APPLICATION OF THERMOLUMINESCENCE TECHNIQUE TO GEOTHERMAL EXPLORATION

N. TSUCHIYA & K. NAKATSUKA

Department of Geoscience and Technology, Tohoku University, Sendai, Japan

SUMMARY – Thermoluminescence (TL) techniques are well known as a dating method for geological and archaeological materials younger than approximately 1 Ma. Thermoluminescence is a thermally derived phenomena of crystals, with the character of thermoluminescence behaviour of minerals and /or rocks in geothermal area being affected by geothermal activity, in other words, by natural heating. Possibilities of thermoluminescence techniques for geothermal exploration are summarized, and a new approach for geological applications of thermoluminescence is described.

1. INTRODUCTION

Thermoluminescence (TL) phenomena have been effectively applied to date archaeological and geological materials that are younger than 1Ma. Here, the age is defmed as natural TL emission, which may be converted into paleo-dose, divided by annual dose (Aitken, 1985; McKeever, 1985). In principle, once the rate of irradiation from the radioactive nuclides and surrounding rocks is established and the rate of thermal release of the 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 its last heating event (Readhead, 1988; Ypma and Hochman, 1991). However, several hypothesis and/or premise, such as a constant dose rate from surrounding rocks, linearity of TL for unit dose and reset of pre-existing TL are required, in order to obtain an accurate and absolute 'age' of geological materials.

The most important precondition is negligible thermal emission by natural heating *or* constant thermal release of TL during sample lifetime. Strictly, the 'age' is the length of time that the sample has been subjected to irradiation following its last heating. Natural thermal events and thermal manifestations are commonly recognized in the geothermal setting, and it is difficult to obtain an accurate 'age' of rocks from geothermal areas. The disadvantage for TL dating, namely detrimental natural thermal effects, actually provides the possibility for applying TL to geothermal exploration techniques, because TL is evidently a thermally derived phenomena.

The TL phenomena of rock-forming minerals reflect their lattice defects and chemical impurities, so the origin and formation process of such minerals could be estimated by change in their TL characteristics. Representative physical and chemical key factors of TL phenomena are thermal effects, chemical impurities and structural defects. The thermal factor suggests potential for TL to be a geothermal sensor, and chemical factors such as inferred impurities and crystal defects highlight possible applications as a geochemical sensor.

In this paper, we describe examples of the application of TL for the evaluation of geothermal activity, and propose other possibilities for TL resolving a variety of geochemical processes, e.g. hydrothermal alteration and mass transport in rock matrix.

2. THERMOLUMINESCENCE

2.1 Background

Quartz, feldspar and calcite are well known as typical rock-forming minerals, and show measurable TL emission. In particular, quartz is often applied for dating, due to its chemical stability for emission. Quartz is also a suitable mineral for evaluation of thermal effects in geothermal areas by the TL analysis.

Feldspar may reflect hydrothermal events, because feldspar is easily altered, in comparison to quartz. For the application of TL to understand geochemical processes, feldspar is a suitable mineral.

The majority of TL measurements have been made using photomultiplier tubes (PMT). These devices offer high sensitivity, however, such standard equipment integrate the luminescence signal from the entire sample and provide no indication of spatial variations in luminescence intensity within a given sample. When we measure TL of a selected mineral, we spend much time to separate the mineral from the rock. Duller *et al.* (1997) developed a luminescence imaging system using a highly sensitive CCD camera. These instruments have the advantage of obtaining a luminescence imaging map, by which spatial variations of TL may be evaluated.

Telfer and Walker (1978) described long wavelength emissions of feldspar at 560 nm and

700-780 nm, being unequivocally associated with the presence of Mn^{2+} and Fe^{3+} impurity ions. Kirsh and Townsend (1988) discussed possible energy transfer mechanisms leading to red emission of the feldspar. Holes created by ionising radiation and subsequently mobilized during heating, oxidize to Fe^{2+} to Fe^{3+}, leaving it in an excited state, followed by emission. Spectral information of TL can thus be expected to provide knowledge that may identify diffusion of impurities in rock matrix.

