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**Groundmass pyroxene crystal trachts record syneruptive magma dynamics in glassy  
pyroclasts**

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## Abstract

Explosive eruptions produce various types of pyroclasts including those contain few with groundmass crystals. The textural variation such as the number density, size, and volume of groundmass crystals is expected to reveal the complex magma dynamics during syneruptive ascent in conduits; however, we have no quantitative method to investigate the magma dynamics based on crystal texture when the pyroclasts show glassy texture with a small number of microlites. Here we show that the variation of combination of crystallographic faces (i.e., tracht) of groundmass pyroxenes enables us to derive the degree of effective undercooling ( $\Delta T_{\text{eff}}$ ) and magma ascent histories even from the glassy pyroclasts. We conducted decompression experiments and analyzed trachts of groundmass pyroxenes in the run products in addition to those in natural pumices from the 1914 Plinian eruption of Sakurajima volcano. These results show that the glassy white pumices experienced higher  $\Delta T_{\text{eff}}$  than the crystal-rich gray pumice, and corroborated that they originate from the magmas at different positions from the conduit walls. The estimate on  $\Delta T_{\text{eff}}$  implies that the magma rapidly ascended in the center of the conduit might experience cooling because of volatile exsolution and expansion.

**Keywords:** pyroxene, crystal habit, crystal size distribution, nanolite, magma ascent

## Introduction

Magma ascent histories in volcanic conduits are important for understanding the evolution of eruptive activities (Cashman and Sparks, 2013; Cassidy et al., 2018). The ascent histories can be recorded in the textures of groundmass crystals; therefore, the investigation of groundmass texture has played an important role to reveal the magma dynamics. Since the crystallization kinetics, especially nucleation rates, vary with the degree of effective undercooling ( $\Delta T_{\text{eff}}$ ) (e.g., Hammer and Rutherford, 2002; Brugger and Hammer, 2010; Shea and Hammer, 2013), the number density of groundmass crystals (Toramaru et al., 2008) and their size distribution (CSDs; Marsh, 1998) have been used to investigate magma ascent processes. These methods are based on the theoretical premise that the nucleation rate and the resultant number density are positively correlated with  $\Delta T_{\text{eff}}$ . However, the positive correlation is not universally true. For instance, explosive eruptions can produce various types of pyroclasts, such as microlite-free white pumice with higher vesicularity and gray pumice with lower vesicularity and abundant microlites (e.g., Polacci, 2005). The glassy texture of the former is attributed to the considerably fast ascent; the ascending magma was probably quenched before crystallization could proceed to a significant extent or within the nucleation delay (e.g., Mollard et al., 2012; Arzilli et al.,



2020; Rusiecka et al., 2020; Rusiecka and Martel, 2022). In this situation, it is difficult to extract syneruptive ascent histories from the crystal number densities in glassy pyroclasts, because the theoretical premise that the nucleation rate and the resultant number density are positively correlated with  $\Delta T_{\text{eff}}$  is not valid. In such scenarios, a new indicator is required to estimate  $\Delta T_{\text{eff}}$  from glassy groundmass with a small number of crystals.

Previous studies have demonstrated that crystal habits reflect  $\Delta T_{\text{eff}}$  (e.g., Lofgren, 1974; Hammer and Rutherford, 2002; Shea and Hammer, 2013; Arzilli et al., 2022). Kouchi et al. (1983) revealed experimentally that the relative growth rates for different faces of clinopyroxene (Cpx) change with  $\Delta T_{\text{eff}}$ . Their data indicates that, with increasing  $\Delta T_{\text{eff}}$ , a pyroxene crystal would have different combinations of crystallographic faces (i.e., tracht; Sunagawa, 2005) by losing some faces. This hypothesis was confirmed by Okumura et al. (2022b), in which it was experimentally revealed that the tracht of groundmass pyroxenes changes from octagonal prisms consisting of  $\{100\}$ ,  $\{010\}$ , and  $\{110\}$  prismatic faces into hexagonal prisms lacking  $\{100\}$  faces as  $\Delta T_{\text{eff}}$  increases. Therefore, the tracht of groundmass pyroxenes is expected to allow us to estimate  $\Delta T_{\text{eff}}$  and magma ascent histories from the glassy pyroclasts with a few number of the crystals.

In this study, we aim to infer magma ascent dynamics from the 1914 Sakurajima

pumices including glassy one by the trachyte analyses of groundmass pyroxenes. By performing decompression experiments on hydrous Sakurajima dacite magma, we investigated the differences in the syneruptive evolution of  $\Delta T_{\text{eff}}$  and the magma ascent histories between the different pumice types. This study proposes the applicable analytical method to glassy pyroclasts towards the investigation of magma ascent dynamics during explosive eruptions.

## Sample description

Sakurajima is located on the southern rim of the Aira caldera, Kyushu, Japan, with two summit vents aligned north–south: Kitadake and Minamidake. Eruptive activity has been summarized by Kobayashi et al. (2013) and is abridged here. Sakurajima started erupting at 26 ka, and its activity is subdivided into four stages based on tephra stratigraphy and chronology: the “Older Kitadake” (26–24 ka), “Younger Kitadake” (13–5 ka), “Older Minamidake” (4.5–1.6 ka), and “Younger Minamidake” (since 764 AD) stages. All eruptive products are andesitic to dacitic. Four large eruptions have occurred during the Younger Minamidake stage: the Tenpyo-Hoji (764–766), Bunmei (1471–1476), An-ei (1779–1782), and Taisho (1914–1915) eruptions. Each of these events had a similar eruptive sequence: an early Plinian eruption followed by lava outflows from lateral vents. In particular, the 1914–1915 Taisho eruption began with seismic activity on 11 January 1914, and the Plinian eruption began at the fissure vents on the western flank on 12 January; ten minutes later, a synchronous Plinian eruption began on the eastern flank. The Plinian phase continued on both flanks for about 36 h and was followed by lava extrusion and intermittent smaller explosions over the following two weeks. Additional outflows of lava began on the eastern flank in February and lasted for 1.5 years.

Seismic and geodetic monitoring of activity at Sakurajima has revealed the structure of the present magma plumbing system (e.g., Omori, 1916; Iguchi, 2013; Iguchi et al., 2013). A major magma reservoir exists at ~10 km depth beneath Aira caldera and minor reservoirs are present at ~5 km beneath the summit, from the north flank of Kitadake to Minamidake (Iguchi, 2013; Iguchi et al., 2013). The diameter of the conduit beneath Minamidake crater is estimated to be 300–500 m at 2 km depth and 40–60 m at the shallowest depths (Iguchi et al., 2013). Araya et al. (2019) analyzed melt inclusions in phenocrysts from the 1914 Sakurajima pumice and showed that the volatile saturation pressure ranged from 20 to 72 MPa. They proposed that the magma was pre-charged in a thick conduit at depths of 0.9–3.2 km below the surface, i.e., shallower than the minor reservoirs that feed present-day Vulcanian eruptions.

The textural analyses of the 1914 Sakurajima Plinian pumice have been performed by Nakamura (2006). The pumice clasts are classified into three types with different color, vesicularity, and microlite content. Type-1 and Type-2 are white pumice with high vesicularity (> 55 vol%): the former is glassy with lower modal abundance of plagioclase microlites than the latter (< 1.0 vs. 1.0–11.0 vol%). Type-3 is gray pumice with low vesicularity (25–50 vol%) and the relatively high modal abundance (8–16 vol%). All the

types have similar bulk groundmass compositions ( $\text{SiO}_2 = 67\text{--}70 \text{ wt\%}$ ) and the same mineral assemblage. Phenocrysts ( $> 100 \text{ }\mu\text{m}$  in length) consist of plagioclase, orthopyroxene (Opx), augite, and magnetite, while microlites ( $< 100 \text{ }\mu\text{m}$ ) are plagioclase, Opx, pigeonite, augite, and titanomagnetite.

We collected pumice clasts erupted by the 1914 eruption from an outcrop at 4.5 km east of the Minamidake summit and examined three pumices as the representative of Type-1, 2, and 3.

### Decompression experiments

We coarsely crushed the Type-1 pumice clasts, removed phenocrysts under an optical microscope, and collected the groundmass fragments as the starting material. Aliquots of 7–9 mg of the starting material and enough water ( $> 5 \text{ wt\%}$ ) to achieve water-saturation were loaded into Au capsules (3 mm outer diameter and 1–2 cm long), sealed by welding. We conducted the experiments using the cold-seal pressure vessels with Ni filler rod at Tohoku University (Okumura et al., 2021). The capsules were heated in the vessels at temperatures of 935 or 955 °C under a pressure of 120 MPa (assuming the minor reservoir at ~5 km depth) for 1 h. Those at 955 °C were then isothermally decompressed to

final pressures,  $P_f$ , of 5, 10, 20, or 50 MPa in different manners: single step decompression (SSD) and continuous decompression (CD). In the SSD experiments, the capsule was rapidly decompressed ( $> 10$  MPa/s) and held at  $P_f$  for 3 h and then quenched by dropping it into a water-cooled zone in the system. The capsule in the CD experiment was decompressed to  $P_f = 5$  MPa with a rate of  $\sim 0.01$  MPa/s and quenched immediately at  $P_f$  in the same way. Control experiments (EQ) were conducted by quenching without any decompression. The conditions of experiments are listed in Table 1.

