〔論文〕

Annual Dose Estimation in TL-Dating Using Alpha-Counter

Masao HAYASHI[1] and Jianming XU[2]

アルファ線計測器を用いた熱蛍光年代測定における年間線量の評価

林　正雄
許　建明

Abstract: Contents of radiometric elements of U and Th have been determined using an alpha-counter. It requires rock samples less than 10g, which are much smaller than those needed for gamma- ray spectroscopy. Numbers of alpha-particles were counted in two ways: one being exposed (Air), and the other is with an aluminum foil cover (Al). The ratio of Air to Al gives a high correlation coefficient in multivariate equations to estimate the content of U or Th. Since all similar equations for K2O contents give smaller correction coefficients, it must be determined by other methods including x-ray spectroscopy. In conclusion, the grain method which measures the age of each grain, is recommended, of which the annual dose is determined by the techniques proposed here.

Keywords: annual dose, TL-dating, alpha-counter, x-ray spectroscopy

1. Introduction

In the thermoluminescence (TL) dating, the annual dose is one of the two parameters to obtain the age of rock samples. There exist some techniques which have been commonly used. In situ observation of the annual dose would be most accurate with some dosimeters set at the place where the dating sample was collected. This technique, however, requires many months to conduct so it is rarely used. Gamma-ray pectroscopy[1)] is commonly used in determining the contents of all the radiometric elements, U, Th and K2O, that are indispensable for the dating. One of the defects of this technique is that it needs large quantities of rock samples. Chemical analysis of those radiometric elements is also adopted as a time-saving technique[2)], although it is impossible to evaluate the degree of disequilibrium in the decay series of U and Th. This study proposes a simple technique to estimate the contents of the three elements, using an alpha-counter for U ant T, and an X-ray spectrometer for K2O.

1) Kyushu Sangyo University, Fukuoka 813-8503, Japan
2) Hunan Agricultural University, Furong, Changsha 410128, China

2. Experiments

2.1 Standard rock samples

The relation between alpha-ray intensities and radiometric elements of U and Th were determined using the standard powdered rock samples from the Geological Survey of Japan. These six samples include: JB-3 (basalt), JA-3 (andesite), JG-3 (granodiorite), JG-1a (granodiorite), JR-1 (rhyolite), and JR-2 (rhyolite). Their chemical compositions, both major and minor elements, have been reported by many experts[3]. Annual doses of the standard samples were calculated by Aitokin's equation (18), in which the diameter of quartz is 1mm (B=0.81) and the water content is ignored (W=0).

2.2 Alpha-counting

The standard rock powder of about 10g was packed in a stainless holder 50mm in diameter and 5mm thick. Numbers of alpha-particles emitted from U and Th were counted using an Alpha-Counter Model ZD-451FU connected to a Basic Scaler TDC-105 from ALOKA Co. Ltd. Measurements were carried out in two different ways: one is "Air" without a cover, and the other is "Al" with an aluminum foil cover with a thickness of 25 micro-meters. Each method was counted three-times per hour to obtain the mean cph (counts per hour).

2-3. X-ray spectroscopy

To determine the contents of K2O, the above standard sample in the same holder was also used in an x-ray fluorescence analyzer, R-233 from Rigaku Co..Ltd. Experimental conditions are as follows: accelerate voltage = 50kV, tube current = 50mA, scanning speed = 1 degree per minute, all performed in a vacuum atmosphere. The rock powder was covered with a very thin sheet of Mylar, which is commonly used, to prevent spilling. The number of x-rays is given in counts per second (cps).

3. Results

3.1 U and Th contents

Table 1 shows the results of the number of alpha-particles counted (Air and Al) and the contents of the radiometric elements of U and Th. The counted numbers are subtracted by the background noise of 2.6 cph. The Air/Al in the table is the ratio of Air to Al that is expected to improve the correction coefficients in equations to estimate the contents of radiometric elements.