Hareyama *et al.* (1998) described a relatively high natural radioactivity in an alteration halo, compared to the unaltered rock matrix, using the two-dimensional radiation imaging plate detector. These facts indicate enrichment of radioactive nuclides, such as potassium, associated with the secondary minerals and diffusion of hydrothermal fluid, containing various kinds of chemical species, from the altered rock and through the rock matrix.

2.2 Measurement System

Three types of measurement systems for TL have been developed and installed in our laboratory. Each system has different features, and we can cover a wide variation of TL behaviour in the crystal and/or host rock.

TYPE I: One-dimensional TL intensity measurement system: Conventional TL system for obtaining intensity of TL emission from selected samples, using photomultiplier tube (Hamamatsu Photonics Co. R376). Separation of an objective mineral from whole rock is required, but sensitivity is greater than the other types.

TYPE II: Spatial distribution TL measurement system: Spatial variations of TL from rock slices were measured using position detective TL equipment. A schematic illustration of the equipment, which is chiefly constructed from a high sensitive CCD camera with an image intensifier and computer system, is shown in Fig. 1. The panel heater, of 30 x 30 mm^2 area, is made from a SiC platelet-heating element. The CCD camera used is a C4880-92 type, with a S5466 CCD chip (Hamamatsu Phonics Co.) of 24 mm square, which is arranged with an array of 512 x 512 pixels. The camera comprises an image intensifier, a thermo-electric cooling unit and associated camera control unit, that allows the system to be interfaced directly to a computer. Figs. 2 and 3 show the Type II and TYPE III measurement systems.

TYPE 111: Spectral TL measurement system: Using a spectrometer coupled with high sensitive CCD camera, spectral data can be obtained from ultraviolet through visible light. Infrared emission and radiant heat from the heater was excluded with an infrared cut-off filter. The CCD camera with image intensifier (C4569-01, Hamamatsu Photonics Co.) is connected to spectrometer (C5094) with optical fibre.

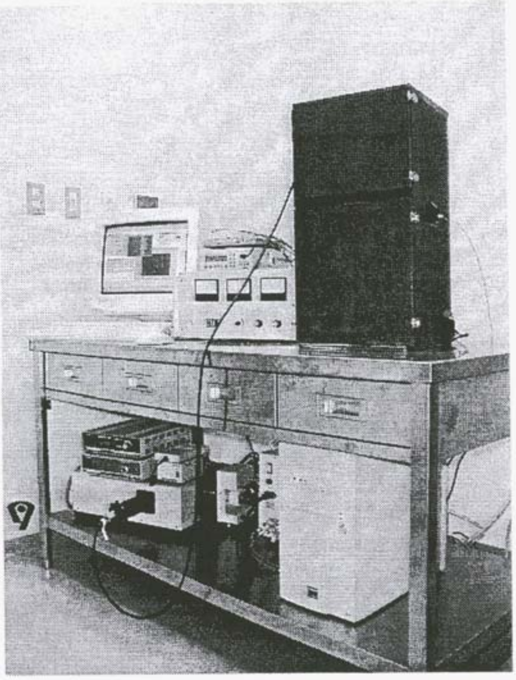

Fig. 1 Overview of TYPE II and TYPE III TL-measurement system.

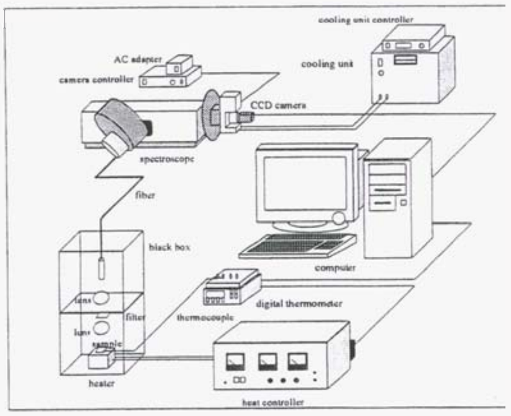

Fig. 2 Schematic block diagram of TYPE II and TYPE III TL-measurement system.

