APPLICATION OF THE THERMOLUMINESCENCE TECHNIQUE FOR EVALUATION OF GEOTHERMAL ACTIVITY

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SUMMARY – Thermoluminescence is a thermally derived phenomenon, with the thermoluminescence signature of minerals and rocks in the geothermal setting strongly affected by alteration processes and natural heating. The technique has potential as a sensitive geothermal/geochemical sensor in geothermal systems. Laboratory analysis of quartz was undertaken to identify the effect of hydrothermal fluid in the Waiotapu Geothermal Field (New Zealand), and two-dimensional TL and spectroscopy of rock samples from Kamaishi mine (Japan) were measured. These results show possibilities of thermoluminescence techniques for geothermal exploration, and a new approach for geological applications.

1. INTRODUCTION

Thermoluminescence (TL) phenomena have been effectively applied to date archaeological and geological materials that are younger than 1Ma. Here, the age is defined 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 hypotheses and/or premises, such as a constant dose rate from surrounding rocks, linearity of TL for unit dose and resetting 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 the sample’s 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 applied for dating, due to its chemical stability for emission. Quartz is a suitable mineral for evaluation of thermal effects in geothermal areas, by TL.

Feldspar may reflect hydrothermal events, because feldspar is easily altered, compared 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 image 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+} impurities. 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*. (2000) 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 a 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 highly sensitive CCD camera with an image intensifier and computer system, is shown in Fig. 1. The panel heater is 30 x 30 mm. The CCD camera (C4880-92) used with a S5466 CCD chip (Hamamatsu Phonics Co.), 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.

TYPE III: Spectral TL measurement system:

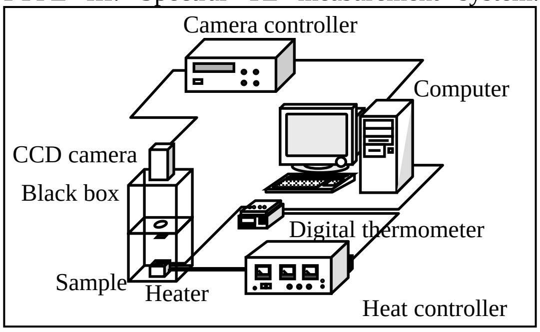

Figure 1 Schematic block diagram of TYPE II TL-measurement system.

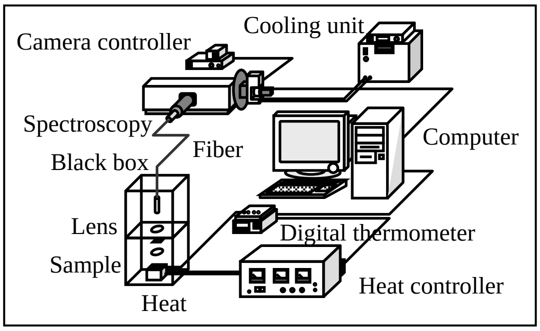

Figure 2 Schematic block diagram of TYPE III TL-measurement system.

Using a spectrometer coupled with high sensitive CCD camera, spectral data can be obtained from ultraviolet through visible light. The CCD camera with image intensifier (C4569-01, Hamamatsu Photonics Co.) is connected to spectrometer (C5094) with optical fibre.

2.3 Samples 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 minute 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 40˚C/min from room temperature to 400˚C is applied. The background component of the TL signal is approximated by the black-body radiation of the platinum plate.

TYPE II &TYPE III systems: The 30 x 30 x 1 mm sliced sample was heated from room temperature to 400˚C with a heating rate of 20, 30 or 40˚C /min. The exposure interval is 5 seconds for each 10 seconds, so the temperature difference during the exposure is less than 2˚C.

3. GLOW-CURVES

Examples of natural TL (NTL) glow-curves of quartz in granitic rocks are shown in Fig. 3, coupled with artificial TL glow-curves after irradiation of ^{60}Co gamma-ray. Generally, quartz from granitic rocks have no, or extremely weak, TL emissions, whilst 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 from 330˚C. The initial rise in temperature is the almost 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), called “afterglow”. The apparent maximum temperature of ATL was