Table 1 CPH for standard rock samples

Sample	Th(ppm)	U(ppm)	K2O(%)	Air(cph)	Al(cph)	Air/Al
JB-3	1.3	0.5	0.8	11.1	5.1	2.17
JA-3	5.0	1.4	1.4	21.4	7.1	3.01
JG-3	8.0	2.0	2.6	43.7	14.7	2.97
JG-1a	12.1	4.7	4.0	83.4	26.4	3.16
JR-1	26.5	9.0	4.4	147.7	36.1	4.09
JR-2	32.2	10.5	4.5	180.4	48.1	3.75

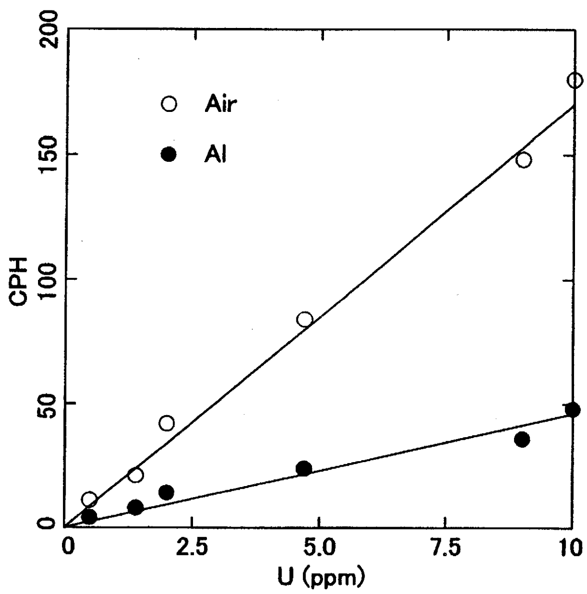

Fig. 1 CPH vs. uranium contents

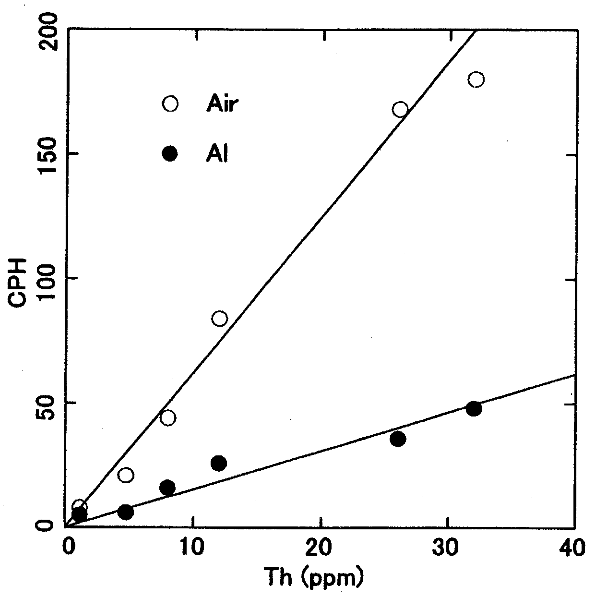

Fig.2 CPH vs. thorium contents

It is made obvious in Figs. 1 and 2 that the numbers of counted alpha-particles of U and Th are proportional to their contents. Through statistical analysis, the results give the following equations, in which Ratio is the ratio of Air to Al, r is the correction coefficient.

$$U(ppm) = 0.059Air \quad (r = 0.999) \qquad (1)$$

$$U(ppm) = 0.222Al \quad (r = 0.995) \qquad (2)$$

$$U(ppm) = 0.082Air - 0.087Al \quad (r = 1.000) \qquad (3)$$

$$U(ppm) = 0.085Air - 0.102Al + 0.041Ratio \quad (r = 1.000) \qquad (4)$$

$$Th(ppm) = 0.176Air \quad (r = 0.998) \qquad (5)$$

$$Th(ppm) = 0.648Al \quad (r = 0.984) \qquad (6)$$

$$Th(ppm) = 0.243Air - 0.253Al \quad (r = 0.998) \qquad (7)$$

$$Th(ppm) = 0.274Air - 0.410Al + 0.424Ratio \quad (r = 0.999) \qquad (8)$$

Of these equations, (4) and (8) have highest correction coefficients of 1.000 and 0.999, in which the parameter of the Air/Al is introduced.

3.2 K2O content

Equations obtained for K2O similar to U and Th are as follows:

$$K2O(\%) = 0.031Air \quad (r = 0.955) \qquad (9)$$

$$K2O(\%) = 0.116Al \quad (r = 0.973) \qquad (10)$$

$$K2O(\%) = -0.450Air + 0.284Al \quad (r = 0.983) \qquad (11)$$

$$K2O(\%) = -0.021Air + 0.158Al + 0.341Ratio \quad (r = 0.990) \qquad (12)$$

Although, the equation (12) has a high correction coefficient of 0.990, it is not enough to estimate accurate annual doses for the TL-dating.

