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Preliminary Study on the Determination of Alteration Age by a Thermoluminescence Method

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Abstract

The specific thermoluminescence (TL) peak heights of quartz separated from Tamagawa welded tuff show a linear correlation to fission-track (FT) ages of 1.1 to 1.8 m.y. However, the specific TL peak heights of altered samples are very low compared to the values expected from FT ages. The ages of 0.13 to 0.48 m.y. are obtained by the TL method. It is considered that the TL was discharged at the conditions of alteration and restored after that. If it is true, the ages of above samples correspond to alteration ones. Moreover, it is quite interesting that the suspected younger alteration areas delineated by the rough TL dating using only their TL peak heights coincide with the hot spring areas or wairakite bearing area.

1. Introduction

The hydrothermally altered land is a good indicator for geothermal activity and is widely studied by many ways. For the estimation of altered ages, the dating of original or associated volcanic rocks were widely used. But, this is the indirect way to determine the alteration ages.

Thermoluminescence (TL) dating was originally applied to archeological field and the dating for pottery was already established (Michels, 1973). For the geological field, TL dating was applied to many minerals, such as calcite (Zeller *et al.*, 1957; Johnson, 1963; Ovchinnikov and Maksenkov, 1969), plagioclase (May, 1977), quartz (Göksu *et al.*, 1974; Hayakawa *et al.*, 1976; Li *et al.*, 1977) etc. However, the attentions of these studies were focused to clear up the ages of the cooling of magma, intrusion of igneous bodies or the settlement of sediments. The application of TL dating to alteration ages has not been carried out so far.

The writer considered that the TL dating may be applicable for the alteration age determination because the stored natural TL was easily discharged by alteration.

In this paper, the results of preliminary experiments are described and the possibility for application of quartz TL to alteration age determination is discussed.

2. Experimental Procedure

The materials under study were phenocrystic crystals of quartz separated from Tamagawa rhyo-dacite welded tuff formation of early Pleistocene and their alteration products. The quartz can easily be separated by hand and almost pure samples were used for all measurements. Also the quartz phenocryst was preserved well during the alteration.

The locations of nine samples and the area of alteration age study were shown in Fig. 1. The rock samples which have large quartz phenocryst of 3 to 10 mm were crushed and shieved to 1 to 2 mm. The quartz grain was hand picked and crushed and shieved to 200 mesh under. Then the

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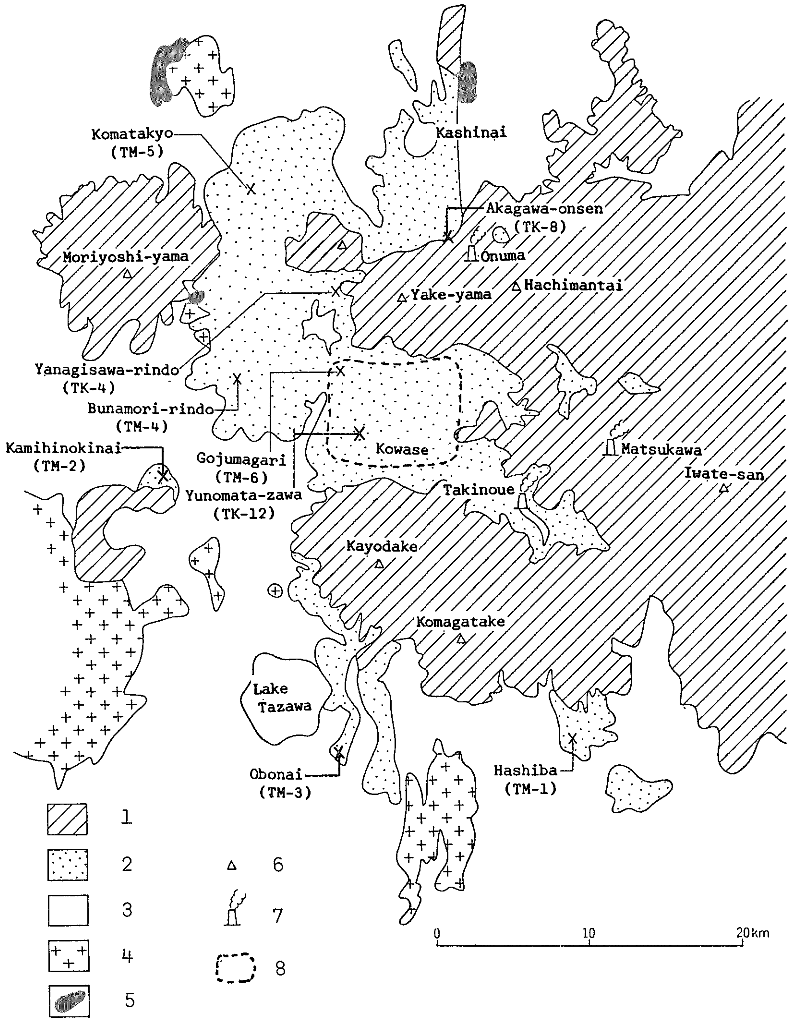
sample was washed three times for removing the fine fraction. After drying, 20 mg of quartz grain was packed into aluminum disc shown in Fig. 2.

