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EVALUATION OF COOLING HISTORY OF THE QUATERNARY TAKIDANI PLUTON USING THERMOLUMINESCENCE TECHNIQUE

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ABSTRACT

The characteristics of thermoluminescence (TL) and Electron Spin Resonance (ESR) of quartz in the Takidani pluton were investigated to evaluate the cooling history of a Quaternary granitoid in an active volcanic chain. The Takidani pluton is composed mainly of granite and granodiorite, and shows a reverse zonal structure in terms of its whole-rock chemical composition. Paleodose values obtained using TL and the ESR-Ti center were similar. However, values for the ESR-Al center are lower. These results indicate that the thermal stability of the ESR-Al center is more unstable than that of the TL and ESR-Ti center. The TL age was strongly dependent on thermal processes that have effected the sample. The closure temperature is considered to be approximately 70°C, and the TL age indicates the elapsed time since cooling below the closure temperature. TL ages of three samples range from 91 to 152 ka. The cooling rate at an early stage in the history of the granitoid, including the solidification process, is considered to be approximately 600°C per million years. During the later stages accompanying uplift, the rate may have decreased to approximately 200°C per million years. TL and ESR techniques are a potentially useful new research tool for evaluating the cooling history of plutonic rocks, particularly the timing of exposure.

1. INTRODUCTION

Thermoluminescence (TL) and Electron Spin Resonance (ESR) phenomena of geological materials have been used as a dating tool and provide an effective technique for obtaining the "absolute age" of volcanic and pyroclastic rocks of the Pliocene-Pleistocene (Takashima, 1985). However, it is difficult to detect the TL of quartz in granitic rocks due to its very weak luminescence. The reasons for the unsuitable luminescence of quartz in granitic rocks are unclear, but most evidence points to the hollocrystalline nature of the rock, and the relatively low cooling rate of plutonic masses compared to volcanic and pyroclastic rocks.

The Takidani granitoid (TG), which is the youngest exposed granitoid pluton on Earth (Harayama, 1992), crops out in the Japan Alps. The TG forms a volcano-plutonic complex of Plio-Pleistocene age. Thermochronological investigations have been performed by Harayama (1994). Discrepancies between ages determined by various dating methods enable the thermal history, and hence, the cooling rate of the pluton, to be deduced. Whole-rock and mineral ages, using Rb-Sr and K-Ar techniques, are useful indicators of cooling rates in the region up to 350°C. The fission track method (FT) provides cooling ages around 200°C. Effective techniques, which provide useful information about temperatures and ages at less than 200°C, has yet to be developed.

In principle, once the rate of irradiation from radioactive nuclides and surrounding rocks is established, and the rate of thermal release of TL during rock irradiation is shown to be negligible, then the age determined by TL is the absolute age of the material since its formation or last heating event. In other words, the TL age indicates the elapsed time since the last thermal event and/or thermal history of the rock coupled with temperature and cooling rate. The TL of rock-forming minerals strongly reflect their thermal history because TL is essentially a thermal phenomena. Recently, the TL technique has been shown to have an application in evaluating geothermal activity (Tsuchiya et al., 1994, 1998, Nambu et al., 1996). Tsuchiya et al. (2000) demonstrate that TL of quartz in pyroclastic and intrusive rocks is a useful method for evaluating the temperature effects and subterranean heat flow patterns of geothermal activity in the Kakkonda geothermal field.

The TG was exposed by rapid uplift in the Quaternary, and its cooling history is considered to be simple compared with other granitoids in Japan, largely due to its young age.

The main purpose of this study is to describe the TL behavior of the Quaternary granitoid and to present a preliminary interpretation of the thermal history of the pluton using TL and ESR techniques. Our results suggest that TL and ESR offer a new exploration technique for evaluating the cooling history of granite-hosted geothermal reservoirs.