2.3 Sample and measurement conditions

TYPE I system: After crushing the rock samples, quartz grains were separated by hand picking. The quartz grains were crushed and the 74-250 μm fraction of quartz was obtained by sieving. The quartz particles were then treated with HCl to remove any carbonates, followed by a 15 min. etch in 40% HF to reduce the alpha dose and to dissolve contaminating feldspar grains.

Light pulses were detected with PMT 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.** The background component of TL signal was approximated by the black-body radiation of the platinum plate.

TYPE II &TYPE III systems: The **30** x **30** x 1 mm sliced samples were heated from room temperature to 400°C with a heating rate of *20°C* /min. The temperature heterogeneity within the sliced rock sample was less than **10°C.** The exposure interval was 5 seconds for each 10 seconds, so temperature differences during the exposure was less than **2°C.**

3. GLOW-CURVES

Fig. 3 shows representative glow curves of quartz **from** a pyroclastic rock (Tamagawa Welded Tuffs). The intensity of the TL emission increased at ca.180°C, with an apparent maximum temperature of the glow-curve **from 270°C** to **290°C.** TL decreased to zero whilst the background (induced by the incandescence of the black radiation of the heating element) increased rapidly above 350°C. Multiple and discrete trap levels were recognized in quartz (Hashimoto *et al.,* **1993).** Tsuchiya *et al.* **(2000)** carried out deconvolution of TL glow-curve, and calculated three elemental peaks, L, M and H, using adequate **trap** parameters (Fig. **3).**

Examples of natural TL (NTL) glow-curves of quartz in granitic rocks are shown **in** Fig. **4,** coupled with artificial TL glow-curves after irradiation of ^{60}Co gamma-ray. Generally, quartz

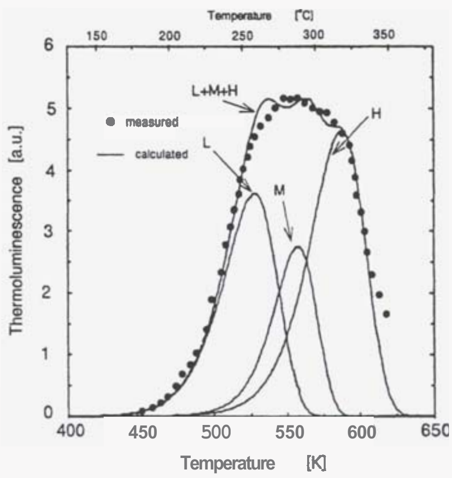

Fig. **3** Representative **TL glow-curve** of **quartz in pyroclastic** rock. L: **Low peak**; M: Medium peak; H: High peak; L+M+H: total **summing** emission (Tsuchiya *et al.,* 2000).

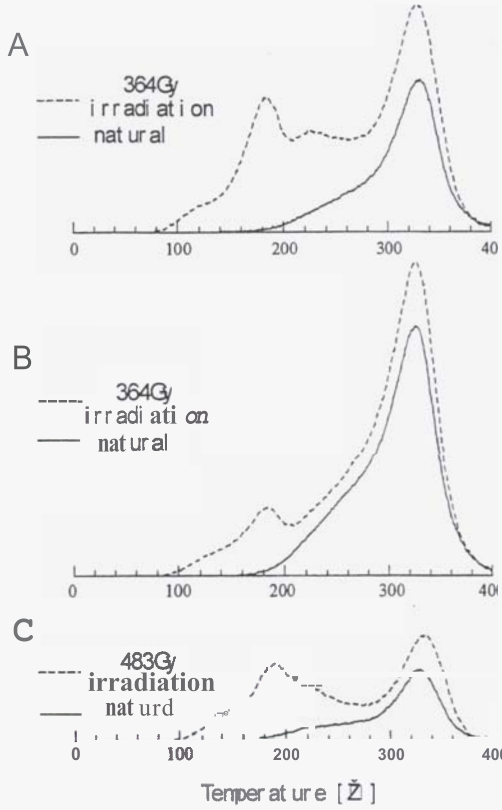

Fig. **4** Examples of TL glow-curve of quartz from the Takidani Granodiorite, a Quaternary pluton reportedly the youngest exposed granitoid on the Earth (after Tsuchiya and Fujino, 2000).