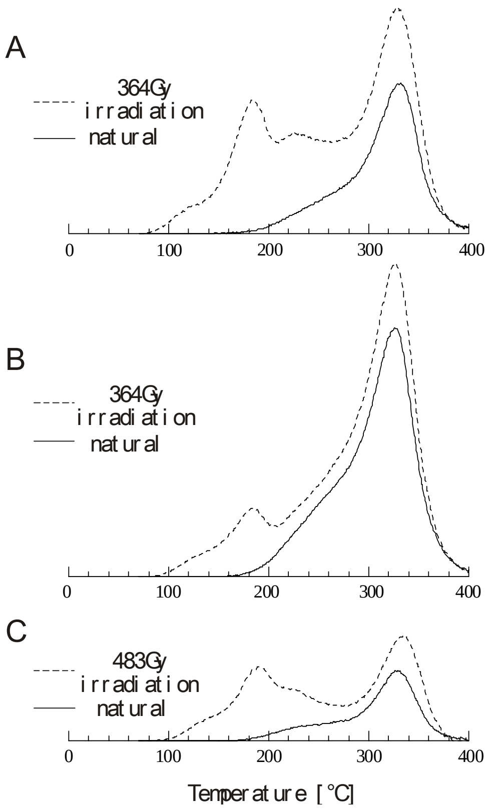

Figure 3. Examples of TL glow-curve of quartz from the Takidani Granodiorite, which is a Quaternary pluton, and it is reported to be the youngest exposed granitoid on the Earth (after Tsuchiya and Fujino, 2000).

the same as the NTL, with an additional unstable peak observed around 180˚C.

4. TL AS A GEOTHERMOMETER

4.1 Samples

Unaltered rock samples, and other samples with a range of alteration intensities were collected from surface exposures and from exploration drill lholes in the Waiotapu Geothermal Field (New Zealand). This field is one of the largest in New Zealand (Fig. 4), with a natural heat flow of about 540 MW_t and a surface area of 17 km^2 (Hedenquist and Browne, 1989). The local stratigraphy comprises flat lying felsic ignimbrite, tuffs, and lacustrine sedimentary rocks (Simmons, 1995).

Samples were crushed and measured by a TYPE I system.

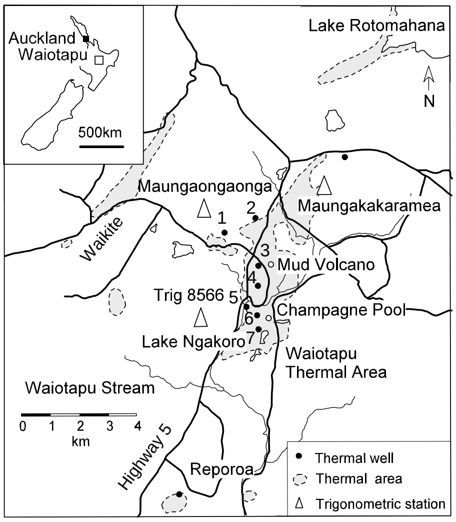

Figure 4. Geography of the Waiotapu area

4.2 Surface distribution of Natural TL

Natural TL (NTL) intensity is strongly affected by geothermal processes and does not preserve its initial paleodose. Fig. 5 shows different NTL intensities with different sampling points (centre, margin or outside thermal area).

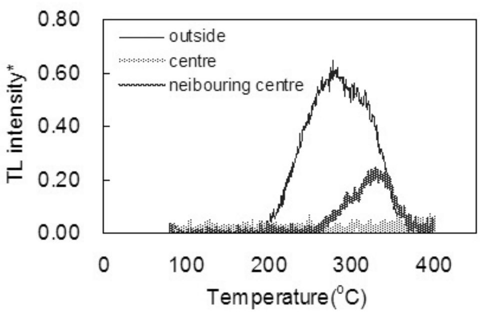

Figure 5. NTL measurement of Waiotapu surface samples depend on their location (inside / outside thermal area)

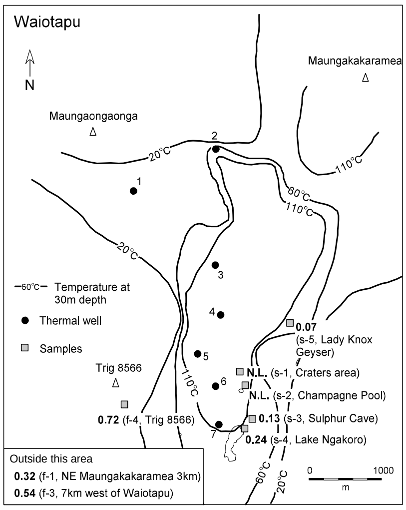

Figure 6. Map showing natural TL emission of surface samples.

NTL intensity of surface samples and the isotherm line are shown in Fig. 6. Isotherm lines for Waiotapu are shown by Hedenquist (1983) and Simmons (1995). Samples from inside the geothermal area show weaker NTL, at around 325°C, compared to samples fro outside the thermal area. Moreover, no NTL emission was observed for samples from the main thermal area (inside the 110°C contour line, for 30 m depth).