The apparatus used was shown in Fig. 2. The TL was detected with 7696 photomultiplier tube which has a maximum sensitivity at 4,400 Å. The TL intensity was expressed as a peak height (cm) which has the proportional relation to stored TL value. The sample was heated from room temperature to about 450 °C at a heating rate of 70 °C/min. in air. The temperatures described in this paper were measured at the center of the sample setting pit and differed from the sample heating ones. So, they were only applicable for this experiment.

In order to estimate the TL accuracy, some preliminary experiments were carried out.

The change of grain size greatly affects the TL intensity. The ratios of TL peak heights for 20–60,



1. Quaternary volcanic rocks	2. Tamagawa welded tuff
3. Tertiary formation	4. Pre-Tertiary granite
5. Paleozoic formation	6. Volcanoes
7. Geothermal power plant	8. Alteration study area

Fig. 1 Distribution of Tamagawa welded tuff and the location of samples.
(Geology was compiled by TAMANYU and SUTO, 1978)

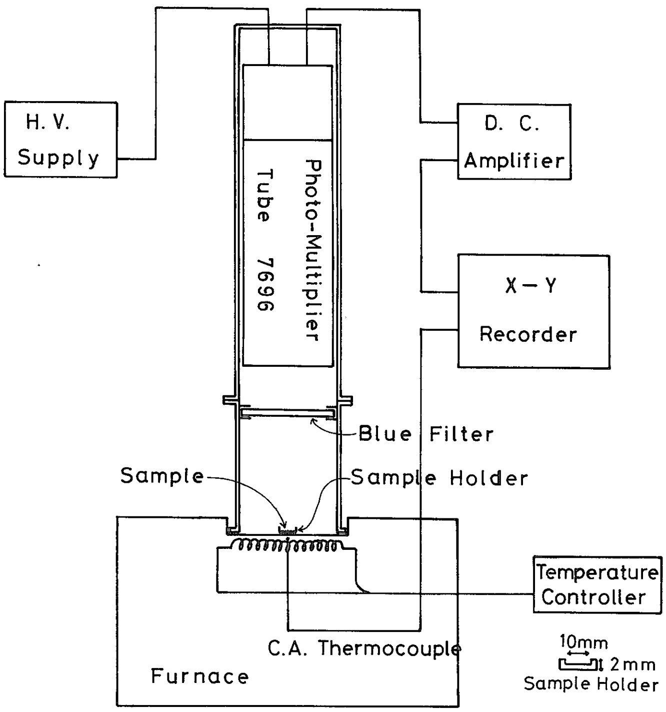


Fig. 2 TL measuring equipment.

