively,

Y_0/X_0 ; ratio of specific activity i.e., calculated ratio of emissions from only thorium series (U=0) to that from only uranium series (Th=0) per unit mass at the condition (a_0, ρ_0).

The calculated value of Y_0/X_0 is 0.3271 for ($a_0=0.97$ cm, $\rho_0=0.20$ cm).

When the values of $\left(\frac{n_{pa}}{n_0}\right)_U$ and $\left(\frac{n_{pa}}{n_0}\right)_{Th}$ are determined by the calibration of standard minerals, the ratio of the thorium to the uranium in a sample mineral can be obtained from the values of $\left(\frac{n_{pa}}{n_0}\right)$ measured with the mineral using Eq. (5). The details of the calculations will be shown in the chapter after next.

4. Measurement of lead

As the quantities of lead treated in this age-determination method is as low as $n \times 10 \sim 100$ ppm., even in the zircon from the paleozoic granite, spectrographic method of measurement was adopted for the quantitative determination of lead, in which the most reliable value will be obtained by the use of standard composed of the same substance as the sample in question.

The spectrographic measurements of lead in zircon were carried out by Dr. MORITA, K.¹⁵⁾ at the Government Industrial Research Institute, in Nagoya, with JACO 3.4 m grating spectrograph. The experimental conditions in his analyses are as follows,

Spectrograph; JACO 3.4 m Plane Grating Spectrograph,

Electrode; Regular Electrode of Nippon Carbon Co.,

Illuminating system; Intermediate imaging,

Grating angle; 9.40° 2nd order,

Slit width; 0.05 mm for 1~100 ppm Pb,

0.02 mm for 50~1000 ppm Pb,

Current; D. C. 7.5 A for 1~100 ppm Pb,

D. C. 9.5 A for 50~100 ppm Pb,

Analytical gap; 3 mm,

Exposure time; 30 sec for 1~100 ppm Pb,

40 sec for 50~1000 ppm Pb,

Photographic process; Fuji Spectroscopic Process Plate Development,

4 min; at 20°C in FD-31; Fixing 5 min, at 20°C in FF-H4,

Densitometry; JACO Recording Microphotometer J. A. 2310,

Emission; Iron arc. Step sector. Each plate was calibrated by the Seidel Method.

Detail of practical measurement

1. *Apparatus*

The equipments used in the radiation measurement are composed of 4π gas-flow counter installed with a preamplifier, a stabilized high voltage source, a linear amplifier, a discriminator and scaling units. The 4π gas-flow counter (type PC-V, Tokyo Atomic Industry Co., Ltd.) is operated at the proportional region by use of PR-gas. This is connected with a linear amplifier used at a gain of 1600, which is a half of its capacity, with regard to the stability. The discriminator is able to set from 3.2 v to 100 v for the input of the entrance of Schmidt-trigger circuit, and is employed as the means of controlling the sensitivity of the counter circuits.

2. *Preparation of sample zircon*

Zircon is very widely distributed as an accessory constituents of all kinds of igneous rocks in concentration of about 0.01~0.05%, but it is especially common in syenite, granite and diorite. The average granite contains about 0.02% of zircon. If 50 mg of sample would be needed for the measurement, it is necessary to separate it from about 1 kg of granite, considering the practical yield rate of zircon to be about 1/4 through the concentrating and selecting process. Separation—About 1 kg of granite taken at random from several kg of fresh rock are crushed with a crusher to pass through a 60 meshes (0.2 mm opening). The rock-powder is separated into relatively heavy minerals with a superpanner. Removing the strong magnetic material in the powder by a permanent magnet, the minerals are washed with dilute HCl to clean the surface, and then with water. The heavies are next separated roughly into fractions with the Frantz Isodynamic separator. The nonmagnetic fraction is separated with methylene iodide into the heaviest mineral fraction. The last heavies are again washed with conc HNO_3 to remove the contaminated lead and some sulphide minerals, then with water. For the final selection of zircons suitable for the measurement, it is necessary to treat them by hand-picking under a binocular microscope.

Zircon samples are ground in a mortar of sintered alumina into powders whose size is less than about 5μ . Being put in the sample-holder, the ground sample should be rested at least for a week to recover the radioactive equilibrium. The sample-holder is a thin aluminium plate, in the center of which exists a circular hole of 0.3–1.5 cm in diameter. The sample is put between the thin aluminium foils like a sandwich and set in the hole. The thickness of the foil and area of the hole should be known exactly. The prepared sample-holder is then put in the center of the gas-flow counter.

For the measurement in the state of "thick" source with respect to alpha-particles of thorium and uranium series, it is sufficient to take about 15 mg of mineral per 1 cm^2 of surface area of the hole. The number of counts no longer

increases with the addition of sample in mass, and becomes proportional only to the surface area of the hole containing the powder.