from granitic rocks have no, or extremely weak, TL emissions, but quartz from younger (Quaternary) granite (Kano and Tsuchiya, **2002;** Bando **et** *al.,* **2002)** give high TL emissions similar to quartz in volcanic and pyroclastic rocks of Tertiary and Quaternary age. The intensity of NTL was found to increase at approximately 160°C, and the apparent maximum temperature of the glow-curve appeared fkom **330°C.** The initial rise in temperature is almost the same **as** that of quartz in the Tamagawa Welded Tuff, shown in Fig. **3.**

After irradiation, artificial TL (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.**

4. TLIMAGES

Fig. 5 shows the TL images of a sliced granitic rock sample (Kurihashi granodiorite in the Kamaishi Mine). This sample was collected from **the** vicinity of a hydrothermal alteration vein, and **an** alteration halo can be recognized in the sample. Fig. 5(a) is an ordinary optical photograph at room temperature. The white wire in the centre is

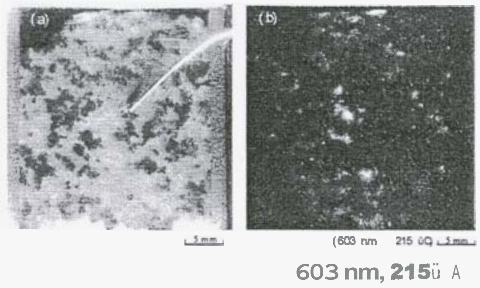

Fig. *5* TL images of sliced granitic rock.

a thermocouple for temperature control. The right-side photograph (Fig.5 (b)) is a TL map through *603* nm interference filter at 215°C. Bright TL spots corresponded to the position of felspar, which is mainly potassium feldspar. TL emission of quartz was very weak, Other samples, collected far from the alteration vein, did not show TL of feldspar. The intensity of TL in the feldspar corresponds to the distance from the hydrothermal vein and the degree of alteration

5. APPLICATION TO GEOTHERMAL EXPLORATION

5.1 Geothermal fields

Kokkonda Geothermal Field The Kakkonda geothermal field is one of the typical liquid-dominated geothermal systems in northeast Japan. The geology of the Kakkonda geothermal field is composed of the Miocene formations, the Tamagawa Welded Tuffs and Quaternary volcanics.

The Tamagawa Welded Tuffs, of Late Pliocene to Early Pleistocene age, are widely distributed in this field and are composed mainly of dacitic and rhyolitic welded tuff. Volcanic rocks of Quaternary age are divided into the Matsukawa Andesite and Iwate Volcanic Products. All samples for TL measuring are quartz, which was collected fkom field outcrop.

Minase Geothermal Field The Minase geothermal field, in Akita Prefecture, is located about 100 km SSW from the Kakkonda geothermal field. The Uenotai geothermal power plant is developed in this field. This area is mainly composed of sedimentary and pyroclastic formations of Neogene age, and Quaternary volcanics associated with pyroclastics are distributed through the field. Quartz for TL measurement was separated from drilling cores fkom these formations.

