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# Groundmass pyroxene crystal habits (trachts) record syneruptive magma dynamics in 3 glassy pyroclasts 4 5 Names: Shota H. Okumura<sup>1\*</sup>, Satoshi Okumura<sup>2</sup>, and Akira Miyake<sup>1</sup> 6 Affiliations: 7 <sup>1</sup>Department of Geology and Mineralogy, Division of Earth and Planetary Sciences, 8 Graduate School of Science, Kyoto University, 9 10 Kitashirakawaoiwake-cho, Sakyo-ku, Kyoto 606-8502, Japan <sup>2</sup> Division of Earth and Planetary Materials Science, Department of Earth Science, 11 Graduate School of Science, Tohoku University, 12 6-3 Aoba, Aramaki-aza, Aoba, Sendai 980-8578, Japan 13 \*Corresponding author: S.H. Okumura, <u>okumura@kueps.kyoto-u.ac.jp</u> 14 15 16

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## Abstract

18	Explosive eruptions produce various types of pyroclasts including those contain
19	few with groundmass crystals. The textural variation such as the number density, size, and
20	volume of groundmass crystals is expected to reveal the complex magma dynamics during
21	syneruptive ascent in conduits; however, we have no quantitative method to investigate the
22	magma dynamics based on crystal texture when the pyroclasts show glassy texture with a
23	small number of microlites. Here we show that the variation of combination of
24	crystallographic faces (i.e., tracht) of groundmass pyroxene crystals enables us to derive the
25	degree of effective undercooling ( $\Delta T_{\rm eff}$ ) and magma ascent histories even from the glassy
26	pyroclasts. We conducted decompression experiments and analyzed trachts of groundmass
27	pyroxene crystals in the run products in addition to those in natural pumices from the 1914
28	Plinian eruption of Sakurajima volcano. These results show that the glassy white pumices
29	experienced higher $\Delta T_{\rm eff}$ than the crystal-rich gray pumice, and corroborated that they
30	originate from the magmas at different positions from the conduit walls. The estimate on
31	$\Delta T_{\rm eff}$ implies that the magma rapidly ascended in the center of the conduit might experience
32	cooling because of volatile exsolution and expansion.

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

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### Introduction

35 Magma ascent histories in volcanic conduits are important for understanding the evolution of eruptive activities (Cashman and Sparks, 2013; Cassidy et al., 2018). The 36 ascent histories can be recorded in the textures of groundmass crystals; therefore, the 37 investigation of groundmass texture has played an important role to reveal the magma 38 dynamics. Since the crystallization kinetics, especially nucleation rates, vary with the 39 degree of effective undercooling ( $\Delta T_{eff}$ ) (e.g., Hammer and Rutherford, 2002; Brugger and 40 41 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 42 investigate magma ascent processes. These methods are based on the theoretical premise 43 44 that the nucleation rate and the resultant number density are positively correlated with  $\Delta T_{\rm eff}$ . However, the positive correlation is not universally true. For instance, explosive eruptions 45 46 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, 47 48 2005). The glassy texture of the former is attributed to the considerably fast ascent; the 49 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., 50

51	2020; Rusiecka et al., 2020; Rusiecka and Martel, 2022). In this situation, it is difficult to
52	extract syneruptive ascent histories from the crystal number densities in glassy pyroclasts,
53	because the theoretical premise that the nucleation rate and the resultant number density are
54	positively correlated with $\Delta T_{\rm eff}$ is not valid. In such scenarios, a new indicator is required to
55	estimate $\Delta T_{\rm eff}$ from glassy groundmass with a small number of crystals.
56	Previous studies have demonstrated that crystal habits reflect $\Delta T_{eff}$ (e.g., Lofgren,
57	1974; Hammer and Rutherford, 2002; Shea and Hammer, 2013; Arzilli et al., 2022). Kouchi
58	et al. (1983) revealed experimentally that the relative growth rates for different faces of
59	clinopyroxene (Cpx) change with $\Delta T_{\rm eff}$ . Their data indicates that, with increasing $\Delta T_{\rm eff}$ , a
60	pyroxene crystal would have different combinations of crystallographic faces (i.e., tracht;
61	Sunagawa, 2005) by losing some faces. This hypothesis was confirmed by Okumura et al.
62	(2022b), in which it was experimentally revealed that the tracht of groundmass pyroxene
63	crystals changes from octagonal prisms consisting of $\{100\}$ , $\{010\}$ , and $\{110\}$ prismatic
64	faces into hexagonal prisms lacking $\{100\}$ faces as $\Delta T_{eff}$ increases. Therefore, the tracht of
65	groundmass pyroxene crystals is expected to allow us to estimate $\Delta T_{eff}$ and magma ascent
66	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

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

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## Sample description

76	Sakurajima is located on the southern rim of the Aira caldera, Kyushu,
77	Japan, with two summit vents aligned north-south: Kitadake and Minamidake. Eruptive
78	activity has been summarized by Kobayashi et al. (2013) and is abridged here. Sakurajima
79	started erupting at 26 ka, and its activity is subdivided into four stages based on tephra
80	stratigraphy and chronology: the "Older Kitadake" (26–24 ka), "Younger Kitadake" (13–5
81	ka), "Older Minamidake" (4.5–1.6 ka), and "Younger Minamidake" (since 764 AD) stages.
82	All eruptive products are andesitic to dacitic. Four large eruptions have occurred during the
83	Younger Minamidake stage: the Tenpyo-Hoji (764-766), Bunmei (1471-1476), An-ei
84	(1779-1782), and Taisho (1914-1915) eruptions. Each of these events had a similar
85	eruptive sequence: an early Plinian eruption followed by lava outflows from lateral vents.
86	In particular, the 1914–1915 Taisho eruption began with seismic activity on 11 January
87	1914, and the Plinian eruption began at the fissure vents on the western flank on 12
88	January; ten minutes later, a synchronous Plinian eruption began on the eastern flank. The
89	Plinian phase continued on both flanks for about 36 h and was followed by lava extrusion
90	and intermittent smaller explosions over the following two weeks. Additional outflows of
91	lava began on the eastern flank in February and lasted for 1.5 years.