Measurements and results

1. Calculation of ratio Th/U

The data obtained from several specimens and standard samples are shown in Table 1.

Table 1. Values of $n_{\rho a}/n_0$ of specimens and standard samples
($a_0=0.97$ cm, $\rho_0=0.20$ cm), ($a=2.91$ cm, $\rho=1.5$ cm)

	TB-1	HU-3	F-5	Stand. 1	Stand. 2	Stand. 3	Stand. 4
$\frac{n_{\rho a}}{n_0}$	0.1005	0.1098	0.0787	0.1247	0.1216	0.1141	0.0983
$\frac{Th}{U}$				22.62	3.44	1.14	0.24

n_0 is the number of counts in the measuring condition in which a sample is merely covered with a holding foil whose thickness has been estimated to be 0.97 cm in air equivalent, and the discriminator is set at a certain fixed point. In this normal conditions the sensitivity or the minimum detectable range (ρ_0) of alpha particle for the instrument is 0.20 cm as stated already. $n_{\rho a}$ is the number of counts in the condition (ρ, a) in which the surface of the sample is covered with added absorber-foils whose thickness is estimated to be 2.91 cm including that of the holding foil, and the discriminator is set at another fixed position corresponding to ca. 30 volts. The value of ρ in this case is probably about 1.5 cm estimated from the relation $n_{\rho a}/n_0$ vs. (ρ, a). It is no use of finding an exact value.

In order to find out the most probable values for $\left(\frac{n_{\rho a}}{n_0}\right)_U (=b)$ and $\left(\frac{n_{\rho a}}{n_0}\right)_{Th}$ ($=a$), the equation (5) is modified into the following expression.

$$\left(\frac{n_{\rho a}}{n_0}\right) (1+x) = ax+b \quad \text{or} \quad y = ax+b$$

The steps of calculation and reduction to the linear form for the method of least squares to obtain $\left(\frac{n_{\rho a}}{n_0}\right)$ for uranium ($Th=0$) or thorium ($U=0$) series only, are shown in Table 2.

The constant terms determined by the above four observed values of the standard sample are 0.1309 for $\left(\frac{n_{\rho a}}{n_0}\right)_{Th}$ and 0.0990 for $\left(\frac{n_{\rho a}}{n_0}\right)_U$. Then the equation (5) is,

$$\frac{n_{pa}}{n_0} = \frac{0.0990 + 0.1309 \cdot 0.3271 \cdot \text{Th/U}}{1 + 0.3271 \cdot \text{Th/U}}$$

Table 2. The steps of calculations for the method of least squares (standard samples)

	Standard sample 1	Standard sample 2	Standard sample 3	Standard sample 4	
$\frac{\text{Th}}{\text{U}}$	22.62	3.44	1.14	0.24	
$\frac{Y_0}{X_0} \frac{\text{Th}}{\text{U}}$	7.399	1.1252	0.3729	0.0785	
$1 + \frac{Y_0}{X_0} \frac{\text{Th}}{\text{U}}$	8.399	2.1252	1.3729	1.0785	$Y_0/X_0=0.3271$
$\frac{n_{pa}}{n_0}$	0.1247	0.1216	0.1141	0.0983	
$\frac{n_{pa}}{n_0} \left(1 + \frac{Y_0}{X_0} \frac{\text{Th}}{\text{U}}\right)$	1.0474	0.2584	0.1566	0.1060	

The Th/U of the above specimens are calculated from this equation, the results are shown in Table 3.

Table 3. Calculated Th/U of the specimens

TB-1	HU-3	F-5
0.15	1.56	negative*

*probable radium deficiency?

2. Determination of age

The relations called for the calculation of the age of zircon are as follows ;

$$t = \frac{m_{\text{Pb}} \cdot 2670}{\bar{N}_{\text{U}} + 1.35 \bar{N}_{\text{Th}}}$$

$$\frac{N_{\text{Th}}}{N_{\text{UI}}} = \frac{6.06 \cdot 10^{23} \cdot \text{Th} \cdot d \cdot \text{Th} / W_{\text{Th}}}{6.06 \cdot 10^{23} \cdot \text{UI} \cdot d \cdot \text{UI} / W_{\text{UI}}} \approx 0.33 \frac{\text{Th}}{\text{U}}$$

$$\bar{N}_{\text{UI}} = 8 \cdot 10^{-3} N_{\text{UI}} / d = 1.74 \cdot 10^{-3} N_{\text{UI}}$$

$$\bar{N}_{\text{Th}} = 6 \cdot 10^{-3} N_{\text{Th}} / d = 1.30 \cdot 10^{-3} N_{\text{Th}} \quad (d=4.6 \text{ for zircon})$$

$$\text{efficiency correction*} \quad + 10.0\%$$

$$n = \frac{4.82}{2} \cdot 10^{-4} (17.62 N_{\text{UI}} + 17.38 N_{\text{Th}})$$

$$n ; (\text{counts/cm}^2/\text{hr})$$

$$\left. \begin{array}{l} a=0.97 \text{ cm} \\ \rho=0.20 \text{ cm} \\ \mu=4.82 \times 10^{-4} \end{array} \right\}$$

* This correction factor was determined by the calibration with the standard zircon sample (R-1932, Geol. Survey Bull., 1097-A, p. 21, Table 6), as described in another paper.¹²⁾

The data of the measurements and steps of calculations following the relation mentioned above, are shown in Table 4.