2. GEOLOGICAL SETTING

Figure 1 shows a generalized geological map around the Hotaka mountains, which forms the main ridge of the Japan Alps. Several active volcanoes form the Norikura Volcanic Chain, which extends along the Japan Alps. An outline of the geology and petrological characteristics of the TG are described in Bando and Tsuchiya (2000). The TG was divided into six lithofacies, consisting of porphyritic-granodiorite, porphyritic granite, biotite-hornblende granodiorite, hornblende-biotite granodiorite, biotite-hornblende granite, and biotite granite, respectively. The TG shows features of a reversely zoned pluton. The form of the pluton was deduced on the basis of the thermal fracture network (Kano and Tsuchiya, 2000; Sekine and Tsuchiya, 2000).

According to Harayama (1994), geochronological data for the TG are as follows: the zircon fission track age is 0.80±0.02 Ma; and K-Ar ages on biotite and K-feldspar range from 1.2 to 1.1 Ma. Detailed investigations on the geochronology are is progress and new age information will be published in due course.

3. EXPERIMENTS

All samples used in this study were collected from surface outcrops. After crushing the rock samples, quartz grains were separated by hand picking. The quartz grains were crushed and the 74-250 μm fraction was obtained by sieving. The quartz particles were then treated with HCl, followed by a 15 minute etch in 40% HF to reduce the effect of the alpha dose and to dissolve any feldspar contaminant.

Thermoluminescence: 30 mg of quartz grains were laid on a platinum plate heating element. Light pulses were detected with a photomultiplier tube coupled to an interference filter with maximum transmittance at 620 nm and half height width of 20 nm. A linear heating rate of 20°C/min was applied from room temperature to 350°C.

Electron Spin Resonance: 100 mg of quartz were used for ESR analysis. ESR spectra were obtained with a commercial ESR spectrometer (JEOL-RE2X). The field modulation width was 0.079 mT with a frequency of 100 kHz for detecting E' centers in quartz at room temperature. Signals of Al and Ti centers were observed with a microwave power 1.0 mW at liquid nitrogen temperature (77K), where field modulation width was 0.05 mT at a frequency of 100 kHz.

Artificial irradiation: After measurement of TL and ESR, the sample was annealed at 320°C for one hour in air to release remanent trapped electrons. The samples were gamma irradiated by a ^{60}Co radiation source. The absorbed dose was evaluated to be air equivalent and the maximum dose was 753 Gy, with a dose rate of 0.309 Ckg-1/h for 72 hours.

Annual dose: TLDs ($CaSO_4$:Tm) had been buried in the rock masses for over two months. In addition, chemical analyses of radioactive elements including K, U and Th were undertaken to calculate the annual dose. In this study, the annual dose of cosmic-ray was determined to be 0.2 mGy/y.

4. RESULTS

Representative thermoluminescence of quartz, including natural and artificial glow curves, are shown in Figure 2. The background component of the thermoluminescence signal, which is approximated by the black-body radiation of the platinum plate, is subtracted from the measured glow curve.
The sample No. 10 was collected from biotite-hornblende granodiorite, No. 20 was biotite granite, and No. 38Q was from a quartz vein in hornblende-biotite granite.

The intensity of natural thermoluminescence, NTL, was found to increase at approximately 160°C, and the apparent maximum temperature of the glow curve appeared from 330°C. Above 400°C, thermoluminescence of the sample decreased to zero while the background, caused by the incandescence of the platinum plate, rapidly increased. The initial rise in temperature is almost the same as that of quartz in the Tamagawa Welded Tuffs distributed in and around the Kakkonda geothermal field (Takashima, 1985; Tsuchiya *et al*., 2000), but the apparent maximum temperature is shifted to a higher value. (the maximum temperature of the Tamagawa Welded Tuffs ranges from 270°C to 290°C under the same experimental conditions).

After irradiation, the artificial thermoluminescence, ATL, is elevated at low temperature (<100°C). The apparent maximum temperature of ATL was the same as the NTL, with an additional unstable peak observed around 180°C. In this study, thermoluminescence intensity was defined as the maximum peak height.