Fig. 3 shows the results of the number of x-rays counted and the content of K2O. They are proportionate to each other, giving a linear equation with a high correction coefficient of 0.999 as follows.

$$K2O(\%) = 9.97 \times 10^{-5} X \quad (r = 0.999) \qquad (13)$$

, where X is the intensity of X-rays (cps).

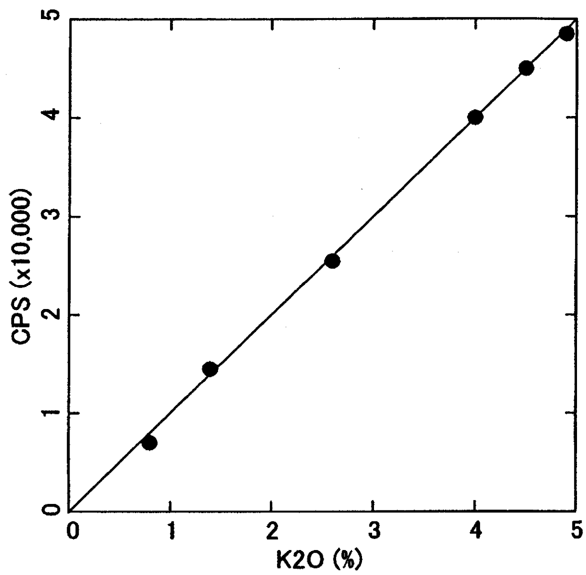

Fig.3 CPS vs. K2O contents

3.3 Annual dose

Equations obtained for the annual dose in the case where the diameter of quartz is 1mm, and water content is zero, are as follows:

$$AD(mGy) = 0.034Air \quad (r = 0.966) \qquad (13)$$

$$AD(mGy) = 0.129Al \quad (r = 0.976) \qquad (14)$$

$$AD(mGy) = -0.210Air + 0.206Al \quad (r = 0.978 \qquad (15)$$

$$AD(mGy) = 0.024Air - 0.026 + 0.625Ratio \quad (r = 0.999) \qquad (16)$$

4. Discussion

4.1 Age equation

An age equation in the TL-dating is given as follows:

$$Age(ka) = \frac{ED}{AD} \quad (17)$$

Here the ED is the equivalent dose (paleodose), which is the total dose of a dating sample from its formation, and, the AD is the annual dose, which is the dose amount per year. Accordingly, the AD is of the same importance as the ED.

Except for the in situ observation, the AD is calculated by the next equation[4],

$$AD(mGy) = (0.1148U + 0.0514T + 0.2609K)$$
$$/(1+1.14W) + (0.1462U + 0.0286T + 0.6893K)$$
$$B/(1+1.25W) \quad (18)$$

Here U is uranium content (ppm), T is thorium content (ppm), and K is K2O content (%), W is water content, and B is the factor controlled by grain size. This equation suggests that the AD depends only upon the contents of radiometric elements, except for the parameters of W and D.

The D factor for grain-size of quartz can be evaluated by some techniques. To measure its grain-size in thin rock sections under a microscope is laborious work and is hard for soft rock samples[5]. It is reasonable to use grains of a limited grain-size, for example, 0.5 to 1mm in diameter[6]. The simplest would be the grain method that dates for each grain whose diameter can be measured under a microscope. One of these techniques would provide a reliable value of the D parameter.

The W parameter will be the most difficult to evaluate. Even in the in situ method, the water content at a place where dosimeters are set might have been changed through geological periods of time. Therefore, there exists no technique to estimate exactly the mean of the water content of dating samples from their formation.

4.2 U and Th contents by alpha counting

The AD in equation (18) should be influenced by the degree of equilibrium in the decay series of U and Th, when dated samples are younger than about half a million years. This implies that the radiometric contents of U and Th by chemical methods, may result in the underestimation of the AD. Consequently, the radiometric techniques are superior to chemical ones for younger dating samples.