5.2 TL distribution

Fig. *6* shows the distribution of TL emissions in the Kakkonda geothermal field. Here, TL intensity represents the intensity relative to that of a reference sample. The contours of TL intensity are concentrically distributed around the Kakkonda geothermal power plant (KGPP). TL intensity decreases towards the power plant, with no TL emissions detected in the central part of the geothermal field, around the KGPP. Natural geothermal heating in the central zone exhausted the paleo-dose accumulated in quartz. Storage

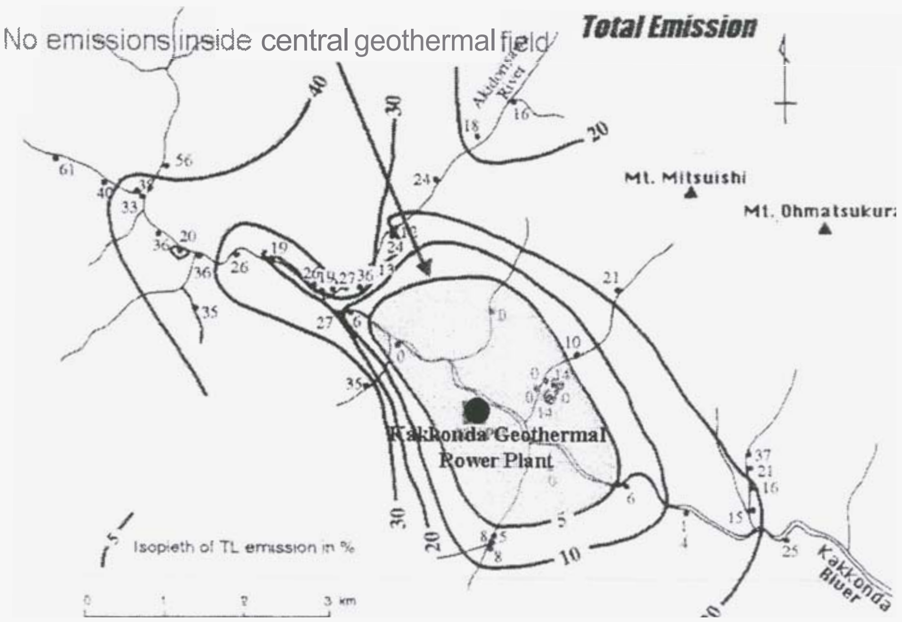

Fig. 6 Distribution of TL intensity in the Kakkonda geothermal field (Tsuchiya ***et al.,*** 2000).