Figure 3 shows examples of ESR spectra of quartz (No. 38Q) obtained at liquid nitrogen temperature. The signals of Al and Ti centers are identified. The signal intensity, used for age evaluation, was defined as peak-to-peak height at g=2.018 for the Al center; the Ti center had a peak position below the baseline at g=1.913 (Toyada *et al*., 1995).

Growth curves of thermoluminescence after irradiation are shown in Figure 4. The TL intensity increases nonlinearly with increasing irradiation dose, and it approaches a saturation level in the range of 753 Gy. The TL intensity of sample No. 20 is approximately three times greater than that of No. 38Q. Clearly, the saturation level does depend on the sample. The capacity of trapped electrons, indicating the saturation level, reflects crystal defects and impurities in the rock. The discrepancy of saturation values also indicates differences in the formation condition of quartz.

The paleodose of the sample could be obtained by using the growth curve. The paleodose of each sample was less than approximately one third of the saturation level of each sample. ESR growth curves for both the Al and Ti centers were obtained. Some growth curves of ESR were evaluated by linear functions, with high correlation factors, but others were approximated by a nonlinear function, similar to that for TL growth curves.

Figure 5 represents the paleodose values obtained using TL and ESR-Al and ESR-Ti centers, respectively. The paleodoses of the TL and ESR-Ti center are similar for each sample, although the paleodose of the ESR-Al center differs from that of the TL and ESR-Ti center for samples No. 20 and 38Q. Overall, the three samples show different paleodoses, which range from 172 Gy to 281 Gy in TL.

Using the annual dose calculated from the contents of K, Th and U, the preliminary TL ages are as follows: No. 10 is 104 ka, No. 20 is 152 ka, and No. 38Q is 91 ka. It is necessary to evaluate beta-ray attenuation, which depends on the quartz grain size, to obtain an accurate annual dose (Mejdahl, 1979). Sample No. 38Q, which is a quartz vein 150 mm in thickness, contains no radioactive nuclides. In this case, it may be argued that the gamma-ray dose of the surrounding rocks can be used for the annual dose of the quartz vein. Thus, the TL age of No. 38Q is more accurate than that of the other two samples.

5. DISCUSSION

5.1 Thermal stability of TL

The theoretical treatment for thermoluminescence was given by Randall and Wilkins (1945), who assumed no retrapping and a TL intensity proportional to the rate of charge of the concentration of trapped electrons. The fundamental equation of the thermoluminescence mechanism, including storage and thermally stimulated processes, is described as follows (Tsuchiya et al., 1998):

$$I(T)=\frac{dn_1}{dt}=-n_1 s\exp\left(-\frac{E}{kT}\right)+\lambda r\left(N-n_1\right) \quad (1)$$

where I is the TL intensity, s is a "pre-exponential factor", N, n and n_1 are the concentrations of traps in question, of free electrons and of trapped electrons, respectively. E is the activation energy, which is the same as trap depth, T is the absolute temperature, k is Boltzmann's constant, λ is a proportionality constant relating to the recombination probability and irradiation efficiency, and r is the irradiation dose rate. The first term in the right side of equation (1) represents the thermally stimulated process and the second term is the storage process of radiation energy. The concentration of the trapped electrons (n_1) is very low compared with N at the early stage of irradiation after zero set, so the second term effectively becomes λrN. By integration of equation (1), the following equation (2) for n_1 was obtained,

$$n_1=\frac{\lambda rN}{s\exp\left(-\frac{E}{kT}\right)+\lambda r}\left\{1-\exp\left[-\left\{s\exp\left(-\frac{E}{kT}\right)+\lambda r\right\}t\right]\right\} \quad (2)$$

Figure 6 shows the relationship between heating duration and thermoluminescence intensity. With increasing temperature, the TL intensity is depleted. The TL mechanism of solid materials is composed of thermally stimulated and radiation storage processes. In other words, both are compensated at a temperature 70°C. According to the preliminary calculation in Figure 6, the TL age indicates the elapsed time after cooling below approximately 70 °C, which is considered to be the closure temperature for TL dating.