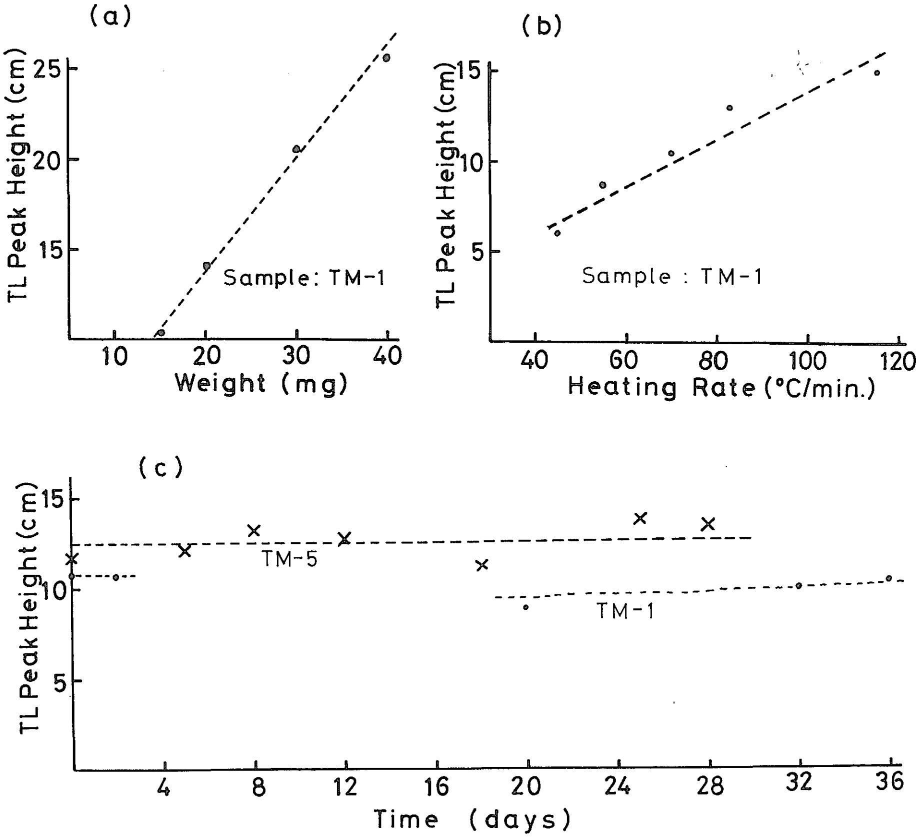
60–150, 150–200, under 200 mesh fraction (from which the fine fraction was removed by washing), and the finest fraction (which was gathered from the washing water) were 1, 0.88, 0.56, 0.50 and 0.32, respectively.

The TL peak heights are proportionally changed with the sample weight (Fig. 3-a). So, the sample was precisely weighted. The heating rate was an important factor to control the TL intensity (Fig. 3-b). To compensate this factor, heating rate was always monitored and the TL intensity was calibrated based on this figure.

In this experiment, there was no fading effect (Fig. 3-c) in peak height. However, new TL peak appeared around 370°C as a fading effect (Fig. 4). The grinding effect was observed as a small peak formation around 370°C (Fig. 4). However, it was not important because its temperature differs from that of main natural TL peak around 320°C.

For the determination of the susceptibility of samples to radiation, all samples were exposed to the X-ray source (Cukα, 40 kV, 50 mA). However, the dose rate was not determined.

The U, Th and K_2O contents of whole rock were analyzed by γ-spectroscopic method. The dose rate of these elements which was reported by BELL (1976) is shown in Table 1. In this experiment, α-ray contribution was neglected because the diameter of quartz phenocryst was much bigger than the affecting range of the alpha particles.

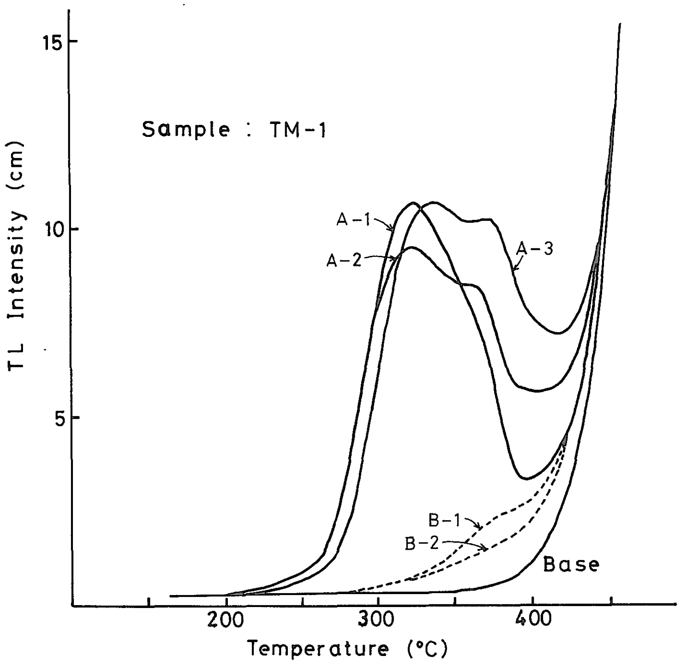


(a) Weight variation
(b) Change of heating rate
(c) Fading effect

Fig. 3 The results for some TL measurements.