93	structure of the present magma plumbing system (e.g., Omori, 1916; Iguchi, 2013; Iguchi et
94	al., 2013). A major magma reservoir exists at ~10 km depth beneath Aira caldera and minor
95	reservoirs are present at $\sim$ 5 km beneath the summit, from the north flank of Kitadake to
96	Minamidake (Iguchi, 2013; Iguchi et al., 2013). The diameter of the conduit beneath
97	Minamidake crater is estimated to be 300-500 m at 2 km depth and 40-60 m at the
98	shallowest depths (Iguchi et al., 2013). Araya et al. (2019) analyzed melt inclusions in
99	phenocrysts from the 1914 Sakurajima pumice and showed that the volatile saturation
100	pressure ranged from 20 to 72 MPa. They proposed that the magma was pre-charged in a
101	thick conduit at depths of 0.9-3.2 km below the surface, i.e., shallower than the minor
102	reservoirs that feed present-day Vulcanian eruptions.
103	The textural analyses of the 1914 Sakurajima Plinian pumice have been performed
104	by Nakamura (2006). The pumice clasts are classified into three types with different color,
105	vesicularity, and microlite content. Type-1 and Type-2 are white pumice with high
106	vesicularity (> 55 vol%): the former is glassy with lower modal abundance of plagioclase
107	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

109	types have similar bulk groundmass compositions (SiO <sub>2</sub> = 67–70 wt%) and the same
110	mineral assemblage. Phenocrysts (> 100 $\mu$ m in length) consist of plagioclase,
111	orthopyroxene (Opx), augite, and magnetite, while microlites (< 100 $\mu$ m) are plagioclase,
112	Opx, pigeonite, augite, and titanomagnetite.
113	We collected pumice clasts erupted by the 1914 eruption from an outcrop at 4.5
114	km east of the Minamidake summit and examined three pumices as the representative of
115	Type-1, 2, and 3.
116	
117	<b>Decompression experiments</b>
118	We coarsely crushed the Type-1 pumice clasts, removed phenocrysts under an
119	optical microscope, and collected the groundmass fragments as the starting material.
120	Aliquots of 7-9 mg of the starting material and enough water (> 5 wt%) to achieve
121	water-saturation were loaded into Au capsules (3 mm outer diameter and 1-2 cm long),
122	sealed by welding. We conducted the experiments using the cold-seal pressure vessels with
123	Ni filler rod at Tohoku University (Okumura et al., 2021). The decompression experiments
124	were conducted in different manners: single step decompression (SSD) and continuous

126	at a certain $\Delta T_{\text{eff}}$ , which allows us to investigate the relation between $\Delta T_{\text{eff}}$ and pyroxene
127	crystal habits. By changing the degree of decompression, we can modulate the applied $\Delta T_{\rm eff}$
128	during the SSD experiment. On the other hand, the CD experiment is an analogy for the
129	actual ascent of magma, where the alleviation of $\Delta T_{\rm eff}$ due to the crystallization can occur
130	simultaneously with the increase in $\Delta T_{\rm eff}$ induced by dehydration. The capsules were heated
131	in the vessels at a temperature of 955 °C under a pressure of 120 MPa (assuming the minor
132	reservoir at ~5 km depth) for 1 h and then isothermally decompressed to final pressures, $P_{\rm f}$ .
133	In the SSD experiments, the capsule was rapidly decompressed (> 10 MPa/s) and held at $P_{\rm f}$
134	of 5, 10, 20, or 50 MPa for 3 h and then quenched by dropping it into a water-cooled zone
135	in the system. The capsule in the CD experiment was decompressed to $P_{\rm f} = 5$ MPa with a
136	rate of ~0.01 MPa/s and quenched immediately at $P_{\rm f}$ in the same way. Control experiments
137	(EQ) were conducted by quenching without any decompression at temperatures of 935 or
138	955 °C to confirm the liquidus temperature at 120 MPa. The conditions and estimated $\Delta T_{eff}$
139	(discussed later) of experiments are listed in Table 1.
140	During the experiments, the oxygen fugacity ( $f_{O2}$ ) was buffered at NNO to NNO+1
141	(Okumura et al., 2021). The error in temperature is assumed to be within $\pm$ 5 °C. Since the

142 duration at the initial condition (955 °C) was restricted by the durability of pressure vessel,

143	there is the possibility that the inherent heterogeneity of the starting material remains
144	without equilibrium and affect the resultant textures of run products. For this reason, the
145	error in $\Delta T_{\text{eff}}$ could be larger than the thermal error of ± 5 °C.
146	The recovered capsules were carefully opened; however, the recovered materials
147	broke into pieces because of their high vesicularity and fragility, and lost their positional
148	relations within the capsules. The materials were then mounted in resin for polishing and
149	subsequent analyses.

