〔論文〕

TL and ESR Dating of Quartz Phenocrysts in Volcanic Rocks

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火山岩斑晶石英のＴＬおよびＥＳＲ年代測定

許　建明
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Abstract: Quartz separated from volcanic rocks of known age from 0.2 to 690 ka has been dated using the TL and the ESR methods. Although there are no essential differences in the obtained ages between the two, the TL method appears to be superior to the ERS method in the following points: (1) The TL signal of quartz is extremely and highly susceptive and it can be also applicable for samples younger than 1 ka. (2) The TL signal of quartz increases in intensity quasi-linearly with radiation doses at least up to 10,000 Gy, which is much larger compared to various ESR signals. (3) The TL single grain method, consequently, can be used to solve the grain-size problem for the annual dose determination.

Keywords: quartz phenocrysts, volcanic rocks, TL-dating, ESR-dating

1. Introduction

The TL (thermoluminescence) method is one of prevailing techniques used to determine the age of geologic material younger than a few million years, such as volcanic rocks, altered rocks, and, fault gauge.

Conversely, the ESR (electron spin resonance) method is less popular restricted to material from fault gauge and shell fossils.

This study aims to compare both methods in determining the age of formation of quartz phenocrysts in volcanic rocks from Kyushu, southwest Japan.

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The results show that there are no essential differences between the two, but the TL method appears to be superior to the ESR method in sensitivity and linearity of signals.

2. Rock samples

Eight rock samples used in the age determination range of known age from 0.2 to 690 ka were as follows:

(1) Q0690 is hornblende dacite in Kusu Formation at Hosenji, Oita (33°11'05"N, 131°10'35"E). The K-Ar age of the whole rock is 0.69 ± 0.08 Ma[1).]

(2) Q1981 is Nodake Lava of Unzen Volcano from Fukae, Shimabara (32°44'20"N, 130°17'12"E). The K-Ar age of the whole rock gives (0.08±0.02) Ma[2).]

(3) Q1982 is Yugawachi Pyroclastic Flows from

Arima, Shimabara (32°41'56"N, 130°18'13"E). The TL age of quartz is (77±6) ka[3].

(4) Q1983 is Furue Pyroclastic Flows from Fukae, Shimabara (32°44'13"N, 130°19'02"E). The TL age of quartz is (23±1) ka[3].

(5) Q1984 is Mutsugi Pyroclastic Flows from Mutsugi, Shimabara (32°47'10"N, 130°20'28"E). The TL age of quartz is (4.9±0.4) ka[3].

(6) Q1985 is Shinyake Lava from Senbongi, Shimabara (32°46'55"N, 130°19'13"E). This lava is from an eruption which occurred in 1772[4].

(7) Q1986 is Kureishibaru Pyroclastic Flows from Nishimachi, Shimabara (32°47'20"N, 130°19'20"E). The TL age of quartz is (23±1) ka[3].

(8) Q1723 is Handa Pyroclastic Flows of Kuju Volcano from Hosenji, Oita (33°11'11"N, 131°09'52"E). The C-14 age of charcoal is over 32.3 ka[5].

3. Experimental

3.1 Sample preparation

Quartz phenocrysts (0.5–1.0 mm in diameter) were picked up under a stereoscopic microscope, and then they were reacted with HF (48%) solution at room temperature for 1 h, at which the TL emission became strongest.

3-2. TL measurement

In order to detect weak TL emissions, a highly-sensitive TL-measurement system was developed [7]. Five mg of quartz grains were put in a platinum container 5mm diameter and 1mm thick. The container was set on the heater and then heated from room temperature to 450 °C at 1 °C/s. The emitted TL signals were then detected after passing through an infrared cut-off filter (IRC-65L) by a photomultiplier tube (R649). The tube was cooled to −30 °C. The number of photons per second in the temperature range 100–450 °C was recorded.

TL emissions were measured 10 times for each of the natural and irradiated samples, and the number of photons emitted in the temperature range 100–450 °C was averaged. The TL intensities referred to hereafter represent the total numbers of photons minus the number of incandescent photons emitted in the same temperature range.

Fig. 1 illustrates the TL glow curve of Q1985, which was separated from a historical lava flow occurred in 1772. It should be noted that the 200 year old quartz has a clear curve, the same as that of much older samples. The highest intensity, however, is only 18 cps (counts per second). The glow curve consists of two signals with peak temperatures of 320°C and 370°C.