During the experiments, the oxygen fugacity ( $f_{O_2}$ ) was buffered at NNO to NNO+1 (Okumura et al., 2021). The error in temperature is assumed to be within  $\pm 5$  °C. Since the duration at the initial condition (955 °C) was restricted by the durability of pressure vessel, there is the possibility that the inherent heterogeneity of the starting material remains without equilibrium and affect the resultant textures of run products. For this reason, the error in  $\Delta T_{\text{eff}}$  could be larger than the thermal error of  $\pm 5$  °C.

The recovered capsules were carefully opened; however, the recovered materials broke into pieces because of their high vesicularity and fragility, and lost their positional relations within the capsules. The materials were then mounted in resin for polishing and subsequent analyses.



## **Analytical methods**

### **FE-SEM-EDS**

The polished pumices and run products mounted in resin were observed using field-emission scanning electron microscopes (FE-SEMs) at Kyoto University. The chemical compositions of glasses were measured using a FE-SEM (JEOL JSM-7001F) coupled with an Oxford Instruments X-Max150 energy dispersive X-ray spectrometer (EDS) and its associated analytical software AZtec. The measurements were performed on multiple square regions ( $\sim 1 \times 1 \mu\text{m}^2$ ) at an accelerating voltage of 15 kV and a beam current of about 0.10 nA for 60 s. Alkali (Na and K) losses during the measurements were corrected by calibration against analyses of larger areas in the sample or in closely relevant samples. The average glass compositions of pumices and run products are described in Table 2 and 3, respectively.

### **Textural analyses with SEM-BSE images**

Backscattered electron (BSE) images for textural analyses of groundmass pyroxene crystals in samples and run products were obtained using a Thermo Fisher Scientific Helios NanoLab G3 CX FE-SEM operating at accelerating voltages of 5–10 kV



and at working distances of 4–10 mm. In run products, 3–12 regions were selected for analysis in each sample and their BSE images were obtained at 3,500× magnification (corresponding to regions ~40–60 μm on a side and pixel sizes of ~20 nm). Using ImageJ software, we measured the areas of the rectangular regions examined (i.e., the entire image) and those occupied by vesicles and individual pyroxene cross sections, as well as pyroxene cross-sectional widths ( $w$ ), which was measured as the minor axis of the best-fit ellipse. We discriminated pyroxenes from other minerals based on their contrast in BSE images and SEM-EDS chemical maps. In addition, BSE images of pyroxene crystals larger than 0.2 μm wide were acquired at higher magnifications (10,000–65,000×) for trach classification.

In the natural pumice samples, because of the low number densities of groundmass crystals, large rectangular areas were required to include a sufficient number of pyroxenes within the BSE images (e.g., >1 mm on a side for Type-1 pumice). However, it was difficult to acquire and process such a large image at a resolution adequate for the measurement of crystal size and trach. Therefore, we obtained magnified images of the pyroxene crystals (magnifications of mainly 8,000–50,000× and pixel sizes of <20 nm) at an accelerating voltage of 5 kV and a working distance of 4 mm, as well as larger, lower-resolution (54 nm/pixel) BSE maps of the analyzed areas at an accelerating voltage

of 10 kV and a working distance of 10 mm. The images of the analyzed areas were used to measure vesicularity and analyze the groundmass areas (i.e., excluding vesicles), and the magnified images were used to measure the sizes and trachts of groundmass pyroxenes in the same manner as in the run products. Analyzed crystals were larger than  $10^{-0.8} \mu\text{m}$  ( $\sim 0.16 \mu\text{m}$ ) wide in accordance with the size interval of crystal size distributions (CSDs).

In BSE images at higher magnifications, each pyroxene cross section could be classified into one of several tracht types based on the number of faces between pairs of parallel faces: octagonal, heptagonal, hexagonal, pentagonal, or parallelogrammatic (Okumura et al. 2022b, 2023; Fig. 1). For example, the octagonal and hexagonal trachts have three and two faces between any pair of parallel faces, respectively, and the heptagonal tracht has the properties of the octagonal tracht on one side and those of the hexagonal tracht on the other (Fig. 1c). Pyroxenes that were difficult to classify as having a particular tracht were classified as “other”. If cross sections were incomplete (e.g., chipped), we classified them based on the remaining parts; however, those with no pairs of parallel faces were classified as “other” (Fig. 1d). Intermediate trachts, such as heptagonal, could only be identified when the cross section had almost all of the constituent faces (Fig. 1c). When multiple crystals were attached to each other, each individual crystal was classified

separately (Fig. 1e).

## **Acquisition of tracht-specific CSDs**

We acquired the size distributions of pyroxene cross sections in each tracht group (hereafter, tracht-specific CSDs). In run products, we counted the number of crystal cross sections of each tracht in each 0.4- $\mu\text{m}$  size interval (i.e., 0.2–0.6  $\mu\text{m}$ , 0.6–1.0  $\mu\text{m}$ , etc.) and plotted the number at the center of the size interval. We did not calculate crystal number densities because the textures in the run products were extremely heterogeneous and thus could not be represented adequately by a single value.

For the natural pumice samples, the datasets of analyzed crystals were converted to tracht-specific CSDs using CSDCorrections v. 1.61 (Higgins, 2000), which converts cross-sectional widths into 3D long-axis lengths ( $L$ ) using the 3D aspect ratio  $S:I:L$  ( $S$  and  $I$  being the short- and intermediate-axis lengths, respectively). We estimated the representative 3D aspect ratio of pyroxenes regardless of tracht for each sample by using ShapeCalc (Mangler et al., 2022), which compares a measured distribution of cross-sectional aspect ratios with model distributions based on the algorithm used in CSDCorrections and returns estimated 3D aspect ratios (and the cumulative goodness of fit,

$R_c^2$ ) for a range of shapes from 1:1:1 to 1:20:20. Although CSDCorrections yields 3D CSDs expressed as a function of  $L$ , we expressed the CSDs as a function of 3D short-axis lengths ( $S$ , or cross-sectional width) with additional corrections (i.e.,  $S$ -plot CSDs; Okumura et al., 2022a). The details of the procedure for acquiring tracht-specific CSDs are described in Okumura et al. (2023). The CSDs were plotted in logarithmic size intervals with five intervals per decade larger than  $0.1\ \mu\text{m}$  (i.e., each interval is  $10^{0.2}$  times as large as the next smaller interval:  $10^{-1.0}$ – $10^{-0.8}\ \mu\text{m}$ ,  $10^{-0.8}$ – $10^{-0.6}\ \mu\text{m}$ ,  $10^{-0.6}$ – $10^{-0.4}\ \mu\text{m}$ , etc.). We note that the effective image resolutions restricted our analysis to crystals larger than  $10^{-0.8}\ \mu\text{m}$ .

## **TEM analyses**

Groundmass pyroxene crystals in the samples were observed under a transmission electron microscope (TEM; JEOL, JEM-2100F) equipped with a Gatan Orius 200D CCD camera and a JEOL JET-2300T EDS detector at an accelerating voltage of 200 kV. Ultrathin sections approximately 100 nm thick were prepared using a focused ion beam system (Thermo Fisher Scientific, Helios NanoLab G3 CX): the  $\text{Ga}^+$  ion gun was operated first at 30 kV and 21–0.083 nA, then at 16 kV and 0.13 nA for thinning, and finally at 2 kV and 77 pA for final processing. To determine mineral phases and crystallographic

orientations, selected-area electron diffraction (SAED) patterns were analyzed using Gatan's proprietary DigitalMicrograph software and ReciPro (Seto and Ohtsuka, 2022). In addition, we obtained compositional maps of pyroxenes by EDS analyses in scanning TEM (STEM) mode. For quantitative X-ray analyses by STEM, we used the  $\zeta$ -factor method (Watanabe and Williams, 2006). To achieve the accurate electron beam current measurements required for the  $\zeta$ -factor method, appropriate calibration was performed with the CCD camera beforehand. Furthermore, we acquired annular dark-field STEM (ADF-STEM) images of pyroxene crystals perpendicular to their *c*-axes to observe compositional zoning at higher resolution.

## Results

### Textural variation of the Sakurajima pumice

**Groundmass texture.** As described in Nakamura (2006), groundmass crystals consisted of plagioclase, clinopyroxene (Cpx) including augite and pigeonite, orthopyroxene (Opx), and Fe-Ti oxide, with a trace amount of apatite. Their size range was mainly 0.1–10  $\mu\text{m}$  in width.