5.2 TL and ESR

At least in principle, paleodose values obtained using TL and ESR techniques should be equal to each other. However, Figure 5 shows that further consideration is required to understand the differences between mutual paleodoses. The TL and ESR-Ti center values could be treated equivalently in spite of slight differences, but the paleodose for the ESR-Al center for Nos. 20 and 38Q, is much lower than the TL and ESR-Ti center values for the same samples. Toyoda and Ikeya (1991) described the thermal stability of paramagnetic defect and impurity centers in quartz for ESR dating. According to their results, the closure temperature at which the “ESR clock” starts was estimated to be 78°C and 31°C for Al and Ti centers respectively, with a cooling rate of approximately 10°C / My. In other words, the ESR-Al center was more stable compared to the ESR-Ti center. However, they also showed that the thermal stability of the Al and Ti centers depends on the nature of the sample. For quartz in the TG, the paleodose obtained using the ESR-Al center is lower than that using the ESR-Ti center.

Further studies will be required to evaluate the accurate closure temperatures of TL and ESR signals of different centers, including kinetic equations of thermally stimulated and radiation storage processes under different cooling rate conditions. TL and ESR behaviors, however, could be possible candidates for evaluating the thermal history of igneous rocks.

5.3 Cooling history

The cooling history of the TG, including K-Ar for biotite, fission track for zircon and TL for quartz, is summarized in Figure 7. Harayama (1994) estimated the temperature of magma crystallization for the TG at approximately 760°C, and an upper limit of the solidification age to be 1.76 Ma, (unpublished data by Harayama, 1998) based on the fission track age of Nyukawa pyrocrastic rocks covering the TG. Quaternary geological samples, particularly igneous rocks, contain excess argon, so careful treatment is required to obtain accurate age data using the K-Ar method. In the near future, age data, using K-Ar and FT techniques, will be revised. However, the final age of the cooling process may be defined by the TL ages.

The preliminary age of TL, which is the elapsed time since cooling below 70°C, ranges from 91 ka to 152 ka. Based on closure temperatures and K-Ar and FT ages, the cooling rate during the early stages of magma plumbing, solidification and emplacement is estimated to be approximately 600°C/My. During the later stage, accompanying uplift, the rate may have decreased to approximately 200°C/My. Rapid uplift occurred in the Japan Alps region during the Quaternary, and our results give a new insight into the formation mechanisms of plutonic rocks in volcanic chains in the island arc plate boundary setting.

Bando and Tsuchiya (2000) have described the petrochemistry and estimated emplacement pressures of various lithofacies of the TG, and Harayama (1994) has mentioned differences in cooling rates within the pluton. Paleodose values of quartz differ in the pluton, suggesting hydrothermal activity and differential movement during uplift. These results indicate that thermoluminescence behavior is a potentially useful new exploration technique for evaluating the thermal history and heat source in neo-granitoids, particularly in active/fossil geothermal systems.

6. CONCLUSIONS

Thermoluminescence and electron spin resonance behaviors of quartz in the Takidani granite were investigated. The following results were obtained.

The thermoluminescence age is strongly dependent on the thermal history of the samples. Its closure temperature is considered to be approximately 70°C, and the TL age indicates the elapsed time since cooling below the closure temperature. Three selected samples suggest that TL ages of the TG range from 91 ka, to 152 ka.

Paleodose values obtained using TL and the ESR-Ti center were almost the same. However, values of the ESR-Al center are lower. These results indicate that the thermal stability of the ESR-Al center is weaker than that of TL and the ESR-Ti center. By coupling TL and ESR methods, including Al and Ti centers of ESR signals, the thermal history of a pluton can be evaluated.

The cooling rate during the early stage of magma plumbing, solidification and emplacement of the TG is estimated to be approximately 600°C/My, and at a later stage, during uplift, might be approximately 200°C/My.

Thermoluminescence behavior is a new and potentially very useful exploration technique for evaluating the thermal history and heat source of Quaternary granitoids in the active geothermal fields.