151

## **Analytical methods**

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

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

163

## 164 **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

168	and at working distances of 4-10 mm. In run products, 3-12 regions were selected for
169	analysis in each sample and their BSE images were obtained at $3,500 \times$ magnification
170	(corresponding to regions ~40–60 $\mu m$ on a side and pixel sizes of ~20 nm). Using ImageJ
171	software, we measured the areas of the rectangular regions examined (i.e., the entire image)
172	and those occupied by vesicles and individual pyroxene cross sections, as well as pyroxene
173	cross-sectional widths $(w)$ , which was measured as the minor axis of the best-fit ellipse. We
174	discriminated pyroxene from other minerals based on their contrast in BSE images and
175	SEM-EDS chemical maps. In addition, BSE images of pyroxene crystals larger than 0.2 $\mu$ m
176	wide were acquired at higher magnifications (10,000–65,000×) for tracht classification.
177	In the natural pumice samples, because of the low number densities of groundmass
178	crystals, large rectangular areas were required to include a sufficient number of pyroxene
179	crystals within the BSE images (e.g., >1 mm on a side for Type-1 pumice). However, it was
180	difficult to acquire and process such a large image at a resolution adequate for the
181	measurement of crystal size and tracht. Therefore, we obtained magnified images of the
182	pyroxene crystals (magnifications of mainly $8,000-50,000\times$ and pixel sizes of $<20$ nm) at
183	an accelerating voltage of 5 kV and a working distance of 4 mm, as well as larger,
184	lower-resolution (54 nm/pixel) BSE maps of the analyzed areas at an accelerating voltage

185	of 10 kV and a working distance of 10 mm. The images of the analyzed areas were used to
186	measure vesicularity and analyze the groundmass areas (i.e., excluding vesicles), and the
187	magnified images were used to measure the sizes and trachts of groundmass pyroxene
188	crystals in the same manner as in the run products. Analyzed crystals were larger than $10^{-0.8}$
189	$\mu m$ (~0.16 $\mu m)$ wide in accordance with the size interval of crystal size distributions
190	(CSDs).

In BSE images at higher magnifications, each pyroxene cross section could be 191 192 classified into one of several tracht types based on the number of faces between pairs of parallel faces: octagonal, heptagonal, hexagonal, pentagonal, or parallelogrammatic 193 (Okumura et al. 2022b, 2023; Fig. 1). For example, the octagonal and hexagonal trachts 194 195 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 196 197 hexagonal tracht on the other (Fig. 1c). Pyroxene crystals that were difficult to classify as having a particular tracht were classified as "other". If cross sections were incomplete (e.g., 198 199 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, 200 could only be identified when the cross section had almost all of the constituent faces (Fig. 201

202 1c). When multiple crystals were attached to each other, each individual crystal was203 classified separately (Fig. 1e).

204

## 205 Acquisition of tracht-specific CSDs

We acquired the size distributions of pyroxene cross sections in each tracht group 206 (hereafter, tracht-specific CSDs). In run products, we counted the number of crystal cross 207 sections of each tracht in each 0.4-µm size interval (i.e., 0.2–0.6 µm, 0.6–1.0 µm, etc.) and 208 209 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 210 could not be represented adequately by a single value. 211 212 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 213 214 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 215 216 representative 3D aspect ratio of pyroxene crystals regardless of tracht for each sample by using ShapeCalc (Mangler et al., 2022), which compares a measured distribution of 217

218 cross-sectional aspect ratios with model distributions based on the algorithm used in

219	CSDCorrections and returns estimated 3D aspect ratios (and the cumulative goodness of fit,
220	$R_c^2$ ) for a range of shapes from 1:1:1 to 1:20:20. Although CSDCorrections yields 3D CSDs
221	expressed as a function of $L$ , we expressed the CSDs as a function of 3D short-axis lengths
222	(S, or cross-sectional width) with additional corrections (i.e., S-plot CSDs; Okumura et al.,
223	2022a). The details of the procedure for acquiring tracht-specific CSDs are described in
224	Okumura et al. (2023). The CSDs were plotted in logarithmic size intervals with five
225	intervals per decade larger than 0.1 $\mu$ m (i.e., each interval is 10 <sup>0.2</sup> times as large as the next
226	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
227	effective image resolutions restricted our analysis to crystals larger than $10^{-0.8}$ µm.

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## 229 **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 Ga<sup>+</sup> 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

236	and 77 pA for final processing. To determine mineral phases and crystallographic
237	orientations, selected-area electron diffraction (SAED) patterns were analyzed using
238	Gatan's proprietary DigitalMicrograph software and ReciPro (Seto and Ohtsuka, 2022). In
239	addition, we obtained compositional maps of pyroxene crystals by EDS analyses in
240	scanning TEM (STEM) mode. For quantitative X-ray analyses by STEM, we used the
241	$\zeta$ -factor method (Watanabe and Williams, 2006). To achieve the accurate electron beam
242	current measurements required for the $\zeta$ -factor method, appropriate calibration was
243	performed with the CCD camera beforehand. Furthermore, we acquired annular dark-field
244	STEM (ADF-STEM) images of pyroxene crystals perpendicular to their <i>c</i> -axes to observe
245	compositional zoning at higher resolution.