Table 1 Radiation dose rate of U, Th, K. (Bell, 1976)

Element	Content	Radioactive Ray	Radiation Dose (rad/y)		
U	1 ppm	α	0.2783	0.3056 (total)	0.0273 (α=0)
		β	0.0146		
		γ	0.0127		
Th	1 ppm	α	0.0740	0.0819 (total)	0.0079 (α=0)
		β	0.0029		
		γ	0.0050		
K	1 % (K_2O)	α	0.0682	0.0887 (total)	—
		β	0.0205		



(A-1) Just after crushing (A-2) After 12 days (A-3) After 36 days (B-1) Kept at room temperature
(B-2) Kept at 70°C, 10 minutes both after 2 minutes grinding of heated sample

Fig. 4 Change of TL pattern by fading and grinding.

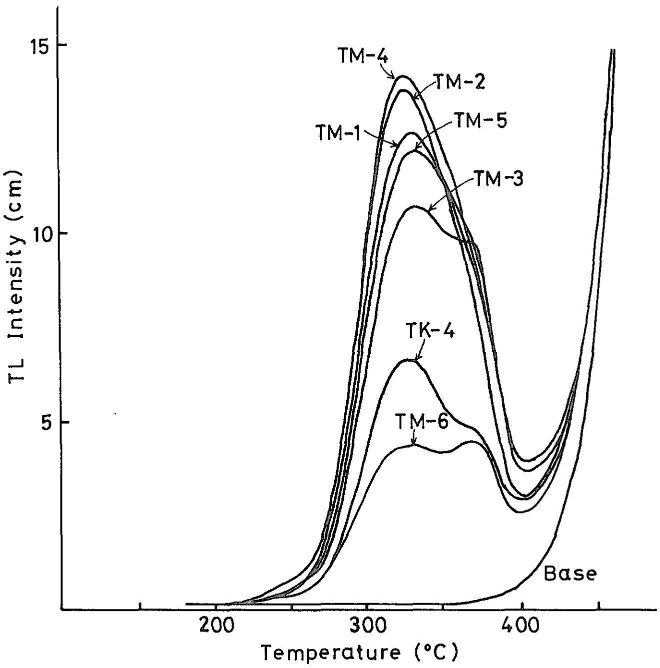


Fig. 5 Natural TL glow curves for Tamagawa welded tuff.

3. Experimental Results

Natural and artificial (X-ray exposed) TL glow curves of seven samples were shown in Figs. 5 and 6, respectively. The natural TL glow curves of four samples had a sharp single peak around 320 °C and the other three had the doublet peaks around 320 and 370 °C. The latter two samples were taken from altered and weathered rocks.

The artificial TL glow curve had a more complicated pattern than a natural one and it is difficult to estimate the 320 °C peak height. So, it was excluded from the calculation of specific TL intensity.

All samples were measured three times and the results were averaged. The variations of the natural TL peak height for each sample were not so large.

Table 2 is the results of U, Th and K_2O analyses and calculated dose rate values.

The TL of about 30 altered samples were also measured. The TL patterns of these samples were the same as that of TM–6 shown in Fig. 5.

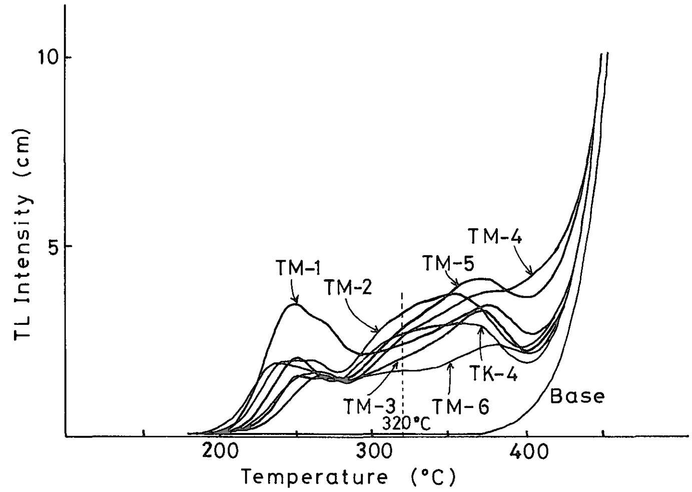


Fig. 6 Artificial TL glow curves for Tamagawa welded tuff (X-ray radiated).