Tsuchiya *et al.* (2000) applied the TL method to the Kakkonda geothermal field, in NE Japan, who reported NTL intensity (especially lower temperature peaks) decreased towards the centre of the geothermal field because of natural annealing, and tmal exploration technique to evaluate thermal activity.

Therefore, NTL behaviour can indicate the presence of natural temperature manifestations and their paleo-temperature history. Furthermore NTL can represent a geothermal exploration technique to evaluate surface thermal manifestations and infer subterranean heat flow in geothermal systems.

4.3 Underground distribution of Artificial TL

Compared to surface samples, it is difficult to evaluate underground temperature using NTL measurement values because the minerals could be affected by temperature annealing with increasing depth. Little NTL emission could be observed from drill core samples, even at shallow depth. The Artificial TL (ATL) measurement has some possibility to assess underground temperature.

To compare three glow curves shown in Fig. 7, ATL intensity of low temperature peaks (270 °C) decrease and the higher temperature peak (325°C) is relatively stable. Hydrothermal water, especially higher temperature water, could affect the quartz lattice defects and reduce ATL

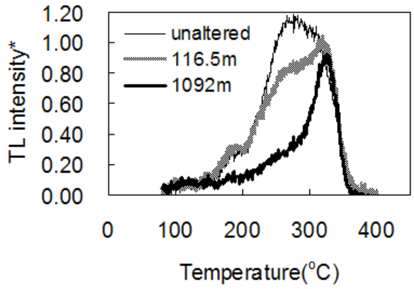

Figure 7. ATL measurement of Waiotapu drillcore samples from different depths (different temperature).

intensity. Our laboratory experiment conducted at hydrothermal conditions is described by Yamamoto and Tsuchiya (2004).

Figure 8 show the results of ATL measurement (ATL intensities for around 270°C peaks) for drill core samples from Waiotapu, with isotherm lines from Hedenquist and Browne (1989). Samples from the deeper part of the Waiotapu system show a weaker luminescence for the 270°C peak than shallow samples. In particular, ATL intensity decreases at points above the 250°C isotherm, irrespective of rock type or stratigraphic unit. ATL intensity at the 325°C peak is quite stable even at high temperature hydrothermal conditions. The middle stable ATL peak at 270°C reflects underground high temperature conditions and is most suitable as a geothermal temperature indicator.

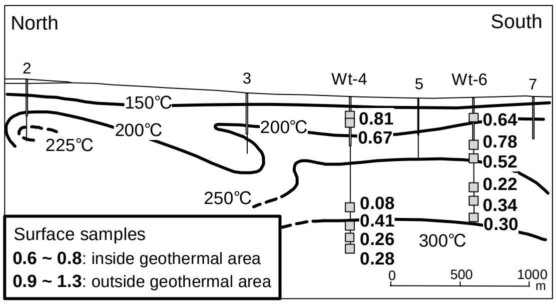

Figure 8. Map showing ATL at 270°C of drill core samples

5. TL AS A GEOCHEMICAL SENSOR

5.1. Samples

The sample is the Kurihashi granodiorite from the Kamaishi Mine, Japan (Fig. 9) that shows both altered and unaltered zones around the vein. XRD shows that the vein is mainly composed of zeolite, and the altered zone is mainly composed of quartz, K-feldspar and plagioclase (anorthite). The sample was cut out as a plate (vertical to the targeted vein); the size is 30 x 30 x 1 mm, and measured by TYPE II & III systems.

5.2. TL distribution around hydrothermal vein

A sample image pictured in natural light and pictured in luminescence is shown on Fig. 10,

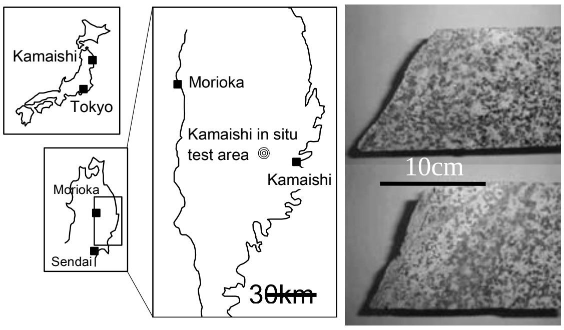

Figure 9. Location of the Kamaishi Mine and samples from the Kamaishi Mine.

showed strong luminescence around the vein. Tens of feldspar crystals are selected in one sample and for each the TL intensity is plotted versus the distance from vein edge (Fig. 11). The distance from the left end of the sample image is the distance from the vein. Yamamoto and Tsuchiya (2004) described detailed methodology for obtaining other information related to detect a mass transport front.