247	Results
248	
249	Textural variation of the Sakurajima pumice
250	Groundmass texture. As described in Nakamura (2006), groundmass crystals
251	consisted of plagioclase, clinopyroxene (Cpx) including augite and pigeonite,
252	orthopyroxene (Opx), and Fe-Ti oxide, with a trace amount of apatite. Their size range was
253	mainly 0.1–10 μm in width.
254	Figure 2 shows the variation in groundmass texture among the pumice samples.
255	The Type-1 pumice had higher vesicularity (75.0 %; Table 4), deformed vesicles, thinner
256	vesicle walls, and few groundmass crystals (Fig. 2). The Type-2 pumice showed a vesicle
257	texture similar to the Type-1 pumice but had more groundmass crystals and more-evolved
258	glass composition (SiO <sub>2</sub> $\sim$ 70 wt%, 1 % higher than Type-1; Table 2). The groundmass
259	pyroxene crystals were characterized by hexagonal and parallelogrammatic trachts in the
260	Type-1 and Type-2 white pumice samples (Fig. 2).
261	In contrast, the Type-3 pumice showed distinct textures (Fig. 2): lower vesicularity
262	(67.1 %; Table 4), less-round vesicle shapes, thicker vesicle walls, abundant groundmass
263	crystals, and the most evolved glass composition (SiO <sub>2</sub> $\sim$ 73 wt%; Table 2). In addition, the

groundmass crystals in the Type-3 pumice were loosely oriented (lateral direction in Fig. 2). 264 265 The crystals were slightly rounded, and groundmass pyroxene crystals were characterized by rounded octagonal tracht. 266 267 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 268 1.00:1.30:4.60 for the Type-1, Type-2, and Type-3 samples, respectively (Table 5). Note 269 270 that the number of crystal cross sections used for the estimation (n = 90) was fewer than the 271 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 273 274 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 275 slopes were steepest at the size range of nanolite (i.e., crystals smaller than 1 µm in width; 276 Mujin et al., 2017). In contrast, the conventional CSD in the Type-3 pumice (Fig. 4c) 277 showed a gentler slope. The Type-3 pumice contained large crystals (mainly  $< 5 \mu m$  in 278 279 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. 280

281	The tracht-specific CSDs exhibit an obvious difference between the white pumice
282	(Type-1 and Type-2) and the gray pumice (Type-3). In the white pumice samples, octagonal
283	pyroxene crystals had larger widths (mainly > 1 $\mu$ m), and parallelogrammatic ones were
284	distributed in the size range of nanolites (i.e., $< 1 \ \mu m$ in width), where the conventional
285	CSDs showed the steepest slope (Fig. 4a and 4b). Hexagonal crystals were distributed
286	throughout the size range of both microlite and nanolite. The CSD slopes of these trachts
287	were consistent with those of the conventional CSDs at the corresponding size ranges.
288	Heptagonal and pentagonal trachts, which are the transient shapes between hexagonal and
289	octagonal or parallelogrammatic trachts, exhibited the intermediate size ranges and slopes
290	(Fig. 4b). In contrast, the tracht-specific CSD of the gray pumice sample (Type-3) was
291	characterized by only the octagonal distribution with a gentle slope (Fig. 4c). Almost all of
292	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 pyroxene crystals were mainly Cpx composed of augite (*C*2/*c*) and pigeonite

298	$(P2_1/c)$ (Figs. 6 and 7), and occasionally Opx (Fig. 5). The Opx domain, when present, was
299	distributed along (100) plane at the center of crystal (Fig. 5). The center of pyroxene
300	crystals tended to be occupied by Ca-poor phases (i.e., pigeonite and Opx), and both sides
301	were generally Cpx including augite (Figs. 5-7). Pigeonite domains showed anti-phase
302	boundaries (Fig. 6h).
303	The pyroxene crystals sometimes showed concentric zonation. The pyroxene
304	crystals tended to be Al-poor at their cores (Figs. 5c, 6c, and 7c); moreover, the Al content
305	decreased again at their rims in those from the Type-3 pumice sample (Fig. 7c). In addition,
306	the pyroxene crystals from the Type-3 sample showed normal zoning in Mg# [=Mg/(Mg + $Mg$ )
307	Fe) in mol; Fig. 7d]. We found an octagonal pyroxene microlite with parallelogrammatic
308	zonation from the Type-3 pumice (Fig. 7).
309	
310	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

315	contrast, the experiments at 955 °C showed inhomogeneous textures (Fig. 8c). The mineral
316	assemblage in the control experiment at 955°C (EQ955°C120MPa1h) was augite, Fe-Ti
317	oxides, and apatite; however, most of their groundmass rarely contained pyroxene crystals
318	(Fig. 8c and 8d). The crystallization of Fe-Ti oxides occurred especially at the sample
319	surfaces (i.e., near the capsule walls and vesicles), and pyroxene (augite) showed a similar
320	tendency (Fig. 8c). The pyroxene crystallized locally, and their size and number density
321	showed large variations among locations. The subsequent decompression induced
322	crystallization of augite and plagioclase; however, the large part of groundmass remained
323	glassy, which indicates that the decompression-induced crystallization in these experiments
324	mainly occurred as heterogeneous nucleation on the pre-existing crystals and/or the sample
325	surfaces. On the other hand, apatite crystals were distributed throughout the sample, and
326	their size was generally a few hundred nanometers wide in all the run products.
327	Figures 9 and 10 show the BSE images of groundmass pyroxene crystals and the
328	distributions of trachts for the experiments at 955 °C. The proportion of crystals in each
329	tracht varied in association with the experimental conditions (Fig. 10; Table 6). At the
330	initial condition (EQ955°C120MPa1h), the pyroxene trachts were mainly octagonal and

hexagonal (Figs. 9a and 10a). Although the decompression to 50 MPa 331

332	(SSD955°C50MPa1h) resulted in the predominance of octagonal tracht (Figs. 9b and 10b),
333	further decompression reproduced hexagonal and parallelogrammatic trachts. The texture at
334	20 MPa (SSD955°C20MPa1h) was characterized by all the types of tracht (Figs. 9c and
335	10c), and the proportion of hexagonal tracht increased at the expense of octagonal one at 10
336	MPa (SSD955°C10MPa1h; Figs. 9d and 10d). The texture of SSD955°C5MPa1h was
337	characterized by parallelogrammatic tracht (Figs. 9e and 10e). We did not confirm obvious
338	correlation between tracht and local number densities in these runs. The CD experiment
339	(CD955°C5MPa3h) was characterized by octagonal and hexagonal trachts (Figs. 9f and
340	10f). In the CD experiment, the crystals tended to be hexagonal and parallelogrammatic in
341	the regions where their number density was relatively low, although the proportion of
342	parallelogrammatic crystals was still low. Note that the TEM analyses confirmed that each
343	type of tracht comprises the same prismatic faces as those observed in the pumice samples.
344	
345	Discussion
346	Estimating the pyroxene liquidus for the Sakurajima melt
347	Estimation of the value of $\Delta T_{\rm eff}$ for each experiment required the pyroxene liquidus in the
348	Sakurajima melt. In our experiments at 955 °C, most parts of the groundmass were glassy (Fig. 8d).