Figure 2 shows the variation in groundmass texture among the pumice samples. The Type-1 pumice had higher vesicularity (75.0 %; Table 4), deformed vesicles, thinner vesicle walls, and few groundmass crystals (Fig. 2). The Type-2 pumice showed a vesicle texture similar to the Type-1 pumice but had more groundmass crystals and more-evolved glass composition ( $\text{SiO}_2 \sim 70$  wt%, 1 % higher than Type-1; Table 2). The groundmass pyroxenes were characterized by hexagonal and parallelogrammatic trachts in the Type-1 and Type-2 white pumice samples (Fig. 2).

In contrast, the Type-3 pumice showed distinct textures (Fig. 2): lower vesicularity (67.1 %; Table 4), less-round vesicle shapes, thicker vesicle walls, abundant groundmass crystals, and the most evolved glass composition ( $\text{SiO}_2 \sim 73$  wt%; Table 2). In addition, the

groundmass crystals in the Type-3 pumice were loosely oriented (lateral direction in Fig. 2). The crystals were slightly rounded, and groundmass pyroxenes were characterized by rounded octagonal tracht.

**Tracht-specific CSDs.** Figure 3 shows the results of shape estimation by ShapeCalc. The best-estimated shape (*S:I:L*) was 1.00:1.40:3.60, 1.00:1.75:6.00, and 1.00:1.30:4.60 for the Type-1, Type-2, and Type-3 samples, respectively (Table 5). Note that the number of crystal cross sections used for the estimation ( $n = 90$ ) was fewer than the recommended number ( $> 200$ ; Mangler et al., 2022); therefore, the estimated 3D aspect ratio in the Type-1 pumice should involve large uncertainty, as shown in Figure 3a.

With the estimated 3D aspect ratios, the conventional (i.e., including all crystal trachts) and tracht-specific CSDs were acquired (Fig. 4). The conventional CSDs were concave up, especially in the white pumice samples (black lines in Fig. 4a and 4b). Their slopes were steepest at the size range of nanolite (i.e., crystals smaller than  $1\ \mu\text{m}$  in width; Mujin et al., 2017). In contrast, the conventional CSD in the Type-3 pumice (Fig. 4c) showed a gentler slope. The Type-3 pumice contained large crystals (mainly  $< 5\ \mu\text{m}$  in width) as compared with the two white pumice samples. The crystal number density was the highest in the Type-3 pumice, and the lowest in the Type-1 pumice.

The tracht-specific CSDs exhibit an obvious difference between the white pumice (Type-1 and Type-2) and the gray pumice (Type-3). In the white pumice samples, octagonal pyroxenes had larger widths (mainly  $> 1 \mu\text{m}$ ), and parallelogrammatic ones were distributed in the size range of nanolites (i.e.,  $< 1 \mu\text{m}$  in width), where the conventional CSDs showed the steepest slope (Fig. 4a and 4b). Hexagonal crystals were distributed throughout the size range of both microlite and nanolite. The CSD slopes of these trachts were consistent with those of the conventional CSDs at the corresponding size ranges. Heptagonal and pentagonal trachts, which are the transient shapes between hexagonal and octagonal or parallelogrammatic trachts, exhibited the intermediate size ranges and slopes (Fig. 4b). In contrast, the tracht-specific CSD of the gray pumice sample (Type-3) was characterized by only the octagonal distribution with a gentle slope (Fig. 4c). Almost all of the nanolites showed octagonal tracht.

**Internal texture of groundmass pyroxene crystals.** The results of TEM observation are shown in Figures 5–7. The parallelogrammatic tracht consists of  $\{110\}$  prismatic faces (Fig. 6). The hexagonal tracht is composed of  $\{110\}$  and  $\{010\}$  faces (Fig. 5), and the octagonal tracht has additional  $\{100\}$  faces (Fig. 7). The mineral phases of groundmass pyroxenes were mainly Cpx composed of augite ( $C2/c$ ) and pigeonite ( $P2_1/c$ )



(Figs. 6 and 7), and occasionally Opx (Fig. 5). The Opx domain, when present, was distributed along (100) plane at the center of crystal (Fig. 5). The center of pyroxene crystals tended to be occupied by Ca-poor phases (i.e., pigeonite and Opx), and both sides were generally Cpx including augite (Figs. 5–7). Pigeonite domains showed anti-phase boundaries (Fig. 6h).

The pyroxene crystals sometimes showed concentric zonation. The pyroxenes tended to be Al-poor at their cores (Figs. 5c, 6c, and 7c); moreover, the Al content decreased again at their rims in those from the Type-3 pumice sample (Fig. 7c). In addition, the pyroxenes from the Type-3 sample showed normal zoning in Mg# [ $\text{Mg}/(\text{Mg} + \text{Fe})$  in mol; Fig. 7d]. We found an octagonal pyroxene microlite with parallelogrammatic zonation from the Type-3 pumice (Fig. 7).

### **Decompression experiments**

The SEM and TEM analyses showed that the groundmass of the control experiment at 935 °C (EQ935°C120MPa1h) contained augite, low-Ca pyroxene (Opx and pigeonite), Fe-Ti oxides (magnetite and hematite), plagioclase, and apatite. Groundmass pyroxene crystals were mostly augite and ubiquitous in the groundmass (Fig. 8a and 8b). In

contrast, the experiments at 955 °C showed inhomogeneous textures (Fig. 8c). The mineral assemblage in the control experiment at 955°C (EQ955°C120MPa1h) was augite, Fe-Ti oxides, and apatite; however, most of their groundmass rarely contained pyroxene crystals (Fig. 8c and 8d). The crystallization of Fe-Ti oxides occurred especially at the sample surfaces (i.e., near the capsule walls and vesicles), and pyroxene (augite) showed a similar tendency (Fig. 8c). The pyroxene crystallized locally, and their size and number density showed large variations among locations. The subsequent decompression induced crystallization of augite and plagioclase; however, the large part of groundmass remained glassy, which indicates that the decompression-induced crystallization in these experiments mainly occurred as heterogeneous nucleation on the pre-existing crystals and/or the sample surfaces. On the other hand, apatite crystals were distributed throughout the sample, and their size was generally a few hundred nanometers wide in all the run products.

Figures 9 and 10 show the BSE images of groundmass pyroxenes and the distributions of trachts for the experiments at 955 °C. The proportion of crystals in each tracht varied in association with the experimental conditions (Fig. 10; Table 6). At the initial condition (EQ955°C120MPa1h), the pyroxene trachts were mainly octagonal and hexagonal (Figs. 9a and 10a). Although the decompression to 50 MPa

(SSD955°C50MPa1h) resulted in the predominance of octagonal tracht (Figs. 9b and 10b), further decompression reproduced hexagonal and parallelogrammatic trachts. The texture at 20 MPa (SSD955°C20MPa1h) was characterized by all the types of tracht (Figs. 9c and 10c), and the proportion of hexagonal tracht increased at the expense of octagonal one at 10 MPa (SSD955°C10MPa1h; Figs. 9d and 10d). The texture of SSD955°C5MPa1h was characterized by parallelogrammatic tracht (Figs. 9e and 10e). We did not confirm obvious correlation between tracht and local number densities in these runs. The CD experiment (CD955°C5MPa3h) was characterized by octagonal and hexagonal trachts (Figs. 9f and 10f). In the CD experiment, the crystals tended to be hexagonal and parallelogrammatic in the regions where their number density was relatively low, although the proportion of parallelogrammatic crystals was still low. Note that the TEM analyses confirmed that each type of tracht comprises the same prismatic faces as those observed in the pumice samples.

## Discussion

### Estimating the pyroxene liquidus for the Sakurajima melt

Estimation of the value of  $\Delta T_{\text{eff}}$  for each experiment required the pyroxene liquidus in the Sakurajima melt. In our experiments at 955 °C, most parts of the groundmass were glassy (Fig. 8d). Because superliquidus heating hinders subsequent crystal nucleation (e.g., Sato, 1995; Pupier et al.,

2008; Waters et al., 2015; Arzilli et al., 2015; First et al., 2020; Matsumoto et al., 2023), this result indicates that the pyroxene crystals and any crystal nuclei were dissolved in the melt at the initial conditions (i.e., 120 MPa and 955 °C), and that the liquidus temperature of Cpx in this composition at 120 MPa is below 955 °C. In contrast, the control experiment at 935 °C contained ubiquitous groundmass pyroxene (Fig. 8b). Therefore, the Cpx liquidus temperature at 120 MPa must be within the range 935–955 °C in our experiments.