Of the radiometric techniques, gamma-ray spectroscopy requires a mass of rock powder of about 200g, which is difficult to obtain in some cases. Conversely, the alpha-ray method used here, needs less than 10g of rock powder, because alpha particles penetrate the powder with a depth of only a few tens of microns. In addition, the accuracy of this is almost the same as that of the gamma-ray technique, when the parameter of the Air/Al ratio is introduced as shown in equations (4) and (8). The reason is that both U and Th decay to produce daughter elements, in which those from U have higher energy, resulting in the lower Air/Al ratios. In addition, the U contents is almost to proportional to the Th content with a high correction coefficient as expressed below.

$$U(ppm) = 0.332Th \quad (r = 0.998) \quad (19)$$

4.3 K2O content

The four equations (9) to (12) to evaluate the content of K2O have correction factors lower than 0.990, which is not enough for the annual dose determination. This is natural because the alpha-counter cannot detect beta- and gamma-rays from the radiometric element K. The reason for relatively high correction factors of those equations may depend on the fact that the K2O content increases quasi linearly with both the U and Th contents in unaltered igneous rocks, like the standard samples used here.

$$K2O(\%) = 0.518U \quad (r = 0.946) \quad (20)$$

$$K2O(\%) = 0.172Th \quad (r = 0.944) \quad (18)$$

For altered rocks whose radiometric elements have been dissolved away to different degrees, their correction coefficients must be lowered accordingly.

Accurate K2O contents can be determined by the x-ray spectroscopy used here, or, some other methods including atomic absorption and ICP mass spectroscopy.

4.4 Annual dose

Equation (16) shows that the alpha-counting method makes it possible to evaluate directly the annual dose itself, at least in the case where dating specimens are of fresh igneous rocks. This depends on the fact that the elements of U, Th and K20 have a linear relation in their contents as shown by equations (19) to (21). In the case of altered rocks, however, those radiometric elements must have been dissolved away to different degrees, so that equation (16) cannot be applied to the rocks.

4.5 Recommended method

From the above discussion, the radiometric methods including gamma-ray spectroscopy and the alpha-counting are superior to chemical methods, which cannot evaluate the degree of disequilibrium in the dacay of U and Th series. Of the two radiometric methods, the alpha-counting technique requires a small amounts of samples of about 10g for the annual dose determination. As to the D factor, it is the simplest to measure the major and the minor axes of each grain of quartz. The grain-by-grain method proposed for the fission track dating[7], should be employed for the TL-dating too.

From the above discussion, it can be concluded that the grain method which measures the age of each grain, is recommended whose annual dose is determined by the alpha-counting for U and Th, and x-ray spectroscopy for K2O.

5. Summary

A simple method to evaluate the annual dose in TL-dating is proposed using an alpha-counter.

(1) The alpha-counting method requires rock samples less than 10g.

(2) Both U and Th contents can be determined precisely when a parameter of Air/Al is introduced.

(3) K2O contents must be determined by other methods including x-ray spectroscopy used here.

(4) The annual dose itself can be also determined by the same method for fresh igneous rocks.

(5) The grain method which measures the age of each grain, whose annual dose determined by the techniques proposed here, is recommended.

Acknowledgement

The writers are indebted to Dr. K. Koga for her guidance to the spectroscopy using an x-ray fluorescence equipment from the Advanced Instruments Center, Kyushu Sangyo University.

References

1) Takashima, I. (1985) Thermoluminescence dating of volcanic rocks and alteration minerals and their application to geothermal history. Bull. Geol. Surv. Japan, 36, 321-366.

2) Hayashi, M. (1997) Otake-Hatchobaru geothermal field in Japan in relation to the thermoluminescence index. Proceedings of 18 PNOC-EDC Geothermal Conference, 29-35.

3) Ando, A., Mita, N. and Terashima, S. (1987) 1986 values for fifteen GSI rock reference samples, "Igneous Series". Geostandards Newsletter, 11, 159-166.

4) Aitken, M. J. (1967) Thermoluminescence. Science Jour. 1, 32-38.

5) Shimao, T., Takashima, I., Watanabe, K. and Izawa, E. (1999) Verification of errors and reliability of thermo-luminescence age for volcanic rocks – Precise beta-ray correction age of pyroclastic flow deposits of Unzen volcano, SW Japan. J. Japan. Assoc. Min. Petr. Econ. Geol., 94, 109-119.

6) Xu, J. (2003) Basic studies on ESR and TL dating methods for quatz. Dr. Thesis of Graduate School of Social and Cultural Studies, Kyushu University, 1-114.

7) Neaser, C. W. and McKee, E. H. (1970) Fission track and K-Ar ages of Tertiary ash-flow tuff, north-central Nevada. Bull. Geol. Soc. Amer., 81, 3375-3381.