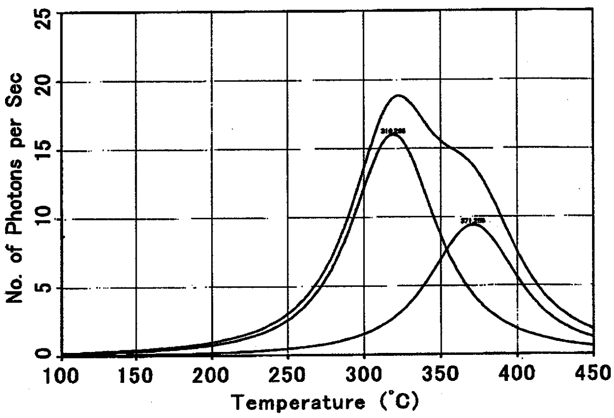

Fig. 1. TL glow curve of quartz phenocrysts (Q1985).

3.3 ESR measurement

ESR spectra were recorded on a JEOL JES-REIX spectrometer under the following conditions:

(1) room temperature and liquid nitrogen temperature.
(2) magnetic field modulation of 100 kHz.
(3) microwave power of 0.01～30 mW.
(4) sweep range of 330±15mT.
(5) amplitude of 0.1 mT.

The measurements were carried out five times for each sample of 100 mg, and the average total dose and its error were computed.

A typical ESR spectrum of the quartz phenocrysts is illustrated in Fig. 2. The Al and Ti centers show a well resolved hyperfine structure consisting of a packet of many lines from low to high fields.

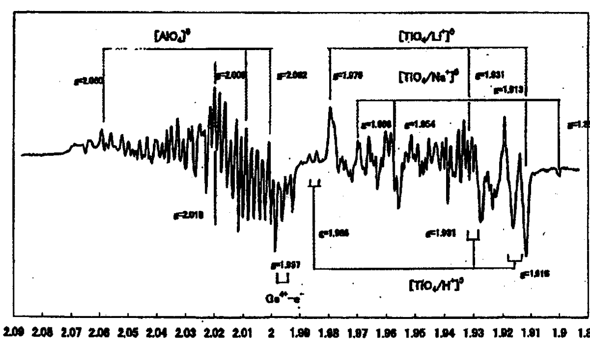

Fig. 2. ESR spectra of quartz phenocrysts (Q1982).

3.4 Gamma-ray irradiation

Artificial ^{60}Co source (Kyushu University) was used to irradiate the samples at a dose rate of ca. 0.45 kGy/h for a total dose of between 100 to 10 kGy.

4. Results and Discussion

The results of the TL and ESR dating are shown in Table 1. In the Table, the AD (annual dose) is given in mGy, most of which was quoted from literature[3] of the known ages for the comparison of the ages obtained.

There are two kinds of ED (equivalent dose) given in Gy: one was determined by the TL growth curve method, and the other by the ESR additive curve method. Both values are quite similar to one another, resulting in the close agreement of the two kinds of ages. Additionally, both the known and the obtained ages are on the 1:1 line in Fig. 3, indicating that the both datings were accurately carried out.

For the youngest sample Q1985, no natural ESR signal could be detected on quartz phenocrysts even of 100 mg, which is 20 times that of the TL dating. This indicates the low susceptibility of ESR signals compared to TL signals. Consequently, its age could not be obtained.

In both of the dating methods, the two parameters of AD and ED must be determined[6]. The AD of quartz grains depends on many factors including the contents of radioactive elements of U, Th and K, the degree of disequilibrium degree of U and Th, water contents, size of grains, etc. Of these, the last is now under debate.

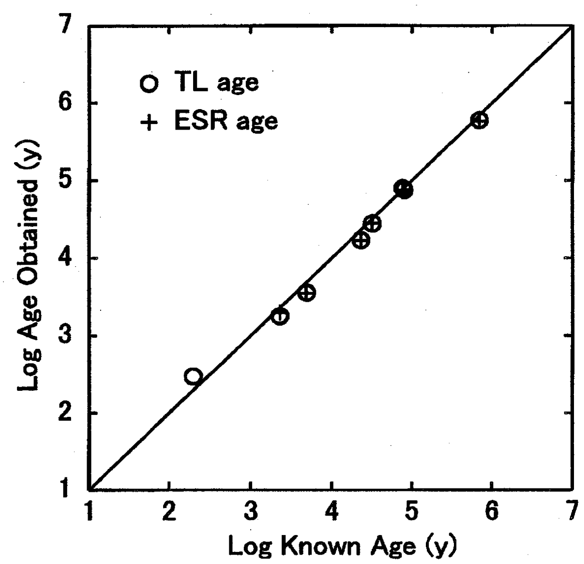

Fig. 3. TL and ESR ages obtained vs known ages.