In contrast, Sekine et al. (1979) estimated pyroxene and plagioclase liquidi by performing phase equilibrium experiments on the groundmass of the pumice from the 1914 Sakurajima eruption under water-saturated conditions at the NNO buffer. Their result at 100 MPa shows that the liquidus temperature of Cpx is below 930 °C, whereas our results show that the liquidus temperature of Cpx is within 935–955 °C at 120 MPa. This difference can be attributed to the Mg content of the melt. The MgO content of the groundmass glass in the Sakurajima pumice used by Sekine et al. (1979; 0.75 wt%) is lower than that in our experiments (around 1.0 wt%; Tables 2 and 3), a discrepancy also reported by Nakamura (2006). Therefore, we thermodynamically modeled the effect of MgO content on the Cpx liquidus using MELTS (Ghiorso and Sack, 1995; Gualda et al., 2012; Ghiorso and Gualda, 2015). We used rhyolite-MELTS in MELTS\_Excel (Gualda and Ghiorso, 2015) and obtained pyroxene liquidus curves for two melt compositions: the starting materials in our

experiments and those used by Sekine et al. (1979) (Fig. 11). This calculation showed that increasing the MgO content of the melt by ~0.3 wt% from that used by Sekine et al. (1979) raises the liquidus temperature by ~25 °C. Additionally, the experimental liquidus can be reproduced by adding 55 °C to the calculated liquidus (Fig. 11).

#### **Magma ascent process deduced from white pumice**

As reported by Nakamura (2006), the white pumice samples, especially Type-1, were characterized by the low crystal number density (Fig. 4a). Assuming that the pyroxenes in the Type-1 pumice crystallized under higher  $\Delta T_{\text{eff}}$ , their shapes might be more elongated than those in other pumice samples (e.g., Kouchi et al., 1983; Shea and Hammer, 2013). On the contrary, the Type-1 pumice had the lowest degree of pyroxene elongation (i.e.,  $L/S$  ratio) of the three samples (Table 5). This might be attributed to the estimation uncertainty due to the insufficient number of analyzed crystals (< 200; Mangler et al., 2022). Although an underestimated value of 3D aspect ratio can affect resultant CSDs, the crystal number density is still the lowest in the Type-1 pumice. When using the 3D aspect ratio of 1.00:1.40:10.00 instead of the estimated value (1.00:1.40:3.60), the population densities ( $N$ ) decrease by approximately one natural log unit in the  $S$ -plot CSD (Fig. 4), and the CSD

shapes are largely maintained. Therefore, the uncertainty in 3D aspect ratio has negligible impact on the following discussion.

The crystallization experiments demonstrated that Cpx tracht changes with the extent of decompression (Fig. 10). Based on the liquidus curve (Fig. 11), the estimated  $\Delta T_{\text{eff}}$  at 955 °C was approximately 35, 75, 95, and 110 °C at 50, 20, 10, and 5 MPa, respectively. Therefore, our results indicate that the Cpx tracht changes from octagonal to hexagonal and then to parallelogrammatic with increasing  $\Delta T_{\text{eff}}$  (Fig. 10). As shown in the tracht-specific CSDs (Fig. 4a and 4b), the tracht of groundmass pyroxenes in the white pumice samples (Type-1 and 2) changes from octagonal to parallelogrammatic with decreasing size. Assuming that smaller crystals were crystallized at shallower part in the conduit, the tracht-specific CSDs indicate that pyroxene crystallization in the white pumice samples changed from octagon- and hexagon-dominant to hexagon- and parallelogram-dominant during syneruptive ascent (Fig. 4a and 4b). Our experimental results (Fig. 10) imply that the magma that produced the white pumice experienced increasing  $\Delta T_{\text{eff}}$  due to accelerated ascent in the conduit. The concave-up shapes of the conventional CSDs (Fig. 4a and 4b) consistently show the accelerated nucleation of crystals, indicating the increase of  $\Delta T_{\text{eff}}$  (e.g., Armienti et al., 1994; Marsh, 1998; Armienti, 2008). This inference is also supported

by the Al-increasing zonation (Fig. 5c and 6c) because the enrichment of pyroxene in Al indicates rapid growth (e.g., Dymek and Gromet, 1984; Mollo et al., 2013; Masotta et al., 2020). Therefore, we conclude that the magma that produced the white pumices ascended sufficiently fast and reached the surface before crystallization could proceed to a significant extent, attaining a high  $\Delta T_{\text{eff}}$ .

The minimum decompression rate of the white pumice magma can be constrained from our CD experiment. Decompression at a constant rate of  $\sim 0.01$  MPa/s mainly produced octagonal and hexagonal Cpx crystals (Fig. 10f), whereas rapid decompression in a single step ( $> 10$  MPa/s) mainly produced parallelogrammatic crystals (Fig. 10e). Therefore, ascent must have been faster than  $0.01$  MPa/s, which is plausible when compared with rates estimated for Vulcanian eruptions:  $0.28\text{--}0.87 \times 10^{-2}$  MPa/s for the 1975–1987 eruptions by Miwa et al. (2009; based on the microlite number density water exsolution rate meter developed by Toramaru et al., 2008);  $0.7\text{--}7.8 \times 10^{-2}$  MPa/s for the 2010 eruption by Miwa and Geshi (2012; based on cracked texture of microphenocrysts).

Because plagioclase and Cpx are the main phenocryst phases in the natural pumices (Nakamura, 2006), the crossing of their liquidus should indicate the magma storage conditions prior to the 1914 Sakurajima eruption. Our Cpx liquidus crosses that of

plagioclase from Sekine et al. (1979) around 50 MPa and 990 °C (Fig. 11), consistent with previous estimates of the storage pressure (20–72 MPa based on the water contents of phenocryst-hosted melt inclusions; Araya et al., 2019) and temperature (940–1010 °C based on the pyroxene thermometry; Matsumoto and Nakamura, 2017). Given that the pumices were quenched at ~5 MPa, based on the water content in the groundmass (ca. 0.4 wt%; Nakamura, 2006), decompression from 50 MPa yields  $\Delta T_{\text{eff}} \approx 75$  °C under water-saturated conditions. On the other hand, the similarity in tracht proportions (Figs. 4 and 10) indicates that the maximum  $\Delta T_{\text{eff}}$  attained during ascent was probably between 70 and 110 °C, which appears to be higher than the estimation based on melt dehydration alone ( $\Delta T_{\text{eff}} \approx 75$  °C). Therefore, the parallelogrammatic tracht common in the white pumices probably resulted from the adiabatic cooling associated with volatile exsolution and expansion (Mastin and Ghiorso, 2001; La Spina et al., 2015; Arzilli et al., 2019). Microlite crystallization may also release latent heat (e.g., Couch et al., 2003b; Blundy et al., 2006; La Spina et al., 2015), but this is unlikely given the low degree of crystallization.

### Implication

The tracht-specific CSD of the gray pumice sloped gently and was dominated by



octagonal pyroxenes spanning a broader size distribution than pyroxenes in the white pumices (Fig. 4c), indicating relatively prolonged crystallization under lower  $\Delta T_{\text{eff}}$ . The decrease in Al content with growth (Fig. 7c) supports the slow or declining growth. In addition, the slightly rounded shapes of the groundmass crystals (Figs. 2 and 7) suggest resorption by heating. We also observed a Cpx microlite with parallelogrammatic zoning, implying that this crystal nucleated under high  $\Delta T_{\text{eff}}$ . These textures indicate that the gray pumice magma initially nucleated microlites under high  $\Delta T_{\text{eff}}$  before prolonged crystallization under decreasing  $\Delta T_{\text{eff}}$ , and that the magma temperature surpassed the pyroxene liquidus just before quenching.

Assuming that the gray pumice originated from portions of the ascending magma nearer to the conduit walls (Nakamura, 2006), two sources of heat are plausible: the release of latent heat by crystallization (Couch et al., 2003a; Blundy et al., 2006) and shear heating (Mastin, 2005; Costa et al., 2007; Hale et al., 2007). Since the crystallization increases magma temperature by 2.3–3.2 °C per 1 % (Couch et al., 2003a; Blundy et al., 2006), the temperature rise by the latent heat release can be up to 50 °C by assuming 15 % crystallization of pyroxene + plagioclase (Table 4; Nakamura, 2006). Moreover, shear heating can increase magma temperature by several tens of degrees (e.g., Mastin, 2005;

Costa et al., 2007; Hale et al., 2007). The weak orientation of the groundmass crystals (Fig. 2) suggests shear deformation. Furthermore, shear heating is enhanced in the presence of crystals (Mueller et al., 2010). Therefore, both heating processes were probably significant during groundmass crystallization in the gray pumice, resulting in the rounding of the groundmass crystals.