Table 2 U, Th, K_2O contents of Tamagawa welded tuff. (Analyst: Hiroshi KANAYA)

No.	Sample No.	Sample Locality	U (ppm)	Th (ppm)	K_2O (%)	Total Radiation Dose (rad/y)[1]
1	TM- 1	Hashiba	1.2±0.1	4.0±0.2	1.48±0.03	0.1957
2	TM- 2	Kamihinokinai	2.2±0.1	6.8±0.3	2.37±0.03	0.3240
3	TK- 4	Yanagisawa-rindo	0.8±0.1	2.7±0.2	1.11±0.03	0.1416
4	TM- 3	Obonai	1.2±0.1	4.8±0.2	1.62±0.03	0.2144
5	TM- 4	Bunamori-rindo	1.4±0.1	4.7±0.2	1.48±0.03	0.2066
6	TM- 5	Komatakyo	1.3±0.1	4.4±0.2	1.55±0.03	0.2078
7	TM- 6	Gojumagari	1.3±0.1	4.0±0.2	2.13±0.03	0.2560
8	TK- 8	Akagawaonsen	1.5±0.1	5.6±0.2	4.46±0.03	0.4808
9	TK-12	Yunomata-zawa	2.0±0.1	7.1±0.3	4.21±0.03	0.4841

1) Total radiation dose are calculated as the α-radiation dose equals to zero.

Table 3 Summary of TL measuring results.

No.	Sample No.	F. T. Age(m.y.)	TL Peak Height (cm) 1	2	3	(A) Mean	(B) Total Radiation Dose (rad/y)	Specific TL (A/B)	Remarks
1	TM- 1	1.1	11.7	12.5	11.7	11.97	0.1957	61.17	
2	TM- 2	1.2	13.2	13.6	13.3	13.37	0.3240	41.27	
3	TK- 4	1.2	6.5	6.5	6.3	6.43	0.1416	45.41	
4	TM- 3	1.7	10.6	10.9	10.5	10.67	0.2144	49.77	Weekly weathered?
5	TM- 4	1.7	14.0	13.0	14.6	13.87	0.2066	67.13	
6	TM- 5	1.8	11.5	11.6	12.0	11.70	0.2078	56.30	
7	TM- 6	2.0	4.5	4.2	4.3	4.33	0.2560	16.91	Altered rock
8	TK- 8	—	2.2	2.0	2.2	2.13	0.4808	4.43	〃
9	TK-12	—	6.6	6.6	6.1	6.43	0.4841	13.28	〃

