



THERMOLUMINESCENCE AGE OF QUARTZ XENOCRYSTS IN BASALTIC LAVA FROM ONINOMI MONOGENETIC VOLCANO, NORTHERN KYUSHU, JAPAN

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Abstract: We determined the eruption age of basaltic rocks by application of thermoluminescence (TL) method, which is often used for TL dating, to quartz. Mafic magma only rarely includes quartz because of their mutual disequilibrium. The basaltic lavas reported herein include quartz as xenocrysts, as corroborated by their rounded or anhedral shape.

The basaltic lava used for this study is from the Oninomi monogenetic volcano in northern Kyushu, Japan. The volcano eruption was estimated as occurring 7.3–29 ka because the lava exists between two widespread tephra: Aira-Tanzawa ash (26–29 ka) and Kikai-Akahoya ash (7.3 ka). We succeeded in collecting *ca.* 200 mg of quartz by decomposition of 30 kg of the lava samples. TL measurements for the lava indicate the eruption age as 15.8 ± 2.5 ka, which is fairly consistent with the stratigraphical estimation. Although the TL method has played a considerable part in constraining the timescale of Quaternary events, its application has been limited to silicic samples. The present result demonstrates the availability of quartz for dating even of mafic rock.

Keywords: Oninomi monogenetic volcano, basaltic rocks, thermoluminescence dating, quartz xenocrysts, eruption age.

1. INTRODUCTION

Quaternary volcanism has had a great impact on recent geographical evolution and human history. A method for precise age determination of young volcanic rocks deserves to be established. The eruption age of volcanic rocks has been determined in many cases using K–Ar or Ar–Ar dating, or by other means. It is nevertheless difficult to apply them to young Quaternary basaltic samples

because of their low K contents and the possible occurrence of excess Ar. Indirect age determination using radiocarbon (¹⁴C) method has served as the only method to estimate the eruption age of young basalts. Some difficulties arise in determining an eruption age via the ¹⁴C method, such as possible effects of bioturbation, absorption of volcanic CO₂, and difficulty in recovering adequate or sufficient carbonic matter. Alternatively the TL age dating method has been applied to quartz-rich young silicic samples (e.g., Gillot *et al.*, 1978; Guerin and Valladas, 1980; Ichikawa *et al.*, 1982; Kanemaki *et al.*, 1991;

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Miallier *et al.*, 1994). It has not been applied, however, to basaltic samples because the mafic magma usually contains no quartz. Although some pioneering reports have described TL and infra-red stimulated luminescence (IRSL) ages of feldspar in basaltic rocks (e.g., May, 1977; Tsukamoto *et al.*, 2011), the former was controversial because of the anomalous fading of luminescence signals (Wintle, 1973). Recently, Takashima *et al.* (2002; 2004) applied the TL method to secondarily formed quartz crystals in altered mafic rocks. Although they demonstrated TL age determination of basaltic rocks, they ascertained not the eruption age but the hydrothermal alteration age of the basalts.

Mafic magma is disequilibrated with quartz, which makes it more difficult to discern the TL age of basaltic rock. Among our observations, however, some basaltic samples include quartz crystals with a resorption rim, which should be regarded as xenocrysts. The TL ages of such quartz xenocrysts are probably reset immediately after interaction with high-temperature basaltic magma because they easily lose luminescence signals by annealing. Therefore, the quartz xenocrysts in mafic samples are expected to be adequate for TL dating.

To assess the availability of the quartz xenocrysts as a probe for TL dating, the TL age of the quartz should be evaluated by comparison with other dating methods. Most earlier research efforts were made on TL dating using quartz extracted from heated siltstone xenolith

(e.g., Chen *et al.*, 2001), phreatic explosion (e.g., Miallier *et al.*, 2004; Preusser *et al.*, 2011; Rufer *et al.*, 2012), pumice (e.g., Shitaoka *et al.*, 2009; Miallier *et al.*, 1994) and baked sediments by a lava flow (e.g., Pilleyre *et al.*, 1992; Bonde *et al.*, 2001). However, very few attempts have been made at TL dating using crustal quartz xenocrysts that have survived in host basaltic magma. Mafic lavas in the Oninomi monogenetic volcano in northern Kyushu, Japan (Fig. 1) include a tiny amount of quartz xenocrysts (Ohta *et al.*, 1992). Although no report in the literature has described the exact age of the Oninomi volcanism, it has been estimated indirectly in tephrochronological studies. Constraints on age are extremely effective in evaluating TL age of the quartz xenocrysts. This report describes the eruption age of the Oninomi basaltic lava estimated by TL dating of the quartz xenocrysts, and presents discussion of the availability of quartz xenocrysts for investigating Quaternary basaltic activities.