Finally, the textural differences between the white and gray pumices indicate that they probably originated from the same magma but different radial positions within the conduit (e.g., Polacci, 2005; Nakamura, 2006): the white pumice represents magma that ascended rapidly in the center of the conduit, whereas the gray pumice originated from magma nearer the conduit walls (Fig. 12). Magma flows more slowly in the vicinity of the conduit walls than in the center of the conduit because of wall-generated friction (e.g., Gonnermann and Manga, 2003; Mastin, 2005; Costa et al., 2007; Hale and Mühlhaus, 2007). The low vesicularity of gray pumice is attributed to the efficient degassing near the walls due to the longer duration for gas escape and the shear deformation (e.g., Stasiuk et al., 1996; Okumura et al., 2009, 2013). The shear deformation also facilitates crystallization (e.g., Kouchi et al., 1986; Kolzenburg et al., 2018; Vetere et al., 2021), favoring the nucleation of groundmass crystals.

461           Our investigation demonstrates the value of pyroxene trach analyses for  
462 unraveling magma ascent dynamics via the evolution of  $\Delta T_{\text{eff}}$ . Because this method is  
463 applicable to crystal-poor glassy samples and achieves high temporal resolution when  
464 crystals are zoned, further such trach analyses should contribute to a better understanding  
465 of syneruptive magma ascent dynamics.

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### Figure captions

**Figure 1.** Classification of pyroxene trachts. **(a)** A 3D shape of pyroxene crystal with octagonal tracht. **(b)** The shape variation of cross section depending on trachts. **(c)** Cross sections of groundmass pyroxene crystals were classified based on the number of faces between a given pair of parallel faces (indicated by circles). **(d)** Incomplete shapes were classified similarly if a pair of parallel faces was present. **(e)** Individual segments of attached crystals were classified in a similar manner.

**Figure 2.** BSE images of groundmass of the Sakurajima pumice samples. Groundmass crystal phases are Fe-Ti oxides, pyroxene, and plagioclase (in order of decreasing brightness). The brightness of the groundmass glass is similar to that of plagioclase. Black regions are vesicles. The images in the right column show the groundmass pyroxene crystals with trachts characteristic of each sample. Abbreviations: Pl = plagioclase; Px = pyroxene; Ox = Fe-Ti oxides.

**Figure 3.** Estimation of 3D aspect ratio of groundmass pyroxenes by ShapeCalc (Mangler



et al., 2022). The best-fitted 300 estimates are plotted in Zingg diagram (Zingg, 1935) with the colors based on the cumulative goodness of fit ( $R_c^2$ ). The uncertainties ( $1\sigma$ ) on the best shape estimate are provided as black bars. Note that the plotted distributions do not necessarily represent the actual variation of crystal shapes: we did not find tabular cross sections of pyroxene through the SEM observation, but most pyroxene crystals seemed to have acicular shapes in 3D as far as we observed. Abbreviations: S = short-axis length, I = intermediate-axis length, L = long-axis length.

**Figure 4.** Tracht-specific CSDs of groundmass pyroxenes in the Sakurajima pumice samples. The conventional CSDs (i.e., including all trachts) are plotted in black for comparison. The size range shown is from 0.16 to 6.31  $\mu\text{m}$  in width (short-axis length). The total number of analyzed crystals and the proportions of crystals in each tracht are shown at top-right in each panel.

**Figure 5.** Internal texture of a hexagonal pyroxene nanolite in the Type-1 pumice.

(a) An ADF-STEM image, (b–d) Ca, Al, and Mg# ( $=\text{Mg}/(\text{Mg} + \text{Fe})$  in mol) compositional maps, respectively, and (e) a SAED pattern were obtained along the [001] zone axis. (f and

g) SAED patterns were obtained from the points 1 and 2 in (a), at another orientation. (g)  
The SAED pattern from Cpx region showed weak reflections of  $P2_1/c$  in addition to those  
of  $C2/c$  (i.e., the  $hkl$  reflections of “ $h + k = \text{even}$ ”). Abbreviations: Opx, orthopyroxene;  
Cpx, clinopyroxene.

**Figure 6.** Internal texture of a parallelogrammatic clinopyroxene nanolite in the Type-2  
pumice.

(a) An ADF-STEM image, (b–d) Ca, Al, and Mg# compositional maps, respectively, and  
(e) a SAED pattern were obtained along the [001] zone axis. SAED patterns were also  
obtained at another orientation from (f) the center and (g) the left side of the crystal. There  
are weak reflections of  $P2_1/c$  in (g). (h) A dark-field image with reflection  $g = 0\bar{3}\bar{1}$  shows  
anti-phase boundaries in the pigeonite (Pgt) domain.

**Figure 7.** Internal texture of an octagonal clinopyroxene microlite in the Type-3 pumice.

(a) An ADF-STEM image and (b–d) Ca, Al, and Mg# compositional maps, respectively,  
were obtained along the [001] zone axis. Note that the Al-rich spot within the crystal is  
groundmass glass. There is a parallelogrammatic zonation inside the rounded octagonal

outline.

**Figure 8.** Backscattered electron (BSE) images of control experiments at 120 MPa. The run products at 935 °C (EQ935°C120MPa1h; **a** and **b**) and 955 °C (EQ955°C120MPa1h; **c** and **d**). The control experiment at 955 °C resulted in inhomogeneous texture: Fe-Ti oxides and pyroxene crystallized locally near the sample surface (upper-left part in **c**) whereas most of the groundmass was glassy (**d**). Abbreviations: Px, pyroxene; Ox, Fe-Ti oxides; Pl, plagioclase.

**Figure 9.** BSE images of groundmass pyroxene crystals in run products at 955 °C. The control experiment at 120 MPa (**a**; EQ955°C120MPa1h). The single-step decompression experiments at the final pressure of 50 (**b**; SSD955°C50MPa3h), 20 (**c**; SSD955°C20MPa3h), 10 (**d**; SSD955°C10MPa3h), and 5 MPa (**e**; SSD955°C5MPa3h). The continuous decompression experiment from 120 to 5 MPa (**f**; CD955°C5MPa3h). Abbreviations: Px, pyroxene; Ap, apatite.

**Figure 10.** Tracht-specific CSDs of pyroxenes produced in experiments at 955 °C. (**a**)

EQ955°C120MPa1h, (b) SSD955°C50MPa3h, (c) SSD955°C20MPa3h, (d) SSD955°C10MPa3h, (e) SSD955°C5MPa3h, and (f) CD955°C5MPa3h. The number of crystal sections ( $n$ ) is plotted on a logarithmic scale against cross-sectional width for each size interval. The size range shown is from 0.20 to 4.20  $\mu\text{m}$  in width. The total number of analyzed crystals and the proportions of crystals in each tracht are shown at top-right in each panel.

**Figure 11.** Estimated magma storage conditions and liquidus curves for the 1914 Sakurajima magma. Cpx (black), Opx (gray), and plagioclase liquid (blue) reported by Sekine et al. (1979) are shown alongside pyroxene liquid for the melt compositions of Sekine et al. (1979; yellow) and this study (red) modeled using MELTS. The calculated curves were shifted by 55 °C from the MELTS calculations to match the experimental results. A black horizontal bar represents the range of possible Cpx liquidus temperatures constrained by the control experiments in this study (i.e., 935–955 °C). The shaded area indicates the magma storage conditions estimated based on pyroxene thermometry (Matsumoto and Nakamura, 2017) and phenocryst-hosted melt inclusion  $\text{H}_2\text{O}$  contents (Araya et al., 2019).

798

799 **Figure 12.** Syneruptive magma ascent dynamics in the conduit. **(a)** Schematic illustration  
800 of the horizontal variation of magma ascent in conduit. **(b and c)** Qualitative illustration of  
801  $\Delta T_{\text{eff}}$  evolution for the gray and white pumice magmas. The magma that ascended rapidly in  
802 the center of the conduit originated the white pumice, whereas the magma that ascended in  
803 the vicinity of the conduit walls produced the gray pumice. Dashed lines in **(a)** are the  
804 isochronal contours of ascending magma at  $t_0$ – $t_3$ . White circles and black objects represent  
805 vesicles and groundmass pyroxene crystals, respectively. The magmatic processes which  
806 increase or decrease  $\Delta T_{\text{eff}}$  are written in blue or red characters, respectively. After the  
807 nucleation delay ( $t > t_2$ ), pyroxene nucleation occurred under high  $\Delta T_{\text{eff}}$ . Then, the white  
808 pumice magma was quenched, whereas the gray pumice magma experienced prolonged  
809 crystallization under the  $\Delta T_{\text{eff}}$  lowered by heating processes (i.e., the release of latent heat  
810 of crystallization and the shear heating).