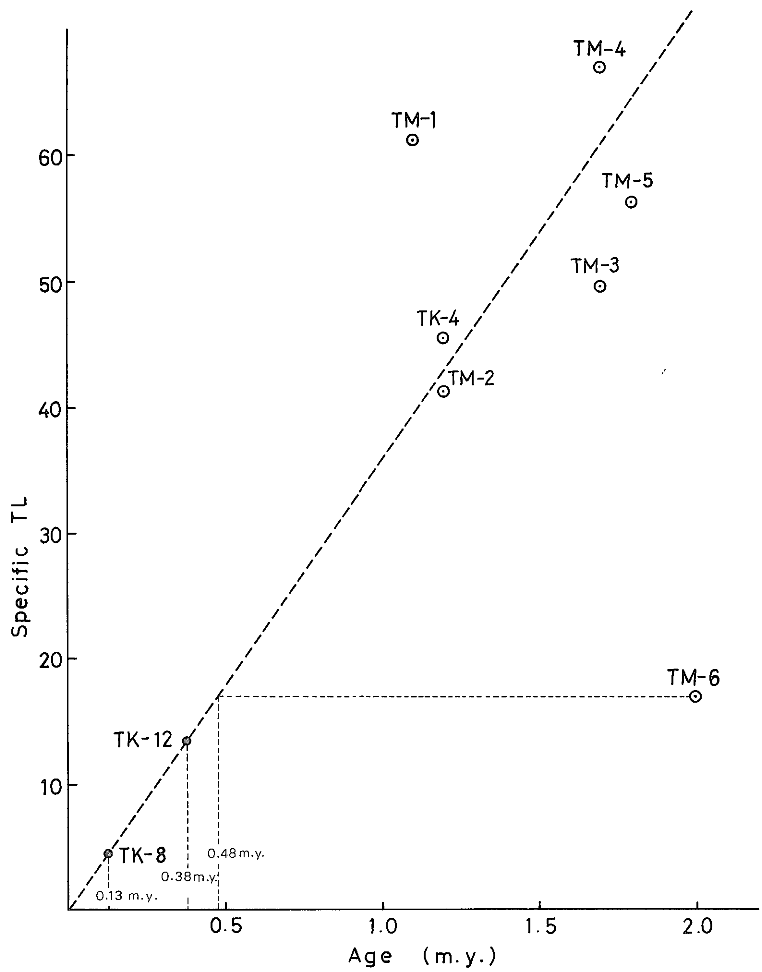


Fig. 7 The relation between specific TL intensity and FT age.

4. Discussion of the Results

Table 3 is the summary of measurements. In this paper, absolute age was not determined by the lack of quantitative dosimetry data. For the check of propriety of quartz TL dating, specific TL intensities and fission-track (FT) ages which were reported by TAMANYU and SUTO (1978) were compared. There is the linear correlation between the FT ages and specific TL for five samples (Fig. 7). It is not clear why TM–1 deviates from the linear line of Fig. 7. There needs more experiment to solve the problem because the both FT and TL methods have the possibility of errors.

The sample taken from altered rock (TM–6) had a low peak height compared to the expected one by its FT age. It is considered that the TL was discharged by the alteration and restored after the alteration. If it is true, the TL age of altered sample corresponds to the alteration age. Based on Fig. 7, the alteration ages were assigned to 0.48, 0.13 and 0.38 m.y. for TM–6, TK–8 and TK–12, respectively.

This idea was applied to the samples taken from the alteration area shown in Fig. 1. For the estimation of the temperature effect during alteration to TL patterns of quartz crystals, the sample was heated from 150 to 300 °C. The change of TL pattern was shown in Fig. 8. This shows that the 320 °C peak shifts to 370 °C when the heating temperature exceeds 230 °C. So, the 320 °C TL peak would become zero when the alteration temperature exceeds 230 °C. At present, the sample taken from the younger alteration zone has a low TL peak if the other conditions are the same.

For the estimation of the alteration ages, the TL of about 30 samples were measured and the results were plotted on Fig. 9. The simplified alteration mineral zoning was compiled from the study of KIMBARA *et al.* (1978). In this case, no corrections using the contents of radioactive elements to TL peak heights were applied. So, they do not show the strict ages and the results are simply indicated