2. GEOLOGICAL SETTING

The subduction zone in the southwestern Japan arc can be characterized by active subduction of the young Philippine Sea plate. The Oninomi monogenetic volcano is located near the intersection of the northern volcanic front of the Kyushu (Fig. 2), southwestern Japan arc with the Beppu graben system (Fig. 2). Quaternary volcanism

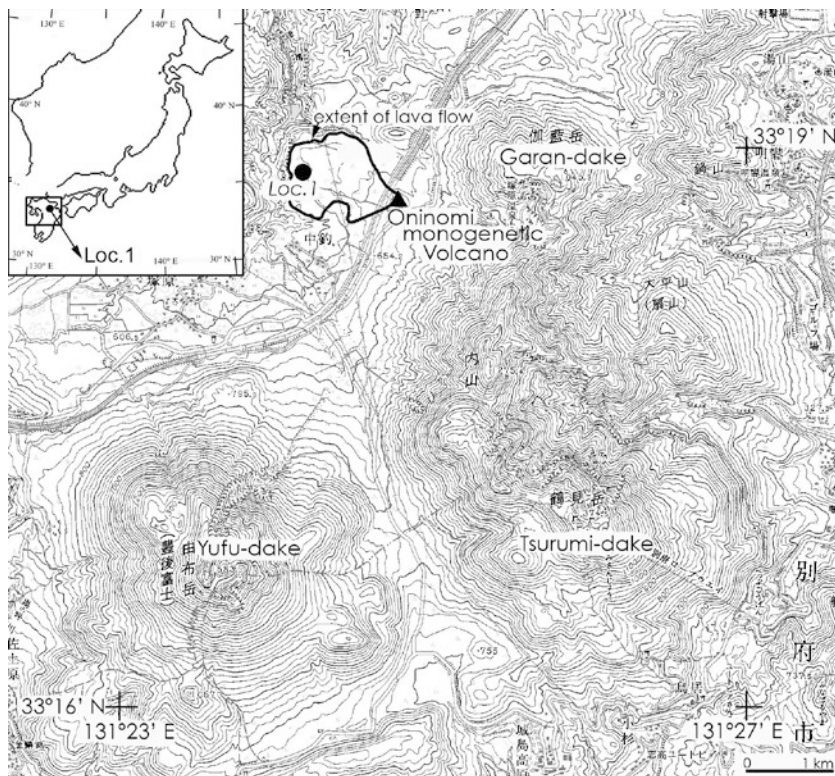


Fig. 1. Location map for points of importance in this study. Loc.1 is the sampling point of a lava flow from Oninomi volcano. Area covered by the rectangle in inset is presented in Fig. 2.

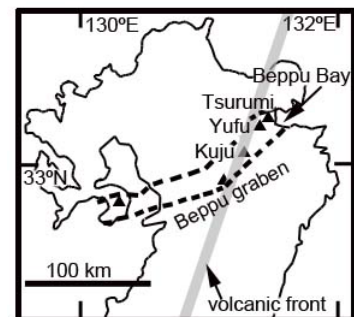


Fig. 2. Schematic map of northern Kyushu. Some volcanoes, the volcanic front, and the Beppu graben are shown.

in the Beppu graben is composed mainly of silicic lava flow and the formation of a lava dome complex in Yufudake, Tsurumi-dake, and Kuju active volcanoes. The basaltic activity of Oninomi monogenetic volcano is quite rare in the graben system. Itoh *et al.* (1998) asserted that the Oninomi basaltic volcanisms occurred at a corner of an active rhomboidal basin in Beppu Bay (Fig. 2). The Oninomi eruption formed a scoria cone near Tsurumi-dake, and effused two lava flow units with no interval (Ohta *et al.*, 1992). Ohta *et al.* (1992) and Kita *et al.* (2001) reported that the Oninomi basaltic lavas are classifiable as within-plate-type basalt based on their trace element compositions. Moreover, Itoh *et al.* (1998) and Yamamoto *et al.* (2009) showed that the strong local extension by a large releasing bend of the Median Tectonic Line (MTL) caused a rupture of the continental crust and subsequent non-arc type magma activity.