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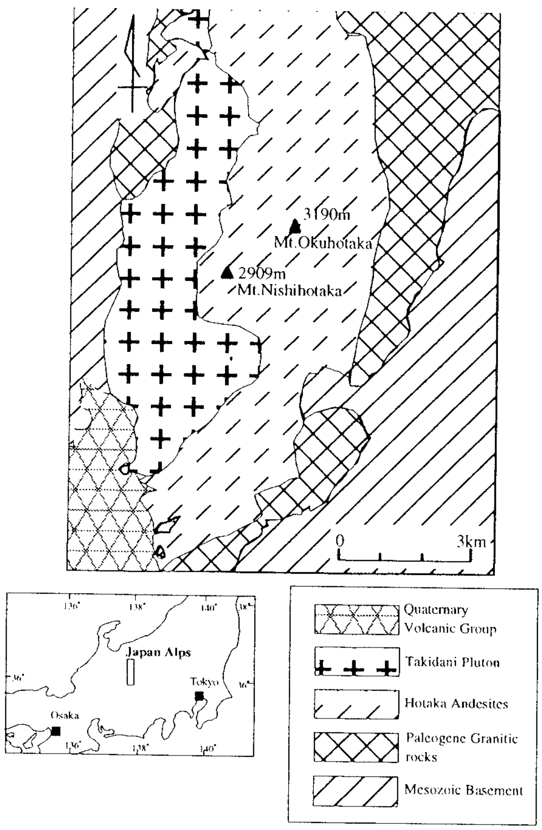

Figure 1 Generalized geological map of the Takidani pluton (simplified after Harayama, 1992 and Bando and Tsuchiya, 2000).

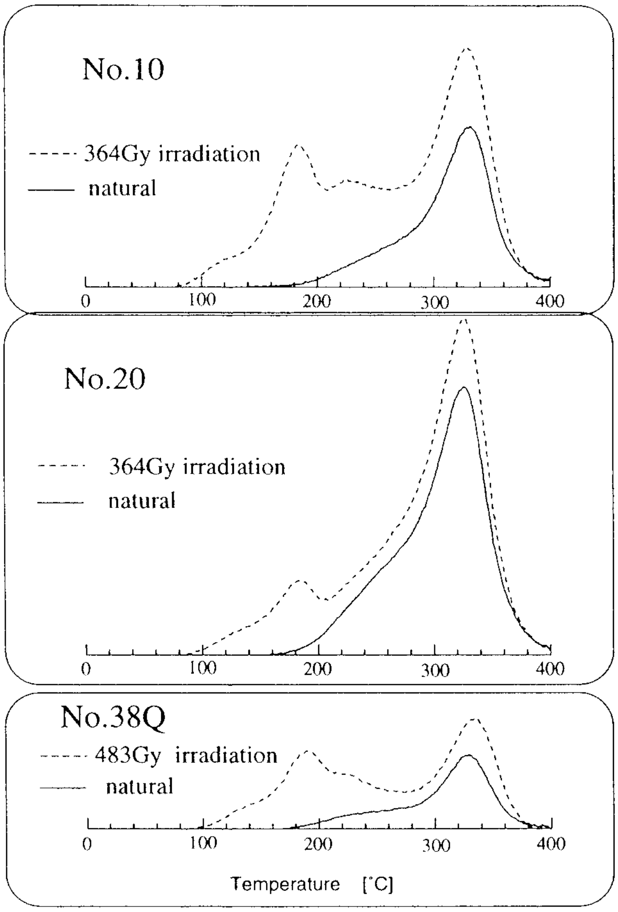

Figure 2 Examples of thermoluminescence glow curves.