811

812

**Tables**

813 Table 1. The experimental conditions and resultant textures.

| Name                | T<br>(°C) | Path | P <sub>i</sub><br>(MPa) | P <sub>f</sub><br>(MPa) | duration<br>(h) | GM phase <sup>a</sup> | Main Px<br>tracht |
|---------------------|-----------|------|-------------------------|-------------------------|-----------------|-----------------------|-------------------|
| EQ935°C120<br>MPa1h | 935       | EQ   | 120                     | 120                     | 1               | Px, Pl, Ox, Ap        | 8, 6              |
| EQ955°C120<br>MPa1h | 955       | EQ   | 120                     | 120                     | 1               | Px, Ox, Ap            | 8, 6              |
| SSD955°C50<br>MPa3h | 955       | SSD  | 120                     | 50                      | 3               | Px, Pl, Ox, Ap        | 8                 |
| SSD955°C20<br>MPa3h | 955       | SSD  | 120                     | 20                      | 3               | Px, Pl, Ox, Ap        | 8, 6, 4           |
| SSD955°C10<br>MPa3h | 955       | SSD  | 120                     | 10                      | 3               | Px, Pl, Ox, Ap        | 6                 |
| SSD955°C5<br>MPa3h  | 955       | SSD  | 120                     | 5                       | 3               | Px, Pl, Ox, Ap        | 4                 |
| CD955°C5M<br>Pa3h   | 955       | CD   | 120                     | 5                       | 3               | Px, Pl, Ox, Ap        | 8, 6              |

<sup>a</sup> Abbreviation: Px, pyroxene; Pl, plagioclase; Ox, Fe-Ti oxide; Ap, apatite.

814

815 Table 2. Average glass compositions of Sakurajima pumices.

|                                | Type-1       | Type-2       | Type-3       |
|--------------------------------|--------------|--------------|--------------|
| (wt%)                          | n = 98       | n = 55       | n = 41       |
| SiO <sub>2</sub>               | 68.80 (0.27) | 70.01 (0.31) | 73.41 (0.24) |
| TiO <sub>2</sub>               | 0.80 (0.07)  | 0.78 (0.08)  | 0.90 (0.06)  |
| Al <sub>2</sub> O <sub>3</sub> | 13.94 (0.13) | 13.64 (0.15) | 11.84 (0.14) |
| FeO                            | 4.62 (0.19)  | 4.29 (0.14)  | 4.08 (0.16)  |
| MnO                            | 0.11 (0.12)  | 0.10 (0.12)  | 0.04 (0.09)  |
| MgO                            | 1.07 (0.05)  | 0.89 (0.06)  | 0.48 (0.07)  |
| CaO                            | 3.70 (0.14)  | 3.34 (0.14)  | 1.96 (0.06)  |
| Na <sub>2</sub> O              | 4.35 (0.12)  | 4.25 (0.14)  | 3.90 (0.11)  |
| K <sub>2</sub> O               | 2.44 (0.07)  | 2.57 (0.07)  | 3.19 (0.07)  |
| P <sub>2</sub> O <sub>5</sub>  | 0.17 (0.11)  | 0.13 (0.12)  | 0.21 (0.11)  |
| Total                          | 100          | 100          | 100          |

NOTES: Values in parentheses are standard deviations.

Oxide concentrations were recalculated to total 100% by cation balance.

816

Table 3. Average glass compositions of run products.

| Starting material              | EQ955°C<br>120MPa1h<br><i>n</i> = 39 | EQ945°C<br>120MPa1h<br><i>n</i> = 30 | EQ935°C<br>120MPa1h<br><i>n</i> = 29 | SSD955°C<br>50MPa3h<br><i>n</i> = 33 | SSD955°C<br>20MPa3h<br><i>n</i> = 37 | SSD955°C<br>10MPa3h<br><i>n</i> = 40 | SSD955°C<br>5MPa3h<br><i>n</i> = 45 | CD955°C<br>5MPa3h<br><i>n</i> = 36 |
|--------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------|-------------------------------------|------------------------------------|
| (wt%) <sup>a</sup>             | <i>n</i> = 45                        | <i>n</i> = 39                        | <i>n</i> = 30                        | <i>n</i> = 29                        | <i>n</i> = 33                        | <i>n</i> = 37                        | <i>n</i> = 40                       | <i>n</i> = 45                      |
| SiO <sub>2</sub>               | 68.99 (0.23)                         | 71.06 (0.42)                         | 71.22 (0.38)                         | 70.58 (0.51)                         | 70.35 (0.32)                         | 71.18 (0.49)                         | 70.96 (0.39)                        | 71.47 (0.40)                       |
| TiO <sub>2</sub>               | 0.79 (0.08)                          | 0.73 (0.10)                          | 0.74 (0.11)                          | 0.73 (0.15)                          | 0.67 (0.07)                          | 0.66 (0.14)                          | 0.64 (0.15)                         | 0.64 (0.11)                        |
| Al <sub>2</sub> O <sub>3</sub> | 13.91 (0.11)                         | 14.43 (0.13)                         | 14.42 (0.11)                         | 14.23 (0.14)                         | 14.3 (0.11)                          | 14.49 (0.13)                         | 14.5 (0.15)                         | 14.59 (0.13)                       |
| FeO                            | 4.47 (0.14)                          | 2.15 (0.19)                          | 2.07 (0.16)                          | 3.24 (0.42)                          | 3.33 (0.13)                          | 2.08 (0.48)                          | 2.27 (0.24)                         | 1.79 (0.19)                        |
| MnO                            | 0.07 (0.11)                          | 0.06 (0.10)                          | 0.1 (0.11)                           | 0.09 (0.12)                          | 0.07 (0.11)                          | 0.11 (0.12)                          | 0.06 (0.10)                         | 0.05 (0.09)                        |
| MgO                            | 1.05 (0.05)                          | 0.89 (0.10)                          | 0.92 (0.07)                          | 0.93 (0.07)                          | 0.91 (0.09)                          | 0.87 (0.17)                          | 0.85 (0.18)                         | 0.84 (0.17)                        |
| CaO                            | 3.57 (0.10)                          | 3.49 (0.21)                          | 3.49 (0.14)                          | 3.42 (0.14)                          | 3.33 (0.14)                          | 3.33 (0.23)                          | 3.36 (0.21)                         | 3.33 (0.31)                        |
| Na <sub>2</sub> O              | 4.47 (0.11)                          | 4.47 (0.44)                          | 4.4 (0.16)                           | 4.14 (0.17)                          | 4.34 (0.07)                          | 4.49 (0.38)                          | 4.54 (0.10)                         | 4.49 (0.13)                        |
| K <sub>2</sub> O               | 2.50 (0.06)                          | 2.57 (0.10)                          | 2.51 (0.07)                          | 2.45 (0.06)                          | 2.56 (0.07)                          | 2.58 (0.17)                          | 2.64 (0.09)                         | 2.62 (0.10)                        |
| P <sub>2</sub> O <sub>5</sub>  | 0.18 (0.12)                          | 0.16 (0.13)                          | 0.13 (0.16)                          | 0.19 (0.14)                          | 0.15 (0.14)                          | 0.21 (0.13)                          | 0.19 (0.13)                         | 0.17 (0.11)                        |
| Total                          | 100                                  | 100                                  | 100                                  | 100                                  | 100                                  | 100                                  | 100                                 | 100                                |

NOTES: Values in parentheses are standard deviations.

Oxide concentrations were recalculated to total 100% by cation balance.



818 Table 4. Texture of the Sakurajima pumice samples.

| Sample | Number<br>of<br>regions | Analyzed area       |                         | Px<br>content<br>(area%) | Number of crystal cross sections |      |       |      |       |       |
|--------|-------------------------|---------------------|-------------------------|--------------------------|----------------------------------|------|-------|------|-------|-------|
|        |                         | ( $\mu\text{m}^2$ ) | (vesicle%) <sup>a</sup> |                          | All                              | Oct. | Hept. | Hex. | Pent. | Para. |
| Type-1 | 4                       | 1,836,593           | 75.0                    | 0.01                     | 90                               | 1    | 1     | 21   | 8     | 30    |
| Type-2 | 1                       | 106,891             | 79.8                    | 0.43                     | 559                              | 6    | 7     | 190  | 40    | 152   |
| Type-3 | 2                       | 49,054              | 67.1                    | 2.47                     | 710                              | 264  | 2     | 1    | 0     | 0     |

<sup>a</sup> The percentage of vesicles in the analyzed rectangle area.

819

820 Table 5. Parameters used in CSDCorrections.

| Sample | 3D aspect ratio |            | Roundness | Size scale<br>length               |
|--------|-----------------|------------|-----------|------------------------------------|
|        | <i>S:I:L</i>    | <i>Rc2</i> |           | (Bins per<br>decade <sup>a</sup> ) |
| Type-1 | 1:1.40:3.60     | 0.997      | 0.5       | 5                                  |
| Type-2 | 1:1.75:6.00     | 0.984      | 0.5       |                                    |
| Type-3 | 1:1.30:4.60     | 0.999      | 0.7       |                                    |

<sup>a</sup> Logarithmic base-10 size scale.