Ohta *et al.* (1992) reported that the Oninomi lava flows exist between two widely spread tephra horizons, which are Aira-Tanzawa ash (AT; *ca.* 26–29 ka by ^{14}C method: Machida and Arai, 2003) and Kikai-Akahoya (K–Ah; *ca.* 7.3 ka by ^{14}C method: Machida and Arai, 2003). In addition, Fujisawa *et al.* (2002) estimated the age of Oninomi lava as 10.5–29 ka indirectly based on the ^{14}C ages for overlying tephra layers of Tsurumi volcano. Therefore, the above constraints on the age of Oninomi volcanism are useful to evaluate the TL age of the quartz xenocrysts.

3. PETROGRAPHY

The modal composition of the Oninomi lava samples was analyzed using an automatic point counter. The Oninomi lava samples included 13–15 vol.% phenocryst content as olivine (4–5 vol.%, <1.5 mm), clinopyroxene (1–4 vol.%, <1 mm), plagioclase (5–6 vol.%, <2 mm), and opaque minerals (<0.8 mm). The olivine was euhedral or subhedral, sometimes showing a skeletal texture. Clinopyroxene was euhedral or subhedral. Clear euhedral plagioclase coexists with dusty anhedral plagioclase. A dusty zone was observed along the resorption rim of anhedral plagioclase. The Oninomi lava samples also contained minor (<1 vol.%) xenocrysts of hornblende (<3 mm) and quartz (<1 mm). Hornblende, observed as pseudomorph, was completely opacitized. The quartz xenocrysts were surrounded by a reaction rim of pyroxene (Fig. 3). Ohta *et al.* (1992) reported that these disequibrated quartz crystals are derived from silicic basement rock based on the description of crustal xenoliths in Oninomi samples. The groundmass showed an intergranular or intersertal texture with microlites of pyroxene, plagioclase, and opaque minerals.

4. SAMPLE PREPARATION AND ANALYSIS

We used approximately 30 kg of rock samples for this study, which were collected from a lava flow of Oninomi

volcano (Fig. 1). The surface layer of the rock samples, which had been exposed to sunlight, thereby reducing their luminescence, was removed using a diamond cutter. Then the samples were sieved into a fraction of 250–50 μm using a crusher and standard sieves. The sieved samples were treated initially with 20% HCl for 90 min. Feldspar and the other minerals except quartz were separated by etching with 20% HF for 90 min. The etched fraction (natural sample) was sieved into 150–75 μm grain size range to remove smaller grains and partially etched feldspars. We made a speculum, IRSL measurement after etching with 20% HF and sieving. If feldspar remained, samples were treated with 46% HF; then IRSL was measured. The acid treatment was repeated to achieve complete elimination of feldspar. After the treatment, we obtained no signal of feldspar by XRD analysis. Approximately 200 mg quartz was obtained using this preparation.

The TL measurements were performed using the original TL/OSL reader NRL-99-OSTL (Nagatomo *et al.*, 2007) at Nara University of Education (NUE). Samples were heated at a rate of $5^\circ\text{C}/\text{s}$ from 100°C to 400°C in a nitrogen atmosphere. The TL signal of 570–700 nm from a sample was guided to a photomultiplier tube (H7360–01; Hamamatsu Photonics K.K., Japan) through two condensing lenses and filters (OG570; Schott AG and HA50; Hoya Corp.). Additive doses were administered using a ^{60}Co gamma-ray source at NUE.

A standard multiple aliquot additive dose (MAAD) protocol (Aitken, 1985) was applied to the treated sample. In the MAAD protocol, TL is measured for both the natural sample and additive dose sample (artificially irradiated samples). The samples were divided into 20 fractions. Then some fractions were exposed to 25, 50, and 75 Gy with the ^{60}Co gamma ray at a dose rate of *ca.* 0.2 Gy/min. The equivalent dose of the sample can be

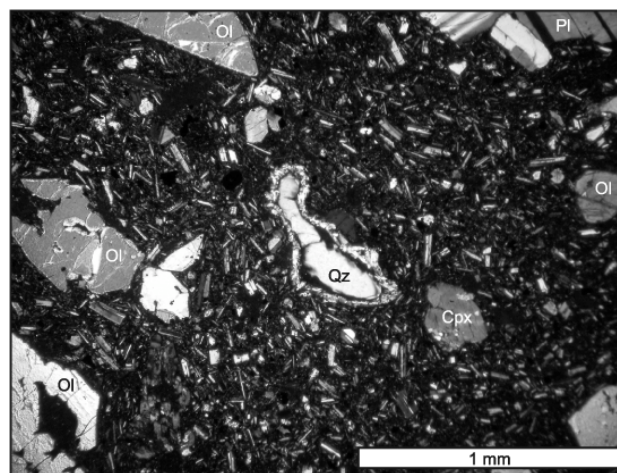


Fig. 3. Photomicrograph of a lava sample from the Oninomi monogenetic volcano. Abbreviations: Ol: olivine; Cpx: clinopyroxene; Pl: plagioclase; Qz: quartz.

estimated by fitting the data assuming linear dose dependence. Consequently, for correcting the nonlinear portion in the low-dose region, we measured TL of samples that had been annealed at 350°C for 60 min.