349	Because superliquidus heating hinders subsequent crystal nucleation (e.g., Sato, 1995; Pupier et al.,
350	2008; Waters et al., 2015; Arzilli et al., 2015; First et al., 2020; Matsumoto et al., 2023), this result
351	indicates that the pyroxene crystals and any crystal nuclei were dissolved in the melt at the initial
352	conditions (i.e., 120 MPa and 955 °C), and that the liquidus temperature of Cpx in this composition at
353	120 MPa is below 955 °C. In contrast, the control experiment at 935 °C contained ubiquitous
354	groundmass pyroxene (Fig. 8b). Therefore, the Cpx liquidus temperature at 120 MPa must be within the
355	range 935–955 °C in our experiments.
356	In contrast, Sekine et al. (1979) estimated pyroxene and plagioclase liquidi by
357	performing phase equilibrium experiments on the groundmass of the pumice from the 1914
358	Sakurajima eruption under water-saturated conditions at the NNO buffer. Their result at 100
359	MPa shows that the liquidus temperature of Cpx is below 930 °C, whereas our results show
360	that the liquidus temperature of Cpx is within 935–955 °C at 120 MPa. This difference can
361	be attributed to the Mg content of the melt. The MgO content of the groundmass glass in
362	the Sakurajima pumice used by Sekine et al. (1979; 0.75 wt%) is lower than that in our
363	experiments (around 1.0 wt%; Tables 2 and 3), a discrepancy also reported by Nakamura
364	(2006). Therefore, we thermodynamically modeled the effect of MgO content on the Cpx
365	liquidus using MELTS (Ghiorso and Sack, 1995; Gualda et al., 2012; Ghiorso and Gualda,

366	2015). We used rhyolite-MELTS in MELTS_Excel (Gualda and Ghiorso, 2015) and
367	obtained pyroxene liquidus curves for two melt compositions: the starting materials in our
368	experiments and those used by Sekine et al. (1979) (Fig. 11). This calculation showed that
369	increasing the MgO content of the melt by ~0.3 wt% from that used by Sekine et al. (1979)
370	raises the liquidus temperature by ~25 °C. Additionally, the experimental liquidus can be
371	reproduced by adding 55 °C to the calculated liquidus (Fig. 11).
372	
373	Magma ascent process deduced from white pumice
374	As reported by Nakamura (2006), the white pumice samples, especially Type-1,
375	were characterized by the low crystal number density (Fig. 4a). Assuming that the pyroxene
376	crystals in the Type-1 pumice crystallized under higher $\Delta T_{\text{eff}}$ , their shapes might be more
377	elongated than those in other pumice samples (e.g., Kouchi et al., 1983; Shea and Hammer,
378	2013). On the contrary, the Type-1 pumice had the lowest degree of pyroxene elongation
379	(i.e., $L/S$ ratio) of the three samples (Table 5). This might be attributed to the estimation
380	uncertainty due to the insufficient number of analyzed crystals (< 200; Mangler et al., 2022)
381	Although an underestimated value of 3D aspect ratio can affect resultant CSDs, the crystal
382	number density is still the lowest in the Type-1 pumice. When using the 3D aspect ratio of

383	1.00:1.40:10.00 instead of the estimated value ( $1.00:1.40:3.60$ ), the population densities ( $N$ )
384	decrease by approximately one natural log unit in the S-plot CSD (Fig. 4), and the CSD
385	shapes are largely maintained. Therefore, the uncertainty in 3D aspect ratio has negligible
386	impact on the following discussion.
387	The crystallization experiments demonstrated that Cpx tracht changes with the
388	extent of decompression (Fig. 10). Based on the liquidus curve (Fig. 11), the estimated $\Delta T_{eff}$
389	at 955 °C was approximately 35, 75, 95, and 110 °C at 50, 20, 10, and 5 MPa, respectively.
390	Therefore, our results indicate that the Cpx tracht changes from octagonal to hexagonal and
391	then to parallelogrammatic with increasing $\Delta T_{\rm eff}$ (Fig. 10). As shown in the tracht-specific
392	CSDs (Fig. 4a and 4b), the tracht of groundmass pyroxene crystals in the white pumice
393	samples (Type-1 and 2) changes from octagonal to parallelogrammatic with decreasing size.
394	Assuming that smaller crystals were crystallized at shallower part in the conduit, the
395	tracht-specific CSDs indicate that pyroxene crystallization in the white pumice samples
396	changed from octagon- and hexagon-dominant to hexagon- and parallelogram-dominant
397	during syneruptive ascent (Fig. 4a and 4b). Our experimental results (Fig. 10) imply that
398	the magma that produced the white pumice experienced increasing $\Delta T_{\rm eff}$ due to accelerated
399	ascent in the conduit. The concave-up shapes of the conventional CSDs (Fig. 4a and 4b)