821

822 Table 6. Tracht analyses on run products at 955 °C.

| Sample |                      | Analyzed<br>area<br>( $\mu\text{m}^2$ ) | Numbers of pyroxene crystals |          |         |          |                   |       |
|--------|----------------------|-----------------------------------------|------------------------------|----------|---------|----------|-------------------|-------|
| Path   | P <sub>f</sub> (MPa) |                                         | Octagon                      | Heptagon | Hexagon | Pentagon | Parallelo<br>gram | Other |
| EQ     | 120                  | 6635                                    | 115                          | 23       | 50      | 5        | 2                 | 49    |
| SSD    | 50                   | 13614                                   | 114                          | 6        | 3       | 1        | 0                 | 36    |
| SSD    | 20                   | 15945                                   | 37                           | 18       | 30      | 16       | 33                | 34    |
| SSD    | 10                   | 23844                                   | 13                           | 13       | 40      | 22       | 11                | 23    |
| SSD    | 5                    | 15905                                   | 2                            | 2        | 13      | 38       | 173               | 31    |
| CD     | 5                    | 15144                                   | 64                           | 9        | 54      | 10       | 4                 | 51    |

823

**Figure 11**

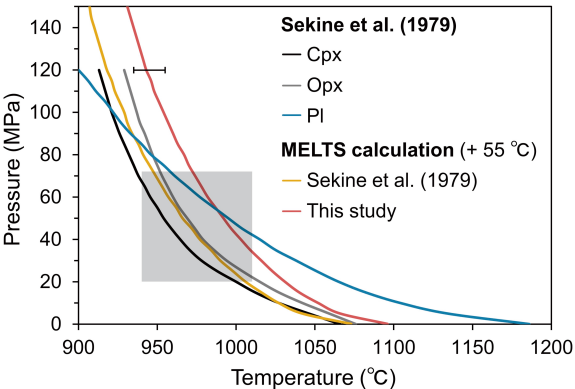
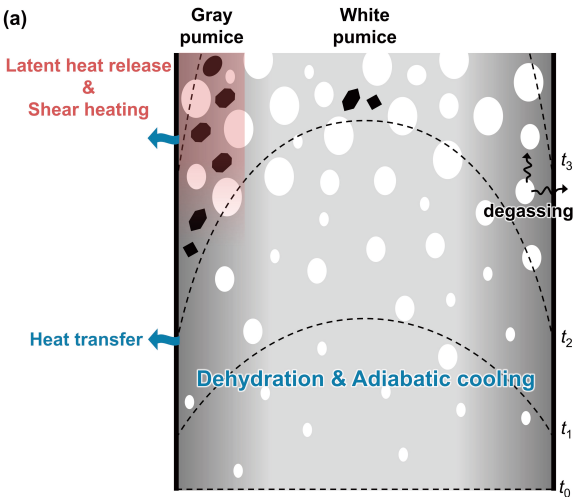
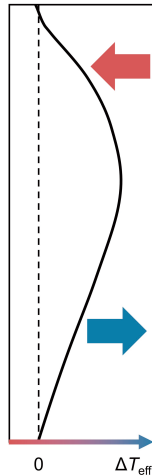


Figure 12

(a)



(b) Gray pumice



(c) White pumice

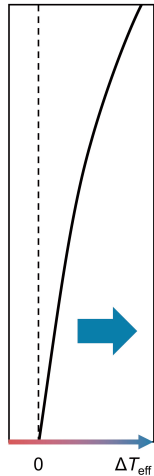


Figure 1

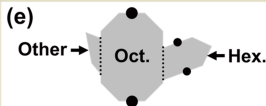
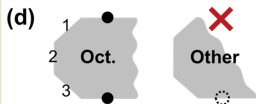
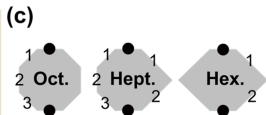
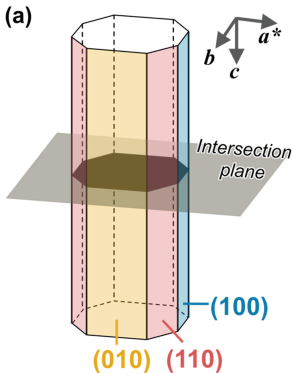
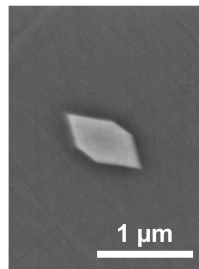
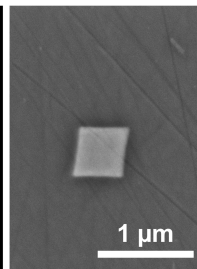
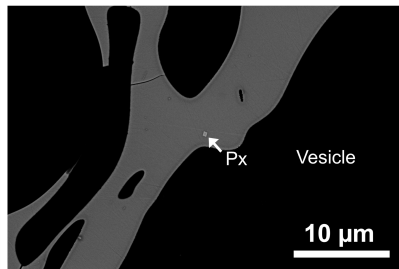
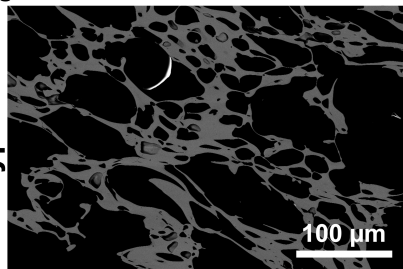
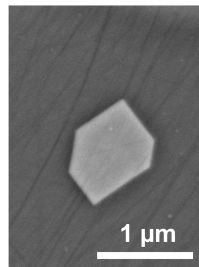
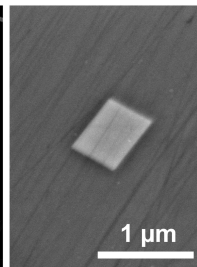
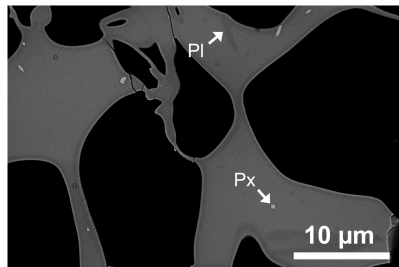
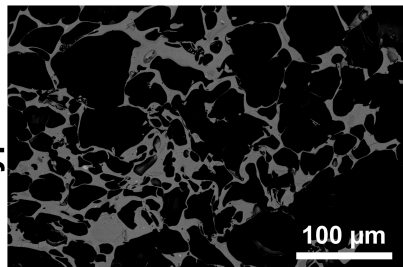


Figure 2

Type-1



Type-2



Type-3

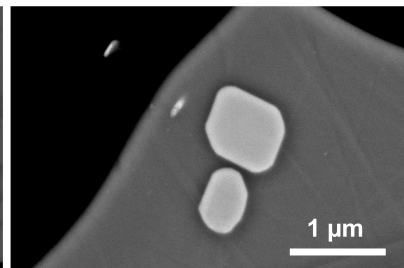
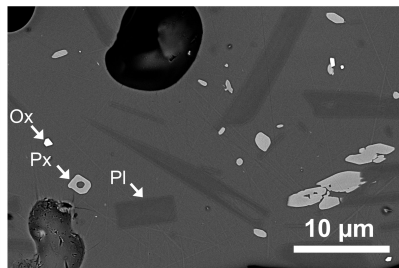
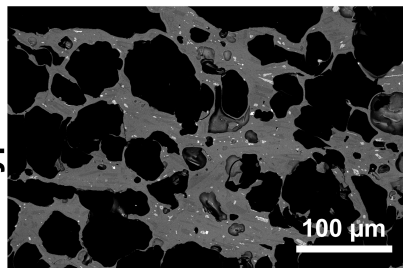
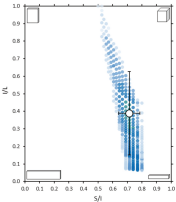
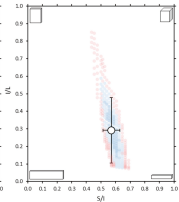