The annual dose (dose rate) measurements were performed using a gamma-ray spectrometer. The concentrations of U, Th, and K in the sample were analyzed using a Ge gamma-ray detector (EGSP 8785; Eurisy Mesures) at NUE. For the annual dose estimation, host lava sample was analyzed. The annual dose was calculated using dose-rate conversion factors (Adamiec and Aitken, 1998). The quartz xenocrysts grain size was from 500–1000 μm . We crushed them to between 150 μm and 75 μm during sample preparation. The beta attenuation factors were taken by Mejdahl (1979) as the grain size 750 μm . Contributions of the cosmic-dose rate to the annual dose were assumed according to methods described in earlier reports by Prescott and Hutton (1994) and by Shitaoka *et al.* (2009).

5. RESULTS

Fig. 4A presents TL measurement results. The equivalent dose was estimated by extrapolation of the linear trend in diagrams of TL growth vs. dose (**Fig. 4B** (a)). The temperature region of 320–335°C was satisfied with the plateau test (Aitken, 1985). The estimation method for nonlinearity correction (Δ in **Table 1**) was analogous to that of the equivalent dose (**Fig. 4B** (b)). The palaeodose of the sample was the sum of the equivalent dose and the nonlinearity correction.

The annual dose was obtained as the sum of beta, gamma, and cosmic dose rates. The palaeodose, annual dose, and TL age are presented in **Table 1**. The obtained TL age of the lava flow of the Oninomi monogenetic volcano was 15.8 ± 2.5 ka.

The error of the palaeodose was *ca.* 15.7%, as shown in **Table 1**, which resulted from scattering of data in the glow curves of additive dose samples (**Fig. 4A**). It is likely to have resulted from instability of the detector, because the red-hot glow could not be completely cut by the optical filters. To reduce the uncertainty of obtained palaeodose, it will be necessary to repeat the TL measurement or to develop a TL detection system to cut the red-hot glow.

6. DISCUSSION

Applicability of quartz xenocrysts for TL dating

To apply quartz xenocrysts as a probe for TL dating, it is necessary that the TL signal in the quartz be removed completely at the time of eruption. The effectiveness of zeroing of the TL signal depends on some annealing conditions such as the temperature, duration, and quartz size. Reaction coronas around the quartz (**Fig. 3**) offer a clue to understanding of the annealing conditions. Sato (1975) proposes that reaction coronas around the quartz xenocrysts in mafic lava can be interpreted as diffusion coronas. He succeeded in reproducing the coronas by annealing of polyhedral quartz with fine powder of basalt at 1400°C for 7 hr. The magmatic temperature in the subduction zone settings has been estimated as 800–

Table 1. Result of TL dating for Oninomi basaltic lava.

Equivalent dose (Gy)	Δ (Gy)	Paleodose (Gy)	U (ppm)	Th (ppm)	K ₂ O (wt%)	β ray (mGy/a)	γ + Cosmic ray (mGy/a)	Annual dose (mGy/a)	TL age (ka)
13.1 ± 2.3	13.0 ± 3.3	26.1 ± 4.1	0.90 ± 0.08	5.50 ± 0.35	1.41 ± 0.10	0.86 ± 0.06	0.80 ± 0.03	1.66 ± 0.07	15.8 ± 2.5