The AD of quartz which contains virtually no radioactive elements in it, depends greatly on its grain size distribution. One of the techniques to solve this problem is to estimate the grain size distribution using rock sections under a microscope[3].

The simplest solution to the grain size problem may be the single grain method, which uses one large grain of quartz whose diameter or weight is known. Accordingly, the beta-factor of the grain can be calculated. This technique is applicable only for TL method, but it is impossible for ESR since it requires a large amount of any mineral because of its low sensitivity.

For the ED determination, there exists another weakness in the ESR dating method. Most ESR signals attain the saturation or a condition to saturation by low doses of gamma-ray irradiation (Fig. 4). The growth curve for the Ge-center becomes almost flat at doses over 3,000 Ge. This means that older samples cannot be dated. In addition, the Al-center shows a irregular curve[8]. Whereas the TL signal of quartz increases quasi-linearly with the amount of gamma-ray irradiation at least up to 10,000 Gy, corresponding to the age of about 3 million years.

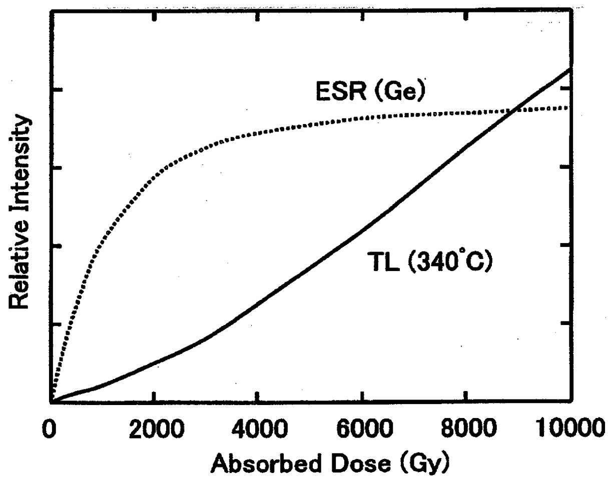

Fig. 4. TL and ESR growth curves of quartz phenocrysts.

The lifetime of TL signals at temperatures above 300°C has been thought to be up to a million years[6]. The annealing experiments[7], however, showed that a TL signal at 310°C weakened more rapidly than we could expected even at low temperatures. This suggests that the age of older quartz samples obtained might have been lowered. In fact, the oldest samples of Q0690 are 14%

Table 1. TL and ESR (Al) ages of quartz phenocrysts in volcanic rocks from Kyushu, southwest Japan. The contents of U, Th, K_2O and water for Q1982, Q1983, Q 1984 and Q1986 are quoted from literature[3].

Sample	U(ppm)	Th(ppm)	K2O(%)	Water(%)	AD(mGy)	ED(Gy,TL)	TL age(ka)	ED(Gy,ESR)	ESR age(ka)	Known age(ka)
Q0690	2.2	8.4	2.12	0	2.68	1591	593	1568	584	690(K-Ar)
Q1981	1.8	7.8	2.22	0	2.63	201	76	20	76	80(K-Ar)
Q1982	2.1	7.8	2.07	1.1	2.55	202	79	186	73	77(TL)
Q1983	2.3	9.4	2.34	1	2.91	49	17	48	17	23(TL)
Q1984	2.6	9.9	2.72	0.25	3.32	12	3.6	11	3.3	4.9(TL)
Q1985	2	8.6	2.37	0	2.84	0.86	0.3	n.d	n.d	0.2(1772)
Q1986	2.4	9.1	2.3	0.5	2.9	52	18	58	20	23(TL)
Q1723	1.9	10.4	1.96	1.6	2.59	73	28	73	28	>32(C-14)

smaller compared to the K-Ar age. The above annealing experiment also suggests that the peak of TL glow curve shift linearly to the higher temperature side with a decreasing total TL intensity. This means that the correction for the obtained age is possible. ESR signals will behave as well as TL signals, so that their ages can also be corrected, if we have annealing data on them.

5. Summary

From the TL and ESR age determination of quartz phenocrysts in volcanic rocks, we can summarize as follows:

(1) Both the TL and ESR ages of quartz phenocrysts are in good agreement with each other, indicating that the latter is also applicable for age determination of the mineral.

(2) The TL signal of quartz is extremely and high susceptive and, consequently it can be dated down to a few hundred years.

(3) The TL signal of quartz increases in intensity qusai linearly with the gamma-ray doses at least up to 10,000 Gy, whereas the ESR signal becomes saturated with much lower radiation.

(4) To solve the grain-size problem for the annual dose determination of quartz, the single grain method can be used for TL dating because of its very high sensitivity.

(5) From the above results, we can conclude that the TL method is superior to the ESR method in reference to various criteria.

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