400	consistently show the accelerated nucleation of crystals, indicating the increase of $\Delta T_{\rm eff}$
401	(e.g., Armienti et al., 1994; Marsh, 1998; Armienti, 2008). This inference is also supported
402	by the Al-increasing zonation (Fig. 5c and 6c) because the enrichment of pyroxene in Al
403	indicates rapid growth (e.g., Dymek and Gromet, 1984; Mollo et al., 2013; Masotta et al.,
404	2020). Therefore, we conclude that the magma that produced the white pumices ascended
405	sufficiently fast and reached the surface before crystallization could proceed to a significant
406	extent, attaining a high $\Delta T_{\rm eff}$ .
407	The minimum decompression rate of the white pumice magma can be constrained
408	from our CD experiment. Decompression at a constant rate of ~0.01 MPa/s mainly
409	produced octagonal and hexagonal Cpx crystals (Fig. 10f), whereas rapid decompression in
410	a single step (> 10 MPa/s) mainly produced parallelogrammatic crystals (Fig. 10e).
411	Therefore, ascent must have been faster than 0.01 MPa/s, which is plausible when
412	compared with rates estimated for Vulcanian eruptions: 0.28–0.87 $\times$ 10 <sup>-2</sup> MPa/s for the
413	1975-1987 eruptions by Miwa et al. (2009; based on the microlite number density water
414	exsolution rate meter developed by Toramaru et al., 2008); 0.7–7.8 $\times$ 10 <sup>-2</sup> MPa/s for the
415	2010 eruption by Miwa and Geshi (2012; based on cracked texture of microphenocrysts).
416	Because plagioclase and Cpx are the main phenocryst phases in the natural

417	pumices (Nakamura, 2006), the crossing of their liquidi should indicate the magma storage
418	conditions prior to the 1914 Sakurajima eruption. Our Cpx liquidus crosses that of
419	plagioclase from Sekine et al. (1979) around 50 MPa and 990 °C (Fig. 11), consistent with
420	previous estimates of the storage pressure (20-72 MPa based on the water contents of
421	phenocryst-hosted melt inclusions; Araya et al., 2019) and temperature (940-1010 °C based
422	on the pyroxene thermometry; Matsumoto and Nakamura, 2017). Given that the pumices
423	were quenched at ~5 MPa, based on the water content in the groundmass (ca. 0.4 wt%;
424	Nakamura, 2006), decompression from 50 MPa yields $\Delta T_{\rm eff} \approx 75$ °C under water-saturated
425	conditions. On the other hand, the similarity in tracht proportions (Figs. 4 and 10) indicates
426	that the maximum $\Delta T_{\rm eff}$ attained during ascent was probably between 70 and 110 °C, which
427	appears to be higher than the estimation based on melt dehydration alone ( $\Delta T_{\rm eff} \approx 75$ °C).
428	Therefore, the parallelogrammatic tracht common in the white pumices probably resulted
429	from the adiabatic cooling associated with volatile exsolution and expansion (Mastin and
430	Ghiorso, 2001; La Spina et al., 2015; Arzilli et al., 2019). Microlite crystallization may also
431	release latent heat (e.g., Couch et al., 2003b; Blundy et al., 2006; La Spina et al., 2015), but
432	this is unlikely given the low degree of crystallization.

## Implication

435	The tracht-specific CSD of the gray pumice sloped gently and was dominated by
436	octagonal pyroxene crystals spanning a broader size distribution than pyroxene crystals in
437	the white pumices (Fig. 4c), indicating relatively prolonged crystallization under lower
438	$\Delta T_{\rm eff}$ . The decrease in Al content with growth (Fig. 7c) supports the slow or declining
439	growth. In addition, the slightly rounded shapes of the groundmass crystals (Figs. 2 and 7)
440	suggest resorption by heating. We also observed a Cpx microlite with parallelogrammatic
441	zoning, implying that this crystal nucleated under high $\Delta T_{\rm eff}$ . These textures indicate that
442	the gray pumice magma initially nucleated microlites under high $\Delta T_{\rm eff}$ before prolonged
443	crystallization under decreasing $\Delta T_{\rm eff}$ , and that the magma temperature surpassed the
444	pyroxene liquidus just before quenching.
445	Assuming that the gray pumice originated from portions of the ascending magma
446	nearer to the conduit walls (Nakamura, 2006), two sources of heat are plausible: the release
447	of latent heat by crystallization (Couch et al., 2003a; Blundy et al., 2006) and shear heating
448	(Mastin, 2005; Costa et al., 2007; Hale et al., 2007). Since the crystallization increases
449	magma temperature by 2.3-3.2 °C per 1 % (Couch et al., 2003a; Blundy et al., 2006), the

450 temperature rise by the latent heat release can be up to 50  $^\circ\mathrm{C}$  by assuming 15 %

451	crystallization of pyroxene + plagioclase (Table 4; Nakamura, 2006). Moreover, shear
452	heating can increase magma temperature by several tens of degrees (e.g., Mastin, 2005;
453	Costa et al., 2007; Hale et al., 2007). The weak orientation of the groundmass crystals (Fig.
454	2) suggests shear deformation. Furthermore, shear heating is enhanced in the presence of
455	crystals (Mueller et al., 2010). Therefore, both heating processes were probably significant
456	during groundmass crystallization in the gray pumice, resulting in the rounding of the
457	groundmass crystals.
458	Finally, the textural differences between the white and gray pumices indicate that
459	they probably originated from the same magma but different radial positions within the
460	conduit (e.g., Polacci, 2005; Nakamura, 2006): the white pumice represents magma that
461	ascended rapidly in the center of the conduit, whereas the gray pumice originated from
462	magma nearer the conduit walls (Fig. 12). Magma flows more slowly in the vicinity of the
463	conduit walls than in the center of the conduit because of wall-generated friction (e.g.,
464	Gonnermann and Manga, 2003; Mastin, 2005; Costa et al., 2007; Hale and Mühlhaus,
465	2007). The low vesicularity of gray pumice is attributed to the efficient degassing near the
466	walls due to the longer duration for gas escape and the shear deformation (e.g., Stasiuk et
467	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.
Our investigation demonstrates the value of pyroxene tracht analyses for