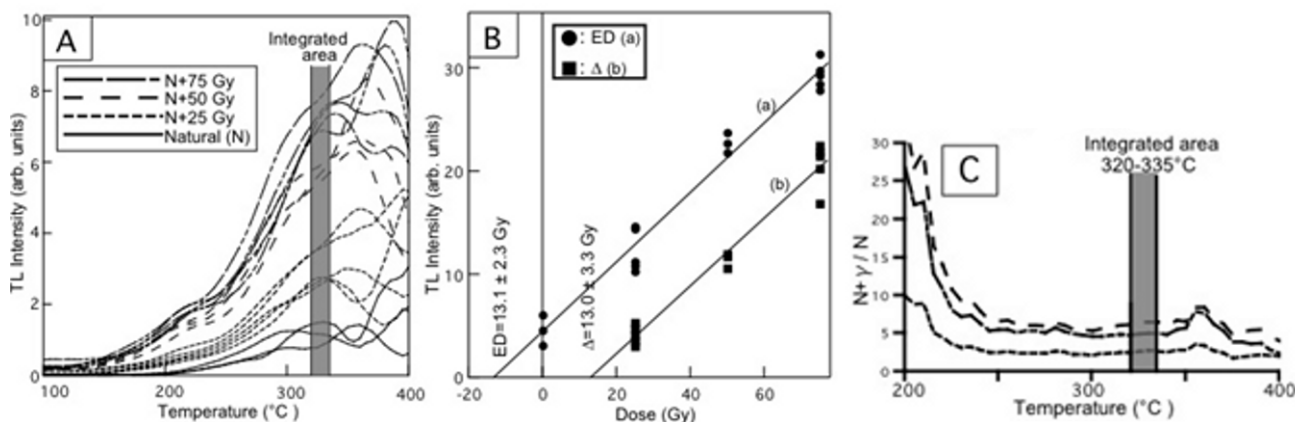


Fig. 4. TL glow curves (A), TL growth curve (B) and Plateau test (C). The plateau test was derived by $N+\text{Gamma} / N$. The plateau area (integrated area) is 320–335°C. The equivalent dose (ED) and nonlinearity correction (Δ) were estimated using the TL growth curve.

1100°C using mineral thermometers (e.g., Tamura, 1994; Straub, 2008). Even in cases of magmatic temperature of 1100°C, reproducing the coronas takes ten times (*ca.* 3 days) as long, or longer, than the time reported by Sato (1975). It is a sufficient time to raise the temperature in quartz xenocrysts of millimeter scale to the surrounding magmatic temperature. Therefore zeroing of the TL signal in the quartz certainly occurred during eruption and subsequent cooling period. The annealing of the quartz xenocrysts did not occur under a specific environment. It is, therefore, safe to say that quartz xenocrysts have the potential of elucidating the eruption age of host magma even if the magma has a mafic composition. The quartz grains used for this study were derived from crustal fragments trapped by ascending magma. Some advantages of using such exotic quartz as a TL index are as follows. One is that it covers young volcanic rocks of all kinds. We often observe quartz xenocryst even in basaltic rock as in the case of this study. Therefore TL dating using quartz xenocryst does not make any distinction according to the volcanic rock type. The other is complete zeroing of TL signal. The xenocryst quartz has survived within the basaltic magma. We did not observe phenocrystic amphibole in the basalt. Crystallization temperature of amphibole for the system of alkali basalt is higher than 1000°C (Yoder and Tilley, 1962), indicating that the eruption temperature of Oninomi basalt exceeded 1000°C. In addition, the existence of pyroxene reaction rim on quartz xenocrysts (Fig. 3) shows that these quartz xenocrysts were partially melted, and reacted with the host basaltic magma at high temperature. Consequently, the TL signal of the quartz was reset completely during entrainment by the basaltic magma.

Comparison of the eruption age of Oninomi lava with that of surrounding tephra was useful to evaluate the accuracy of TL dating using quartz xenocrysts. Ohta *et al.* (1992) showed that Oninomi lava exists between two widespread tephra layers, AT (*ca.* 26–29 ka) and K–Ah (*ca.* 7.3 ka) (Machida and Arai, 2003). In addition, Fujisawa *et al.* (2002) proposed that the age of the Oninomi lava was slightly older than *ca.* 10.5 cal ka BP based on ¹⁴C data of the overlying tephra layer on surrounding volcanoes. The TL age of 15.8 ± 2.5 ka obtained in this study is consistent with the tephrostratigraphical estimation and Fujisawa *et al.* (2002).

7. CONCLUSIONS

We conducted age dating for a Quaternary basaltic lava from the Oninomi monogenetic volcano by analyzing quartz xenocrysts, obtaining the TL age of 15.8 ± 2.5 ka, which is consistent with the previously estimated tephrochronological age and with ¹⁴C data (*ca.* 10.5–29 ka). Our results suggest that TL dating is widely available to quartz xenocrysts even in mafic samples. Further determination of TL age of quartz xenocrysts is expected to

have great potential for clarifying volcanic history in the Quaternary period.

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