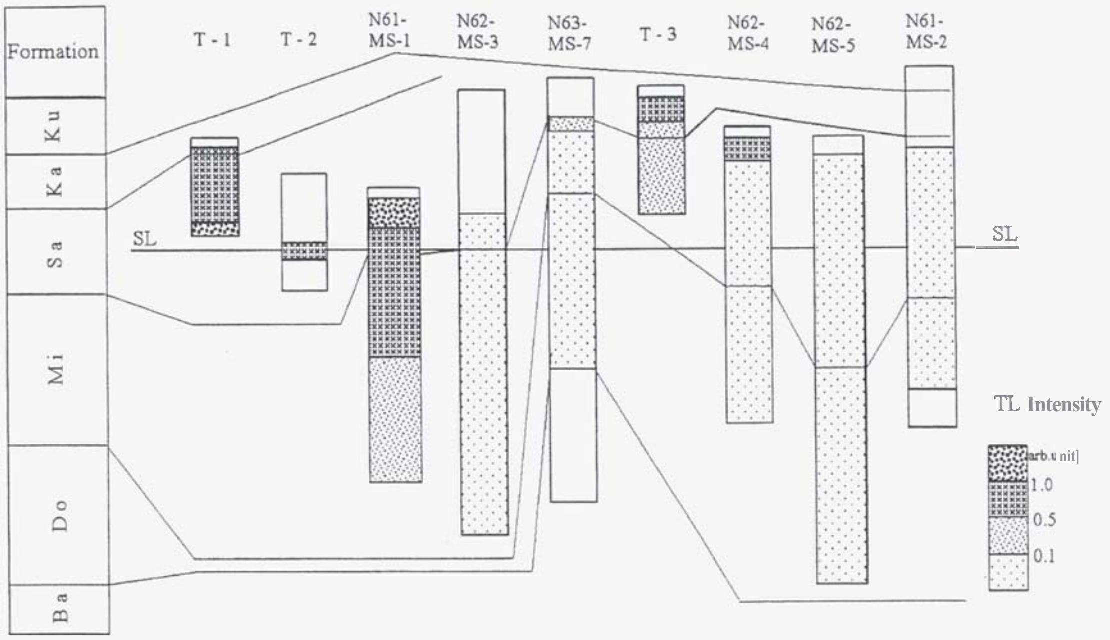

Fig. **7** Stratigraphical column and variation of TL intensity of quartz in drill cores from the Minase geothermal field (Nambu *et al.*, 1996.). Ba; basement, Do: Doyoru F., Mi; Minasegawa F., Sa: Sanzugawa F., Ka: Kabutoyama F., **Ku;** Kurikomayama volcanics, SL: sea level.

processes of ionising natural radiation is greater towards the edge of field, i.e. away from the central geothermal area. **The** total emission indicates relative prevalence of thermal manifestations. Tsuchiya *et al.* (2000) described detailed methodology for obtaining other information related to geothermal activity.

Fig. **7** shows vertical variation of TL intensity of **quartz** in several drilling cores from the Minase geothermal field. In general, deeper formations are older than upper (shallow) formations in the sedimentary sequences and caldera deposits. **A** high TL intensity of upper (younger) formations, was recognized in some boreholes (ex. N61-MS-1 and T-3), compared to the lower (older) formations, but no variation of TL intensity is evident with depth in T-1 borehole. TL intensity is independent of stratigraphy.

Reverse zoning of TL intensity, which means relatively high TL intensity in the upper (younger) formation, is inferred to be caused by geothermal activity. With increasing borehole depth, temperature increases, and the paleo-dose was released due to the high temperature. The T-1 borehole is located far from the central geothermal zone, so the original TL profile was maintained.

Fig. **8** shows the relationship between borehole temperature and TL intensity. The borehole temperature reflects present reservoir temperature, and is closely related to TL intensity. ***So,*** TL

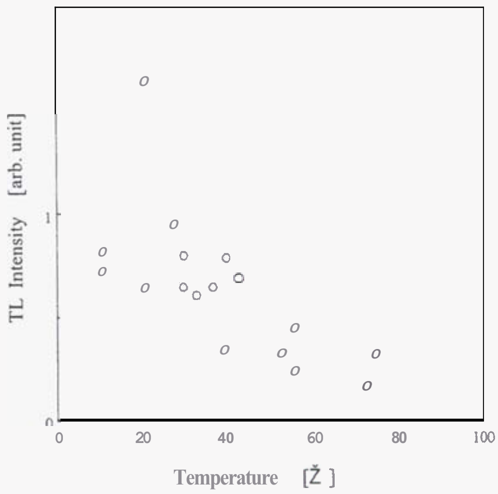

Fig. **8** Relationship between borehole temperature (at sampling point) and TL intensity of quartz in drill core fiom the Minase geothermal field (Nambu *et al.*, 1996).

intensity may prove effective as a qualitative geothermometer.

6. CONCLUSIONS

TL emission in geothermal areas is independent of stratigraphic boundaries, but it is closely related to surface geothermal manifestations and present geothermal temperatures. Thermally stimulated processes of TL caused by natural annealing, and radiation storage processes is initiated as a consequence of the temperature drop. TL behaviour is indicative of natural temperature manifestations, paleo-temperature history and geothermal fluid flow.

TL is an effective exploration tool for evaluating natural temperature manifestations and subterranean heat flow in geothermal systems. TL has been developed as a dating technique for geological materials, but paleo-dose in rock-forming minerals, particularly in quartz, of the geothermal field decayed by natural annealing associated with geothermal activity.

TL of feldspar reflect the effect of impurities and defects inside the crystal, and may be applied as a kind of geochemical sensor to determine trace element occurrences or an indicator of structural defects in crystals.