Figure 3

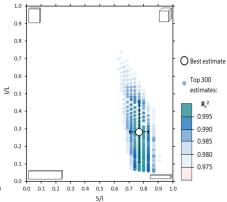
(a) Type-1



(b) Type-2



(c) Type-3



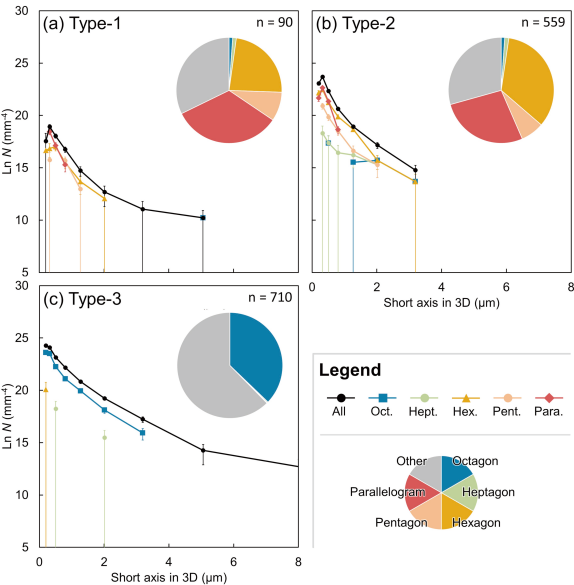
**Figure 4**



Figure 5

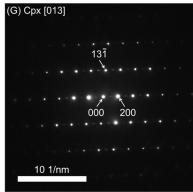
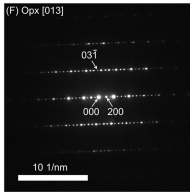
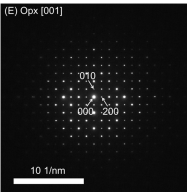
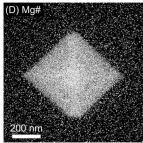
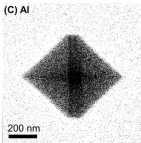
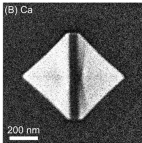
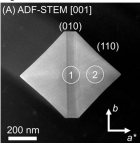
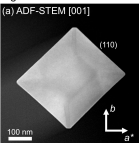
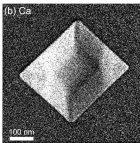


Figure 6

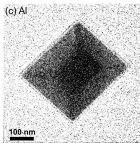
(a) ADF-STEM [001]



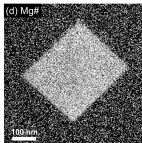
(b) Ca



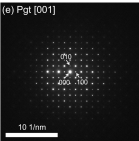
(c) Al



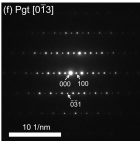
(d) Mg#



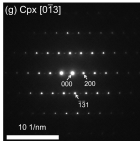
(e) Pgt [001]



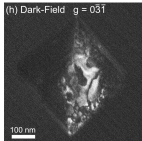
(f) Pgt [013]



(g) Cpx [013]

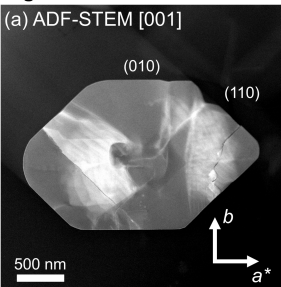


(h) Dark-Field  $g = 0\bar{3}\bar{1}$

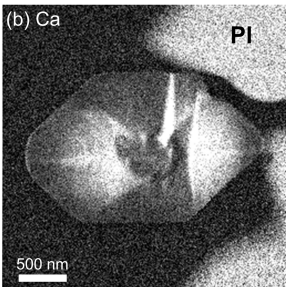


**Figure 7**

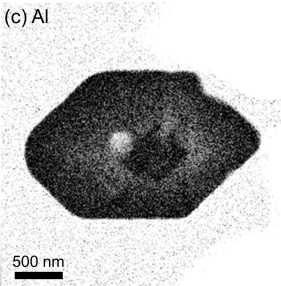
(a) ADF-STEM [001]



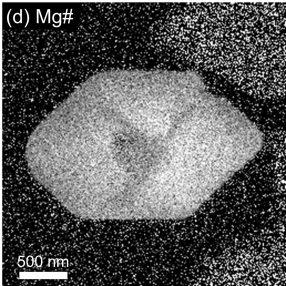
(b) Ca



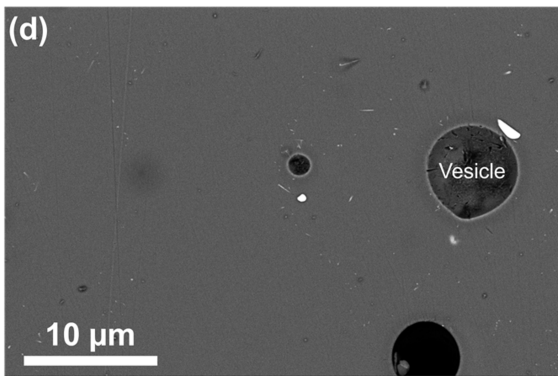
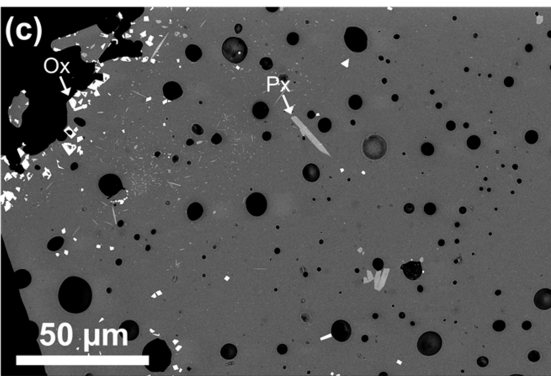
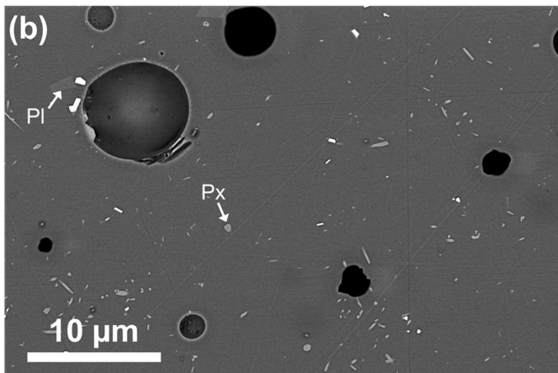
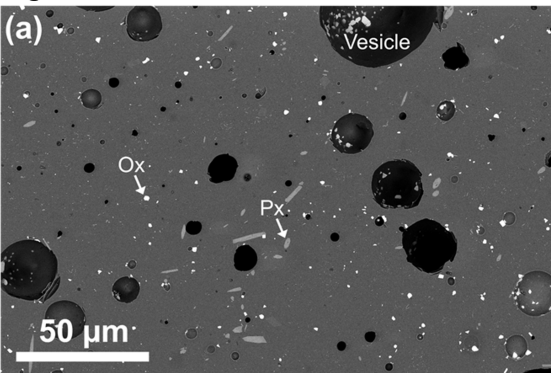
(c) Al



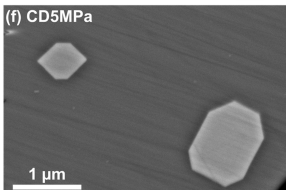
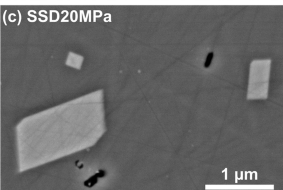
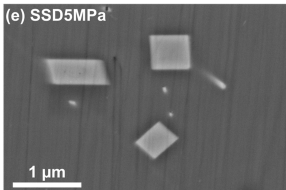
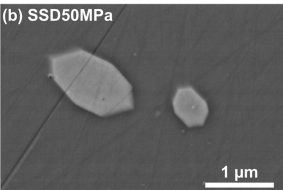
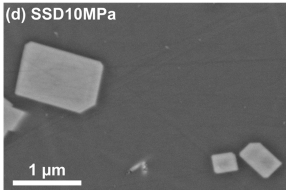
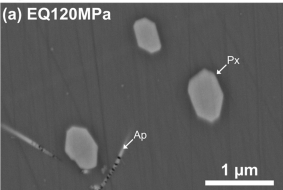
(d) Mg#

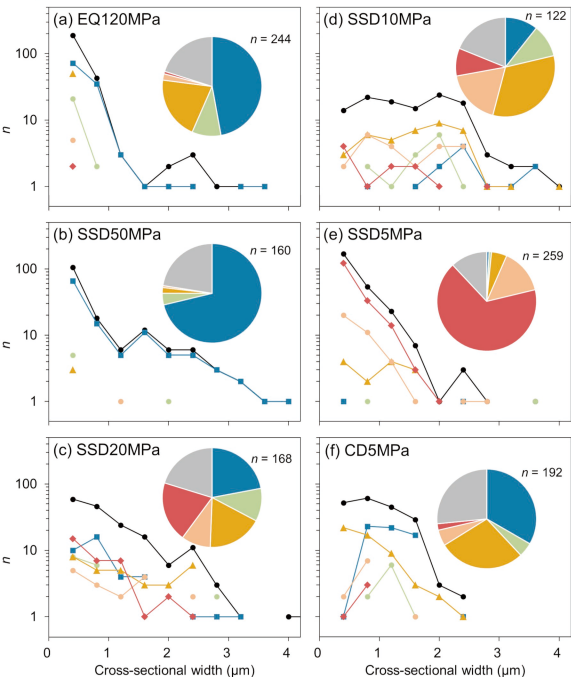


**Figure 8**



**Figure 9**



**Figure 10****Legend**

- All
- Octagon
- Heptagon
- ▲ Hexagon
- Pentagon
- ◆ Parallelogram