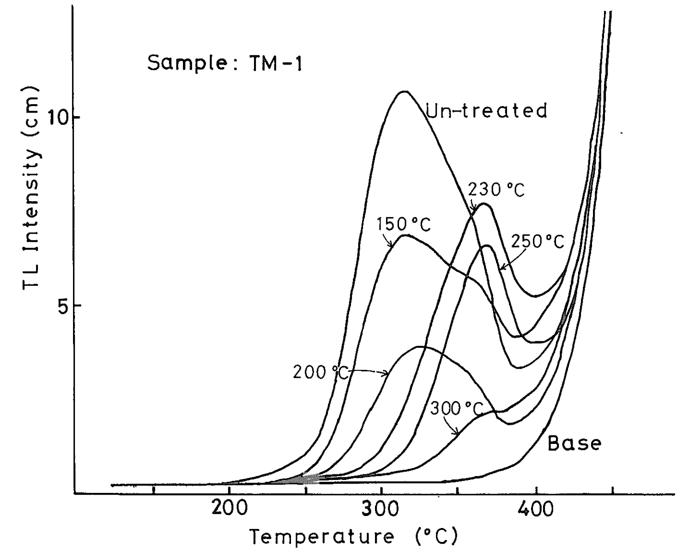
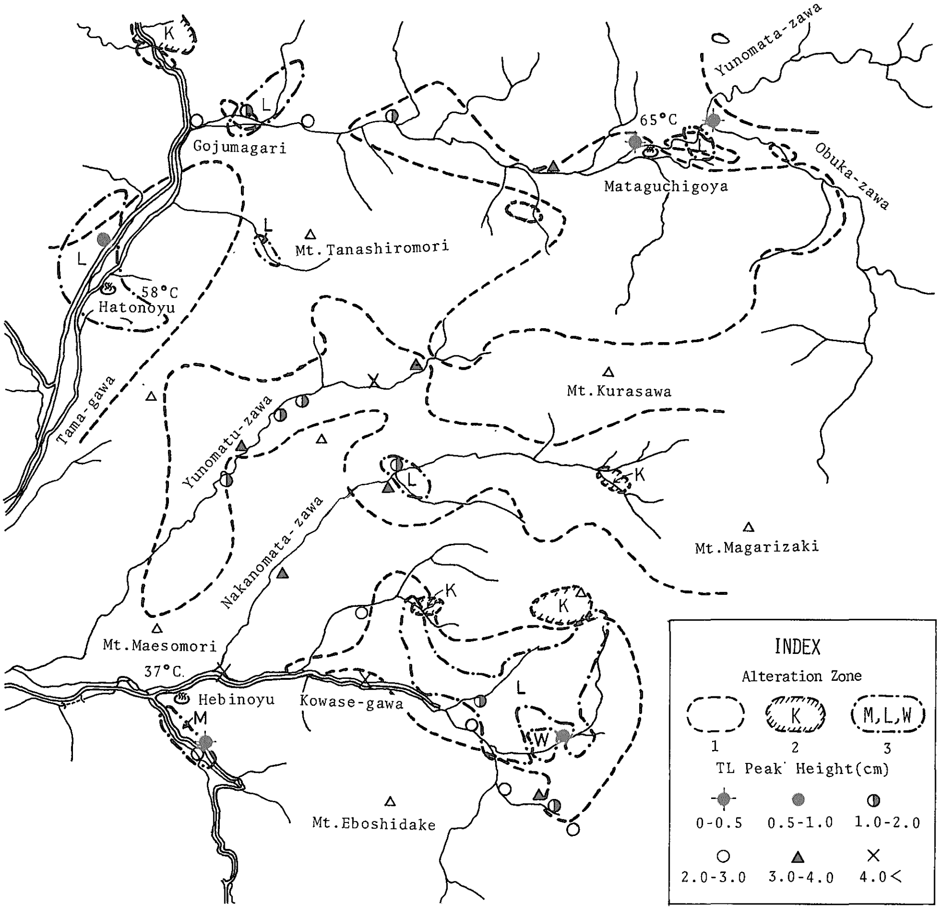


Fig. 8 Change of TL glow curves by heating.



Alteration zone:
1. Argillized zone (without kaolin)
2. Argillized zone (with kaolin)
3. Zeolite zone (M; Mordenite, L; Laumontite, W; Wairakite)

Fig. 9 Alteration zone and TL intensities (Alteration was studied by KIMBARA *et al.*, 1978).

as the non-corrected TL peak heights in Fig. 9. However, roughly estimated results are valuable for the order estimation of altered ages because the correcting coefficient by radioactive elements are about one to three as shown in Table 2. If these roughly estimated values are adopted to above alteration area, it is very interesting that the suspected younger alteration areas delineated by the rough TL dating coincide with the hot spring areas or wairakite bearing area.

5. Conclusions

The TL dating method for quartz provides a good result for the age determination of original rock and would be applicable for alteration age determination. In this study, the quantitative TL assessment for artificially radiated sample was not carried out. However, the assessment of the radiation dose rate for natural minerals and artificial exposure is the important factor to determine the absolute TL ages. Also, the annealing effect may be the key to the precise TL age determination for both original and altered rocks because the TL are very sensitive to their suffered temperature conditions.

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サーモルミネッセンスによる変質年代決定法の基礎的研究

高島　勲

要　旨

秋田県八幡平地域に分布する玉川溶結凝灰岩中の石英のサーモルミネッセンス（ＴＬ）強度とフィッショントラックＦＴ年代がかなり良い対応を示している．しかし，変質を受けた岩石中の石英のＴＬはＦＴ年代から予想される強度より著しく低くなっている．このことは，変質によってそれまでに蓄積し

たＴＬを放出し，変質が終了後，新たにＴＬの蓄積をはじめたことを示すものと思われる．このような仮定にもとづいて，前記地域の変質溶結凝灰岩の変質年代を求めれば，0.13，0.38，0.48 m. y. という値が得られた．さらに，このような方法を同地域の変質帯について広く適用したところ，現在の熱徴候のあるところおよびワイラカイトを含む変質帯が若い変質年代を示すことが明らかになった．