- 471 unraveling magma ascent dynamics via the evolution of  $\Delta T_{\text{eff}}$ . Because this method is
- 472 applicable to crystal-poor glassy samples and achieves high temporal resolution when
- 473 crystals are zoned, further such tracht analyses should contribute to a better understanding
- 474 of syneruptive magma ascent dynamics.

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721	

722	Figure captions
723	
724	Figure 1. Classification of pyroxene trachts. (a) A 3D shape of pyroxene crystal with
725	octagonal tracht. (b) The shape variation of cross section depending on trachts. (c) Cross
726	sections of groundmass pyroxene crystals were classified based on the number of faces
727	between a given pair of parallel faces (indicated by circles). (d) Incomplete shapes were
728	classified similarly if a pair of parallel faces was present. (e) Individual segments of
729	attached crystals were classified in a similar manner.
730	
731	Figure 2. BSE images of groundmass of the Sakurajima pumice samples. Groundmass
732	crystal phases are Fe-Ti oxides, pyroxene, and plagioclase (in order of decreasing
733	brightness). The brightness of the groundmass glass is similar to that of plagioclase. Black
734	regions are vesicles. The images in the right column show the groundmass pyroxene
735	crystals with trachts characteristic of each sample. Abbreviations: $Pl = plagioclase; Px =$
736	pyroxene; Ox = Fe-Ti oxides.
737	

738 Figure 3. Estimation of 3D aspect ratio of groundmass pyroxene crystals by ShapeCalc

739	(Mangler et al., 2022). The best-fitted 300 estimates are plotted in Zingg diagram (Zingg,
740	1935) with the colors based on the cumulative goodness of fit $(R_c^2)$ . The uncertainties $(1\sigma)$
741	on the best shape estimate are provided as black bars. Note that the plotted distributions do
742	not necessarily represent the actual variation of crystal shapes: we did not find tabular cross
743	sections of pyroxene through the SEM observation, but most pyroxene crystals seemed to
744	have acicular shapes in 3D as far as we observed. Abbreviations: $S =$ short-axis length, $I =$
745	intermediate-axis length, $L = long$ -axis length.
746	
747	Figure 4. Tracht-specific CSDs of groundmass pyroxene crystals in the Sakurajima pumice
748	samples. The conventional CSDs (i.e., including all trachts) are plotted in black for
749	comparison. The size range shown is from 0.16 to 6.31 $\mu$ m in width (short-axis length). The
750	total number of analyzed crystals and the proportions of crystals in each tracht are shown at
751	top-right in each panel.
752	
753	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# (=Mg/(Mg + Fe) in mol) compositional

maps, respectively, and (e) a SAED pattern were obtained along the [001] zone axis. (f and

756 g) SAED patterns were obtained from the points 1 and 2 in (a), at another orientation. (g) 757 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 = even"). Abbreviations: Opx, orthopyroxene; 758 759 Cpx, clinopyroxene. 760 Figure 6. Internal texture of a parallelogrammatic clinopyroxene nanolite in the Type-2 761 pumice. 762 763 (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 764 obtained at another orientation from (f) the center and (g) the left side of the crystal. There 765 are weak reflections of  $P2_1/c$  in (g). (h) A dark-field image with reflection  $g = 0\overline{3}\overline{1}$  shows 766 anti-phase boundaries in the pigeonite (Pgt) domain. 767 768



- (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

773 outline.

774

775	Figure 8. Backscattered electron (BSE) images of control experiments at 120 MPa. The run
776	products at 935 °C (EQ935°C120MPa1h; <b>a</b> and <b>b</b> ) and 955 °C (EQ955°C120MPa1h; <b>c</b> and
777	d). The control experiment at 955 °C resulted in inhomogeneous texture: Fe-Ti oxides and
778	pyroxene crystallized locally near the sample surface (upper-left part in c) whereas most of
779	the groundmass was glassy (d). Abbreviations: Px, pyroxene; Ox, Fe-Ti oxides; Pl,
780	plagioclase.
781	
782	Figure 9. BSE images of groundmass pyroxene crystals in run products at 955 °C. The
783	control experiment at 120 MPa (a; EQ955°C120MPa1h). The single-step decompression
783 784	control experiment at 120 MPa ( <b>a</b> ; EQ955°C120MPa1h). The single-step decompression experiments at the final pressure of 50 ( <b>b</b> ; SSD955°C50MPa3h), 20 ( <b>c</b> ;
783 784 785	control experiment at 120 MPa ( <b>a</b> ; EQ955°C120MPa1h). The single-step decompression experiments at the final pressure of 50 ( <b>b</b> ; SSD955°C50MPa3h), 20 ( <b>c</b> ; SSD955°C20MPa3h), 10 ( <b>d</b> ; SSD955°C10MPa3h), and 5 MPa ( <b>e</b> ; SSD955°C5MPa3h).
<ul><li>783</li><li>784</li><li>785</li><li>786</li></ul>	control experiment at 120 MPa ( <b>a</b> ; EQ955°C120MPa1h). The single-step decompression experiments at the final pressure of 50 ( <b>b</b> ; SSD955°C50MPa3h), 20 ( <b>c</b> ; SSD955°C20MPa3h), 10 ( <b>d</b> ; SSD955°C10MPa3h), and 5 MPa ( <b>e</b> ; SSD955°C5MPa3h). The continuous decompression experiment from 120 to 5 MPa ( <b>f</b> ; CD955°C5MPa3h).
783 784 785 786 787	control experiment at 120 MPa ( <b>a</b> ; EQ955°C120MPa1h). The single-step decompression experiments at the final pressure of 50 ( <b>b</b> ; SSD955°C50MPa3h), 20 ( <b>c</b> ; SSD955°C20MPa3h), 10 ( <b>d</b> ; SSD955°C10MPa3h), and 5 MPa ( <b>e</b> ; SSD955°C5MPa3h). The continuous decompression experiment from 120 to 5 MPa ( <b>f</b> ; CD955°C5MPa3h). Abbreviations: Px, pyroxene; Ap, apatite.