Table 4. Observed and calculated data on zircons

	HU-3	F-5	TB-1	unit
counts	9.98 ± 0.09	8.69	3.88	cpm
surface area	0.2248	0.5236	0.5463	cm ²
counts	44.40 ± 0.40	16.60	7.10	$\alpha/\text{cm}^2/\text{min}$
	2662 ± 24	996	426	$\alpha/\text{cm}^2/\text{hr}$
Th/U	1.56	0	0.15	
$N_{\text{UI}}/N_{\text{Th}}$	0.52	0	0.05	
N_{UI}	41.60×10^4	23.5×10^4	9.6×10^4	$\alpha/\text{cm}^3/\text{hr}$
N_{Th}	21.67×10^4	0	0.5×10^4	$\alpha/\text{cm}^3/\text{hr}$
$\bar{N}_{\text{UI}}$	724	411	167	$\alpha/\text{mg}/\text{hr}$
$\bar{N}_{\text{Th}}$	282	0	7	$\alpha/\text{mg}/\text{hr}$
$\bar{N}_{\text{UI}} + 1.35\bar{N}_{\text{Th}}$	1105	411	174	$\alpha/\text{mg}/\text{hr}$
efficiency correction (10.0%)	1215	452	191	$\alpha/\text{mg}/\text{hr}$
mpb	51	59	20	ppm
age	111	343 331*	274	10 ⁶ year

* age corrected for diminution of the parent element using Eq. (1')

Age and locality of granites

1. The specimen HU-3 is the zircon of the Hiei granite, sampled at a quarry in the Uryû-Yama, Kitashirakawa, Kyoto Pref.. The Hiei granite is biotite granite, occasionally with small quantity of hornblende. The accessory minerals are magnetite, zircon, apatite and allanite.

This granite intruded into the Paleozoic formation which are often altered to hornfels by contact metamorphism. The margins of this granite body are holded in several places by the Plio-Pleistocene formations called "Paleo-Biwako".

Concerning the age of intrusion of the Hiei granite, HAYASE, I.¹⁹⁾ estimated that it had occurred in earlier Tertiary; the age is $67.09 \cdot 10^6$ years old by A-K method, while the age of HU-3 zircon determined by the method of lead-alpha is $111 \cdot 10^6$ yr.

2. The specimen TB-1 is the zircon from the granite-porphyry sampled from a quarry at the Binwari-Yama near Ômi-Hachiman, Shiga Pref.. In this granite-porphyry phenocrysts of quartz, orthoclase, plagioclase and some biotite almost altered to chlomite, are recognized. The ground mass is composed of minor crystals of feldspar, quartz, epidote, magnetite and zircon. Generally this rock looks fresh at a glance but essentially metamorphosed by auto- or contact-

metamorphism.

The age obtained for TB-1 zircon is $274 \cdot 10^6$ yr.

It seems that the age calculated is too large as compared with that expected for the granite in this locality. It may be explained by the fact that this granite-porphyry has been suffered hydrothermal metasomatism in which leaching of some radioactive isotopes or less probable contamination by lead might have accompanied. These zircons are mostly composed of a quantity of fine crystals and less amount of those of normal size (ca. $100-150\mu$).

These effects on the age determined by this method require further examination.

3. The specimen F-5 is the zircon separated from the "Gairome"-clay, in Fuzioka Mine, Aichi Pref., offered by Dr. MORITA. The original rock of this zircon is not distinct, and as described in Table 3, the calculated value of Th/U of this specimen is found negative. This suggests that the contained radioelements may be out of radioactive equilibrium, perhaps owing to the conditions in which the sample was placed. Accordingly, the age for F-5, $311 \cdot 10^6$ yr. is unduely large.

Conclusion

The calibration of the constant in the equation expressing the relation between counting and its intrinsic radioactivity has been once carried out, the measurement of alpha-activity of samples is easy to carry into execution. It is relatively laborious and important to prepare samples, especially to select the suitable minerals by hand-picking under a binocular. It requires a strict precaution to keep away from contamination of lead.

Zircons sampled from metamorphosed rock or young sediments seem to be improper for the age determination.

Correction in emissions of alpha rays for the ratio of thorium to uranium in each sample, will be useful for the accurate determination of the age.

The Hiei granite is estimated to be $111 \cdot 10^6$ years of age.

Acknowledgment

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