TL phenomena have several possibilities as indicators for evaluating geological processes, such as geothermometry, geospeedometry and as geochemical sensors. However, the mechanism of TL remains uncertain, so that further studies are required for development of quantitative TL geological indicators.

7. REFERENCES

Aitken, M.J., **1985.** Thermoluminescence Dating. Academic Press, 359pp.

Bando, M., Bignall, G., Sekine, K. and Tsuchiya, N. **(2002),** Petrology and uplift history of the Quaternary Takidani Granodiorite: Could it have hosted a supercritical (HDR) geothermal reservoir? *J. Volcanol. Geotherm. Res.* (in press)

Duller, G. A. T., Bøtter-Jensen, L., and Markey, B. G. **(1997), A** luminescence imaging system based on CCD camera. ***Radiat. Meas.* 27, 91-99.**

Kano, S. and Tsuchiya, N. (2002), Parallelepiped cooling joint and anisotropy of P-wave velocity in the Takidani granitoid, Japan Alps. *J. Volcanol. Geotherm. Res.* **114,465-477.**

Kirsch, *Y*. and Townsend, P. D. **(1988),** Speculations on the blue and red emission bands in the TL emission spectrum of albite and microcline, *Nucl. Tracks Radist. Meas.,* **14,4349.**

Hareyama, **M.,** Tsuchiya, N. and Takebe, M. **(1998),** Two-dimensional measurement of natural radioactivity of rocks by photostimulated luminescence, ***Proc. Water-Rock Interaction-9,* 835-838.**

Hareyama, M. Tsuchiya, N. Takebe, M. and Chida, T. (2000), Two-dimensional Measurement of Natural Radioactivity of Granitic Rocks by Photostimulated Lsuminescence Technique., ***Geochem. J.,* 34, 1 – 9.**

Hashimoto, T., Hayashi, Y., Koyanaga, A., Yokosaka, K. and Kimura, K. **(1986)** Red and blue colouration of thermoluminescence fiom natural **quartz** sands. ***Nucl. Tracks Radiat. Meas.,* 11, 229-235.**

McKeever, **S.W.S., 1985.** Thermoluminescence of solids. Cambridge University Press, 376pp.

Nambu, M., Mikami, K. Tsuchiya, N. and K. Nakatsuka, **(1996),** Thermoluminescence of quartz in the borehole cores from the Minase geothermal area, Akita Prefecture, Japan. ***J. Geotherm. Res. Soc. Japan,* 18 (1), 39 - 49** (in Japanese with English abstract)

Readhead, M.L., **1988.** Thermoluminescence dating study of **quartz** in aeolian sediments from southeastern Australia. ***Quaternary Science Reviews.* 7, 257-264.**

Telfer, D. J. and Walker, G. (1978), Ligand field bands of Mn^{2+} and Fe^{3+} luminescence centres and their site occupancy in plagioclase feldspars, *Mod. Geol.,* **6, 199-210.**

Tsuchiya, N., Yamamoto, **A., and** Nakatsuka, K., **1994.** Thermoluminescence of quartz in volcanic and pyroclastic rocks fiom the Kakkonda geothermal area, Northeast Japan - preliminary study of thermoluminescence geothermometer. ***J. Geotherm. Res. Japan.* 16, 57-70.** (in Japanese with English abstract)

Tsuchiya, N. and **Fujino,** K. **(2000),** Evaluation **Of** Cooling History of the Quaternary Takidani Pluton Using Thermoluminescence Technique, *Proc.* ***World Geothermal Congress 2000,* 3939 – 3944.**

Tsuchiya, N., Suzuki, T. and Nakatsuka, K. **(2000),** Thermoluminescence as a new research tool for the evaluation of geothermal activity of the Kakkonda geothermal system, northeast Japan. ***Geothermics,* 29, 27-50.**

Ypma, P.J. and Hochman, M.B.M., **1991.** Thermoluminescence geothermometry- a case study of the Otway Basin. ***J. Australian Petrol. Produc. Explo. Assoc.* 31, 312-324.**