**Figure 10.** Tracht-specific CSDs of pyroxene crystals produced in experiments at 955 °C.

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

797 Figure 11. Estimated magma storage conditions and liquidus curves for the 1914 Sakurajima magma. Cpx (black), Opx (gray), and plagioclase liquidi (blue) reported by 798 799 Sekine et al. (1979) are shown alongside pyroxene liquidi for the melt compositions of 800 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 801 802 results. A black horizontal bar represents the range of possible Cpx liquidus temperatures 803 constrained by the control experiments in this study (i.e., 935–955 °C). The shaded area 804 indicates the magma storage conditions estimated based on pyroxene thermometry 805 (Matsumoto and Nakamura, 2017) and phenocryst-hosted melt inclusion H<sub>2</sub>O contents (Araya et al., 2019). 806

807

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

## Tables

Name	Т (°С)	Path	P <sub>i</sub> (MPa)	P <sub>f</sub> (MPa)	duration (h)	GM phase <sup>a</sup>	$\Delta T_{\rm eff}$ for Px (K)	Main Px tracht
EQ935C12 0MPa1h	935	EQ	120	120	1	Px, Pl, Ox, Ap	10	8,6
EQ955C12 0MPa1h	955	EQ	120	120	1	Px, Ox, Ap	~0	8,6
SSD955C5 0MPa3h	955	SSD	120	50	3	Px, Pl, Ox, Ap	35	8
SSD955C2 0MPa3h	955	SSD	120	20	3	Px, Pl, Ox, Ap	75	8, 6, 4
SSD955C1 0MPa3h	955	SSD	120	10	3	Px, Pl, Ox, Ap	95	6
SSD955C5 MPa3h	955	SSD	120	5	3	Px, Pl, Ox, Ap	110	4
CD955C5 MPa3h	955	CD	120	5	3	Px, Pl, Ox, Ap	<110	8,6

Table 1. The experimental conditions and resultant textures.

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

823

	Type-1		Type-2		Туре-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)
$Al_2O_3$	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)
$P_2O_5$	0.17	(0.11)	0.13	(0.12)	0.21	(0.11)
Total	100		100		100	

Table 2. Average glass compositions of Sakurajima pumices.

NOTES:

Values in parentheses are standard deviations.

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

Table 3	Average glass	compositions of	f run products.						
	Starting material	EQ955°C 120MPa1h	EQ945°C 120MPa1h	EQ935°C 120MPa1h	SSD955°C 50MPa3h	SSD955°C 20MPa3h	SSD955°C 10MPa3h	SSD955°C 5MPa3h	CD955°C 5MPa3h
$(Wt\%)^a$	n = 45	n = 39	n = 30	n = 29	n = 33	n = 37	n = 40	n = 45	n = 36
$SiO_2$	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)	70.86 (0.39)
$TiO_2$	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)	0.61 (0.08)
$Al_2O_3$	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)	14.42 (0.12)
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)	2.76 (0.15)
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)	0.10 (0.12)
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)	0.87 (0.15)
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)	3.36 (0.19)
$Na_2O$	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)	4.29 (0.08)
$K_2O$	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)	2.59 (0.06)
$P_2O_5$	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)	0.13 (0.13)
Total	100	100	100	100	100	100	100	100	100
NOTES	: Values in pare	ntheses are stan	dard deviations.						

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

Sample	Number of	Analy	Px content	Px content Number of crystal cross sections							
	regions	regions (excluding vesicles)									
		(µm <sup>2</sup> )	(vesicle%) <sup>a</sup>	(area%)	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	

## Table 4. Texture of the Sakurajima pumice samples.

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

## 829

## 830 Table 5. Parameters used in CSDCorrections.

Sample	3D aspect r	atio	Roundness	Size scale length
	S·I·L	Rc2		(Bins per
	5.1.1	Re2		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.

Sample		$\Delta T_{\mathrm{eff}}$	Analyzed		Nu	umbers of py	roxene crysta	als	
		for Px	area						
Path	$\mathbf{P}_{\mathrm{f}}$	(K)	$(\mu m^2)$	Octagon	Heptagon	Hexagon	Pentagon	Parallelo	Other
	(MPa)							gram	
EQ	120	~0	6635	115	23	50	5	2	49
SSD	50	35	13614	114	6	3	1	0	36
SSD	20	75	15945	37	18	30	16	33	34
SSD	10	95	23844	13	13	40	22	11	23
SSD	5	110	15905	2	2	13	38	173	31
CD	5	<110	15144	64	9	54	10	4	51

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



Figure 2



#### Figure 3 (a) Type-1

... 6.7

.... 5 ...

1.1

#### (b) Type-2











#### Figure 7



### Figure 8



Figure 9