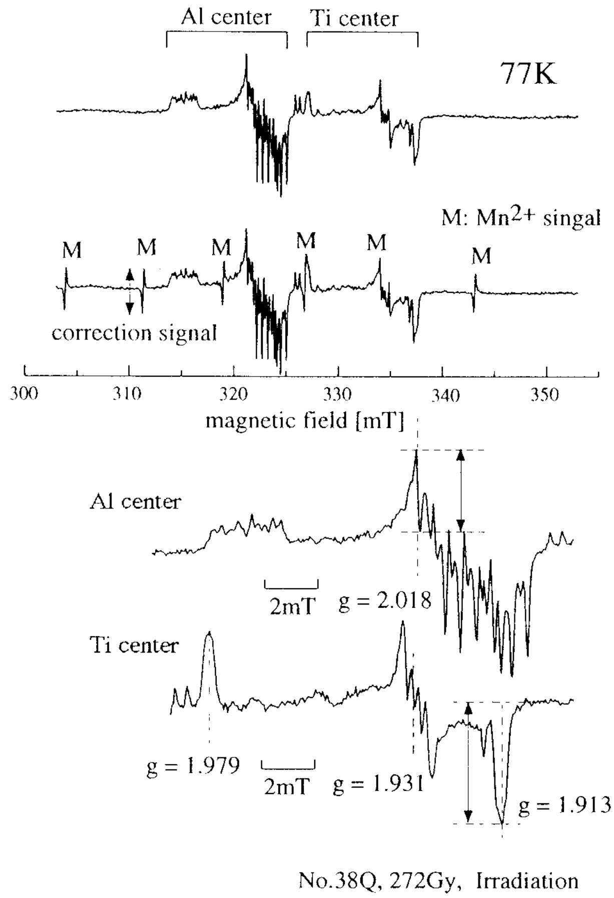

Figure 3 Examples of ESR spectra.

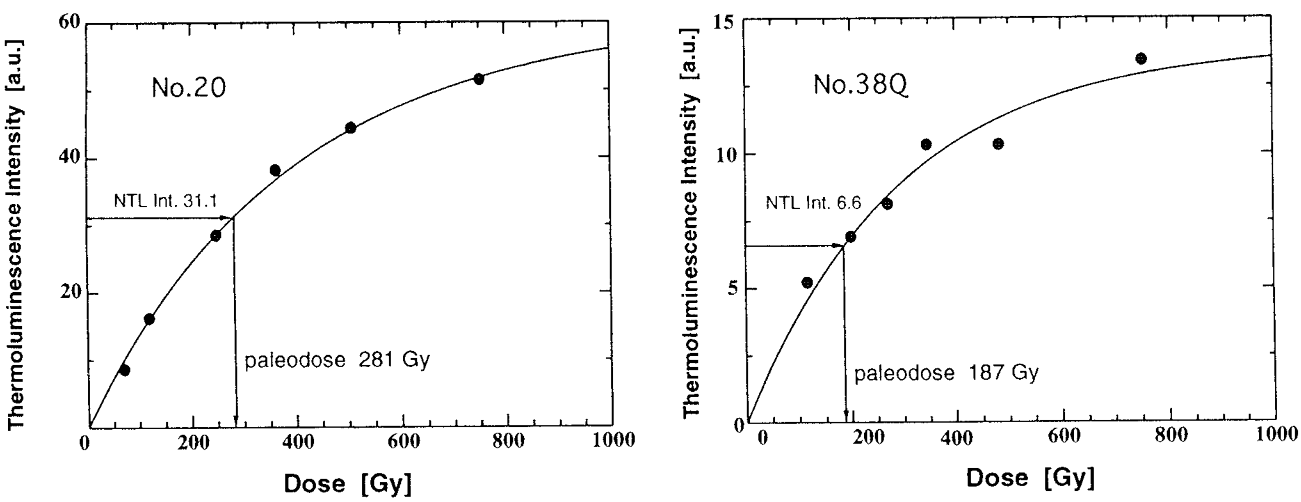

Figure 4 Thermoluminescence growth curves.