The fracture resulted from the reaction of thermal water, in an altered zone forming veins. In this process, an increase of the lattice defects and impurities around the vein affects the TL intensity with material dissolution or addition according to reactions. Therefore, it is possible that TL could be a chemical sensor that detects mass transport front.

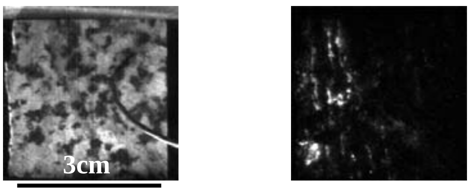

Figure 10. Result of two-dimensional TL measurement. Images on left side are samples in natural light and images on right side are natural TL (620 nm, 220°C).

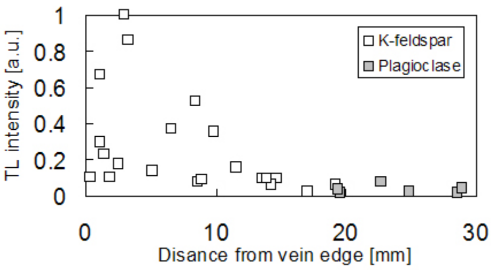

Figure 11. Relationship between TL intensity and distance from vein edge.

5.3. TL spectroscopy

Results are shown in Figure 12 by three dimensions (temperature, wavelength and TL intensity). In the altered feldspar, the main

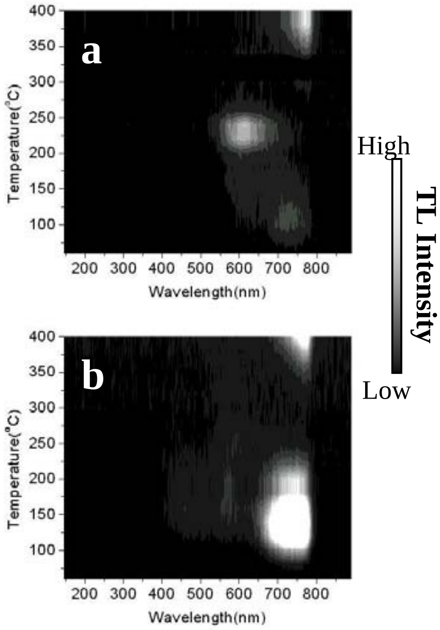

Figure 12. Results of spectral TL measurement for (a) altered feldspar, and (b) unaltered feldspar.

luminescence is at 250°C, 600 nm (red TL). In the unaltered feldspar, the main luminescence is at 150°C, 730 nm (far red TL), also blue TL could be observed (175°C, 450 nm). The peak at around 400°C in both results is the black body radiation of the heater. It may be possible that spectroscopy of TL could detects the alteration front more certainly.

6. CONCLUSIONS

The results of NTL distribution in surface samples correspond with the 110°C isotherm line at Waiotapu, at 30 m depth. NTL intensity was strongly affected by geothermal processes and could not preserve its initial paleodose. NTL behaviour reflects the presence of natural temperature manifestations and their paleo-temperature history.

It is hard to evaluate underground temperature by NTL compared to surface samples, because high temperature annealing occurs. However, ATL measurement values demonstrate its potential a geothermometer in the high temperature geothermal setting. The ATL intensity of lower temperature peaks decreases with increasing temperature conditions, especially samples collected from the >250°C zone, which showed strong reduction for the 270°C ATL peak. An ATL behaviour for the 270°C peak indicates the presence of a high temperature condition in the subsurface.

Two-dimensional and spectroscopy of TL were measured and evaluated around a vein that is a vestige of past hydrothermal activity. The distribution of radioactivity was also measured. Stronger luminescence occurred in the altered zone around veins and differences of spectra between the altered and unaltered zones were confirmed by spectroscopy measurement.

TL phenomena has strong potential for evaluating geological processes, as a geochemical sensor in the geothermal/hydrothermal setting, even though some of the mechanisms of TL remain unresolved. In addition, other minerals (e.g. feldspar and calcite *etc.*) have various TL phenomena, depending on their condition (e.g. style and intensity of alteration, impurities and defects in the crystal), as well as quartz, which may prove useful indicators of material and fluid movement in the geothermal setting.

6. ACKNOWLEDGEMENTS

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