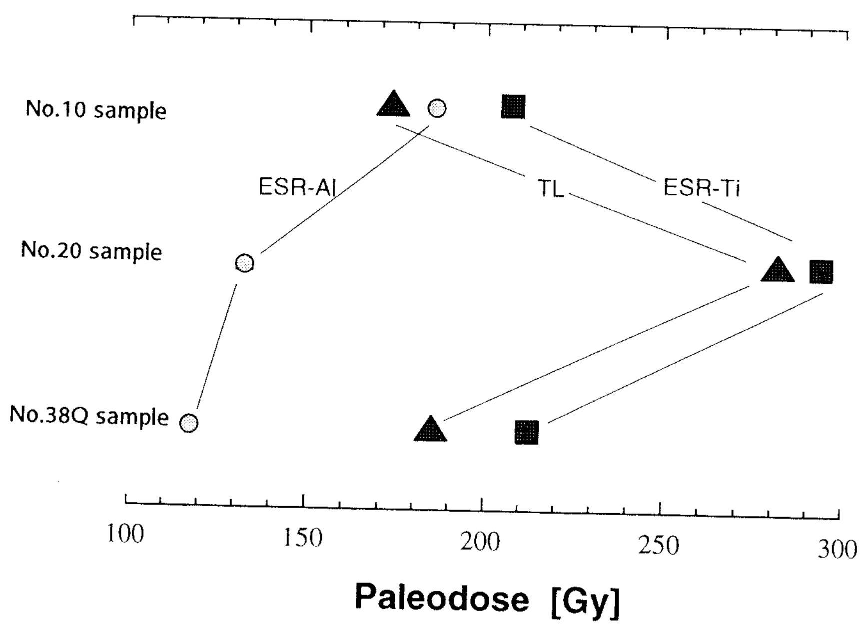

Figure 5 Paleodose of selected three samples of Nos. 10, 20 and 38Q obtained using TL, ESR-Ti and ESR-Al centers.

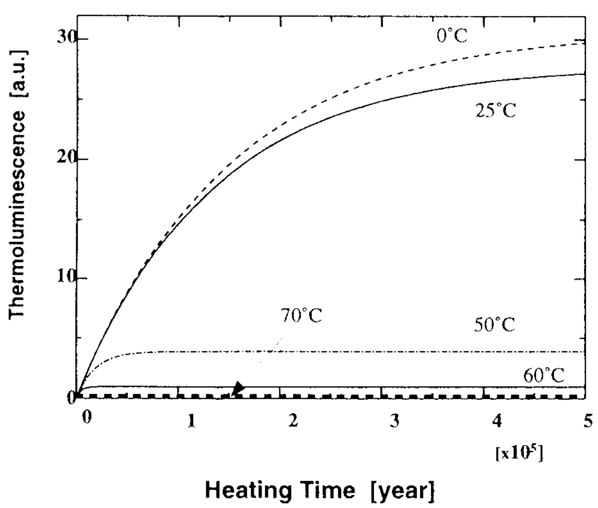

Figure 6 Relationship between heating time and thermoluminescence intensity calculated from equation (2).

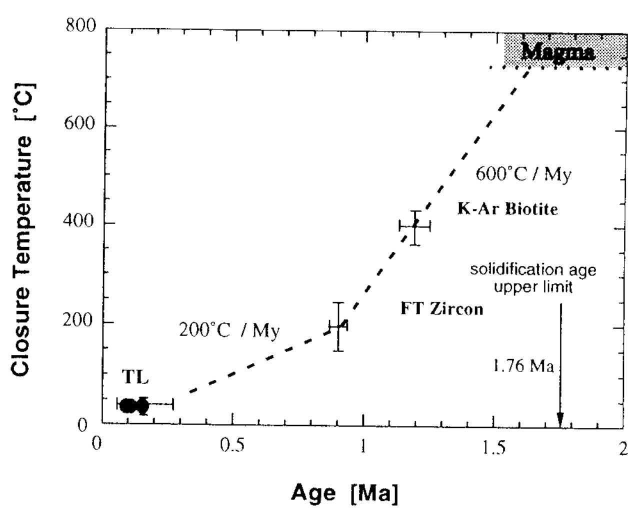

Figure 7 Cooling history of the Takidani pluton. (K-Ar and FT ages are quoted after Harayama, 1994)